STC2026

62nd Symposium on Theoretical Chemistry

Book of Abstracts

STC 2026 Schedule

STC2026

	Monday 14.9.	Tuesday 15.9.	Wednesday 16.9.	Thursday 17.9.	Friday 18.9.
09:00		09:00–09:40 **Saalfrank**	09:00–09:40 **González**	09:00–09:40 **Zgid**	09:00–09:40 **Paulus**
		09:40–10:00 **Brehm**	09:40–10:00 **Picconi**	09:40–10:00 **Hamprecht**	09:40–10:00 **Sierka**
10:00	10:00–13:40 **Registration**	10:00–10:20 **Ahmadkhani**	10:00–10:20 **Runeson**	10:00–10:20 **Steffen**	10:00–10:20 **Hehn**
		10:20–10:40 **Dreßler**	10:20–10:40 **Titov**	10:20–10:40 **Erhard**	10:20–10:40 **Marian**
11:00		10:40–11:10 **Coffee Break**	10:40–11:10 **Coffee Break**	10:40–11:10 **Coffee Break**	10:40–11:10 **Coffee Break**
		11:10–11:50 **Tkatchenko**	11:10–11:50 **Heine**	11:10–11:50 **Ceriotti**	11:10–11:50 **Meyer**
12:00		11:50–12:10 **Helmich-Paris**	11:50–12:10 **Szabla**	11:50–12:10 **Pausch**	11:50–12:10 **Gorfer**
		12:10–12:30 **Meisner**	12:10–12:30 **Röhr**	12:10–12:30 **Mazumder**	12:10–12:30 **Lambie**
13:00		12:30–14:00 **Lunch**	12:30–14:00 **Lunch**	12:30–14:00 **Lunch**	12:30–12:45 **Poster Prizes & Closing**
14:00	13:45–14:00 **Opening**				
	14:00–14:40 **Clary**	14:00–14:40 **Grüneis**	14:00–15:20 **Hückel Award: Domcke and Manz**	14:00–19:30 **Excursions**	14:10–22:00 **After STC Event**
15:00	14:40–15:00 **R. Fink**	14:40–15:00 **K. Fink**			
	15:00–15:20 **Jansen**	15:00–15:20 **Penschke**			
	15:20–15:50 **Coffee Break**	15:20–15:50 **Coffee Break**	15:20–15:50 **Coffee Break**		
16:00	15:50–16:30 **Keller**	15:50–16:10 **Dittmer**	15:50–16:30 **Hellmann Award**		
		16:10–16:30 **Kuc**			
	16:30–16:50 **Mai**	16:30–17:30 **Peyerimhoff Award: Hasecke and Liebert**	16:30–17:30 **AGTC Meeting**		
17:00	16:50–17:10 **von Domaros**				
	17:10–17:30 **Frömbgen**				
	17:30–19:30 **Break**	17:30–19:30 **Break**	17:30–19:30 **Break**		
18:00					
19:00					
	19:30–22:00 **Poster Session 1**	19:30–22:00 **Poster Session 2**	19:30–22:00 **Poster Session 3**	19:30–22:00 **Conference Dinner**	
20:00					
21:00					
22:00					

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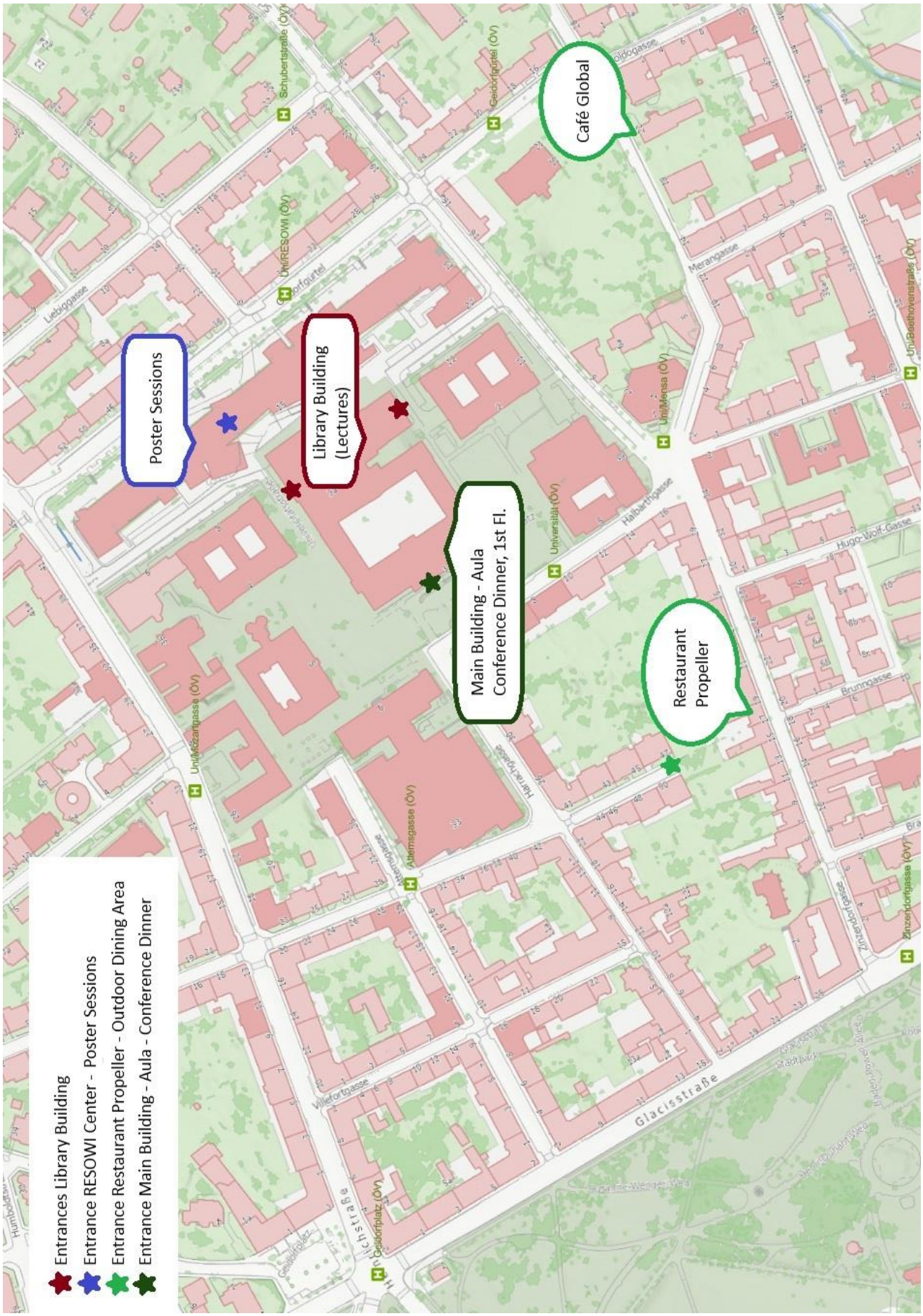
Conference Locations



Please turn over

Conference Locations:

Library Building

Registration and Talks

Universitätsplatz 3a, 8010 Graz

RESOWI Zentrum

Poster Sessions

Universitätsstrasse 15, 8010 Graz

Main Building

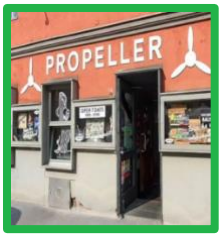
Conference Dinner (1st Floor)

Universitätsplatz 3, 8010 Graz

Lunch:

Café Global

Leechgasse 22, 8010 Graz



Restaurant Propeller

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Meeting Points for Excursions (Thursday)

STC2026

Guided tour of Schlossberg with walk through the city centre:

Start: 14:30
Meeting place: Karmeliterplatz, “Dreifaltigkeitssäule”
Address: Am Fuße des Schlossberges 2, 8010 Graz

Old town tour with a visit to the Zeughaus (armoury):

Start: 14:30
Meeting place: Hauptplatz (Main Square), in front of City Hall
Address: Hauptplatz 1, 8010 Graz

Guided tour of Schloss Eggenberg including the palace gardens:

Start Group 1: 14:30
Start Group 2: 14:45
Meeting place: Schloss Eggenberg Entrance Gate
Address: Eggenberger Allee 95, 8020 Graz

Climbing Garden Hilmteich:

Start: 14:30
Meeting place: Entrance Climbing Garden
Address: Hilmteichstraße 110a, 8010 Graz

Hiking:

Start: 14:30
Meeting place: Entrance Library Building

Conference Program

Monday, September 14

10:00–13:40 Registration

13:45–14:00 **Opening Ceremony**

Session 1 – Chair: Alexander Sax

14:00–14:40 **I1** **David Clary** — University of Oxford, United Kingdom
Schrödinger in Graz

14:40–15:00 **C1** **Reinhold F. Fink** — University of Tübingen, Germany
Wave functions are mapped in intermolecular potential energy surfaces of π-stacked aggregates

15:00–15:20 **C2** **Georg Jansen** — University Duisburg-Essen, Germany
Reformulation of intermolecular perturbation theory using imaginary time

15:20–15:50 Coffee Break

Session 2 – Chair: Tillmann Klamroth

15:50–16:30 **I2** **Bettina Keller** — Freie Universität Berlin, Germany
Learning Reaction Mechanisms from Molecular Dynamics: Isomerization in Retinal and p–Coumaric Acid

16:30–16:50 **C3** **Sebastian Mai** — University of Vienna, Austria
Excited-state dynamics using the SHARC4 package

16:50–17:10 **C4** **Michael von Domaros** — Philipps-Universität Marburg, Germany
From Biased Trajectories to Local Mobility: Residence Times as a Route to Diffusivity

17:10–17:30 **C5** **Tom Frömbgen** — University of Bonn, Germany & EPFL, Switzerland
A Multifidelity Monte Carlo Approach for Simulating the Diffusion Coefficient of Water

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17:30–19:30 Break

19:30–22:00 **Poster Session 1** (Posters P1–P66)

Tuesday, September 15

Session 3 – Chair: Shirin Faraji

09:00–09:40 **I3** **Peter Saalfrank** — University of Potsdam, Germany
Adsorbates at periodic metal and insulating surfaces: From structure to non-equilibrium dynamics

09:40–10:00 **C6** **Martin Brehm** — Paderborn University, Germany
Beyond Verlet: Unlocking a 2x Speedup in Molecular Dynamics with Novel Time Integrators

10:00–10:20 **C7** **Somayeh Ahmadkhani** — Freie Universität Berlin, Germany
Dipole-Renormalized Reservoirs for Adaptive Open-QM/MM Simulations of Liquid Water

10:20–10:40 **C8** **Christian Dreßler** — TU Ilmenau, Germany
Description Is Not Understanding: From Molecular Trajectories to Atomistic Mechanisms

10:40–11:10 Coffee Break

Session 4 – Chair: Oliver Kühn

11:10–11:50 **I4** **Alexandre Tkatchenko** — University of Luxembourg, Luxembourg
How Atoms Interact Within Molecules

11:50–12:10 **C9** **Benjamin Helmich-Paris** — FACCTs GmbH, Germany
Excited-state methods based on state-averaged long-range CASSCF short-range DFT

12:10–12:30 **C10** **Jan Meisner** — Heinrich Heine University Düsseldorf, Germany
Reaction Discovery in Periodic Systems

12:30–14:00 Lunch

Session 5 – Chair: Doreen Mollenhauer

14:00–14:40 **I5** **Andreas Grüneis** — TU Wien, Austria
Status and Future Challenges for periodic Coupled-Cluster Simulations in Materials

14:40–15:00 **C11** **Karin Fink** — Karlsruher Institut für Technologie, Germany
Quantum chemical calculations on surface adsorption

15:00–15:20 **C12** **Christopher Penschke** — University of Potsdam, Germany
Water splitting at two-dimensional carbon-based materials

15:20–15:50 Coffee Break

Session 6 – Chair: Maren Podewitz

15:50–16:10 **C13** **Anneke Dittmer** — FACCTs GmbH, Germany
Revisiting the optical band gaps of Co_3O_4 using molecular methods

16:10–16:30 **C14** **Agnieszka Kuc** — CASUS/HZDR, Germany
Engineering Hybrid Two-Dimensional TMDC/Perovskite Heterostructures from First Principles

Sigrid Peyerimhoff Award

16:30–17:00 **A1** **Lukas Hasecke** — University of Göttingen, Germany
Development of Low-Order Scaling Multicomponent Methods

17:00–17:30 **A2** **Julia Liebert** — Princeton University, USA
Quantum Information Theoretical Approach to Functional Theories

17:30–19:30 Break

19:30–22:00 **Poster Session 2** (Posters P67–P132)

Wednesday, September 16

Session 7 – Chair: Carmen Herrmann

09:00–09:40 **I6** **Leticia González** — University of Vienna, Austria
From Molecules to Materials: Effective Hamiltonians for Spin Dynamics and Magnetic Exchange

09:40–10:00 **C15** **David Picconi** — Heinrich Heine University Düsseldorf, Germany
Photodynamics of multi-mode multi-state molecular systems coupled to a dissipative environment

10:00–10:20 **C16** **Johan Runeson** — University of Freiburg, Germany
A simple way to deal with decoherence in trajectory surface hopping

10:20–10:40 **C17** **Evgenii Titov** — University of Potsdam, Germany
Modelling nonadiabatic dynamics of excitonic and plasmonic photoswitchable systems

10:40–11:10 Coffee Break

Session 8 – Chair: Ralf Tonner-Zech

11:10–11:50 **I7** **Thomas Heine** — TU Dresden, Germany
Long-range magnetic order in metal-free two-dimensional materials

11:50–12:10 **C18** **Rafał Szabla** — Wroclaw University of Science and Technology, Poland
Quantifying photoinduced electron transfer in RNA and DNA

12:10–12:30 **C19** **Merle Röhr** — University of Würzburg, Germany
Topological Photophysics in Molecular Aggregates: The Role of Charge-Transfer States

12:30–14:00 Lunch

14:00–15:20 **Erich Hückel Award: Wolfgang Domcke and Jörn Manz**

15:20–15:50 Coffee Break

15:50–16:30 **Hans G.A. Hellmann Award**

16:30–17:30 **AGTC Meeting**

17:30–19:30 Break

19:30–22:00 **Poster Session 3** (Posters P133–P197)

Thursday, September 17

Session 9 – Chair: Tim Neudecker

09:00–09:40 **I8** **Dominika Zgid** — University of Michigan, USA
Relativistic GW for solids and molecules

09:40–10:00 **C20** **Fred Hamprecht** — Heidelberg University, Germany
Orbital-free DFT is becoming practical

10:00–10:20 **C21** **Julien Steffen** — Friedrich-Alexander-University Erlangen-Nürnberg, Germany
Self-Consistent Fine-Tuning of Machine Learning Interatomic Potentials by Targeted Exploration of Configuration Space

10:20–10:40 **C22** **Jannis Erhard** — McMaster University, Hamilton, Canada
Implementing Reduced Density Matrix Functional Theory within a Kohn–Sham like Framework

10:40–11:10 Coffee Break

Session 10 – Chair: Andreas Dreuw

11:10–11:50 **I9** **Michele Ceriotti** — EPFL, Switzerland
Let them learn: AI models that master materials physics

11:50–12:10 **C23** **Amber Pausch** — Universität Münster, Germany
Beyond Potential Energy Surfaces: A Quantum Geometric View of Molecular Electronic Structure

12:10–12:30 **C24** **Shivnath Mazumder** — Indian Institute of Technology Jammu, India
Theoretically Designed Electro- and Photoactivated Earth-Abundant Metal-Based Catalysts for Dihydrogen Generation from Water

12:30–14:00 Lunch

14:00–19:30 **Excursions**

19:30–22:00 **Conference Dinner**

Friday, September 18

Session 11 – Chair: Thorsten Klüner

09:00–09:40 **I10** **Beate Paulus** — Freie Universität Berlin, Germany
Molecule-2D heterostructures - still a challenge for computational chemistry

09:40–10:00 **C25** **Marek Sierka** — Friedrich Schiller University Jena, Germany
Watching Excitons Move: Real-Time and Linear-Response TDDFT for Periodic Systems in TURBOMOLE

10:00–10:20 **C26** **Anna Hehn** — Christian-Albrechts-University Kiel, Germany
Reshaping the research landscape for the description of excited states in solids: the tool set of the CP2K-Newton-X program package

10:20–10:40 **C27** **Christel Marian** — HHU Düsseldorf, Germany
Spin–orbit Coupling of TDDFT Amplitudes: Left or Right?

10:40–11:10 Coffee Break

Session 12 – Chair: Robert Berger

11:10–11:50 **I11** **Bernd Meyer** — Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany
Accelerated Materials Discovery with Machine-Learning and DFT

11:50–12:10 **C28** **Alexander Gorfer** — Fritz Haber Institute of the MPS, Berlin, Germany
Super-Resolution for Real-Time Electronic Structure Theory in Complex Systems

12:10–12:30 **C29** **Stephanie Lambie** — Max Planck Institute for Solid State Research, Germany
The Subtleties of Calculating Non-Covalent Interactions with Wavefunction Based Methods

12:30–12:45 **Poster Prizes and Closing**

14:10–22:00 (After STC Event)

Poster Session 1 (Monday, P1–P66)

P1 **Tolibjon Abdurakhmonov** — University of Rostock, Germany
Davydov Splitting Without a Davydov Pair in Perylene Red Microcrystals with a Large Unit Cell

P2 **Zarah Aigner** — University of Innsbruck, Austria
Generative Neural Networks to sample PIMC configurations

P3 **Ardhmeri Alija** — Helmholtz Institute Mainz, Johannes Gutenberg-Universität Mainz, Germany
Prospective Parity-Violation NMR Experiments: A Relativistic DFT Study of Chiral Platinum Complexes

P4 **Vahideh Alizadeh** — Center for Advanced Systems Understanding (CASUS), Germany
Quantifying and Eliminating Self-Interaction Error in Density Functionals: A Recipe for Traditional and Machine-Learning DFT

P5 **Vipul Kumar Ambasta** — Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany
Adsorption-Induced Structural and Optical Changes of a thioindigo-type photoswitch on Au(111)

P6 **Mrinal Arandhara** — Ruhr University Bochum, Germany
Molecules in Quantum Solids: Protonated Methane in para-Hydrogen Matrices

P7 **Maximilian Asbach** — Universität München (LMU), Germany
Overcoming Variational Collapse in Excited-State Simulations: A Benchmark of a Optimally Tuned Constraint-Based Orbital-Optimized Excited-State Approach

P8 **Alice Balbi** — University of Warsaw, Poland
Neutral electronic excitations from an iterative solution of the Bethe-Salpeter equation

P9 **Philipp Baltruschat** — University of Hamburg, Germany
Optimizing Input vs Correcting Output in Semiempirical Machine Learning

P10 **Tom Barnowsky** — Technische Universität Dresden & Helmholtz-Zentrum Dresden-Rossendorf, Germany
Beyond van der Waals: High-Throughput Prediction of 2D Materials from Non-Layered Crystals

P11 **Lily Barta** — University of Augsburg, Germany
Exploiting Hard-Core Boson Structure in Quantum Chemistry

P12 **Marco Bauer** — KTH Royal Institute of Technology, Sweden
Cutting Active Spaces Beyond the Strong-Orthogonality Condition via Excitonic Renormalization

P13 **Saman Behjou** — Nicolaus Copernicus University in Toruń, Poland
Analytic Gradients and Geometry Optimization for Orbital-Optimized Pair Coupled Cluster Doubles

P14 **Jonas Bentrup** — University of Bremen, Germany
Simulating Polymer Scission in Ultrasound Baths using Molecular Dynamics

P15 **Florian Bischoff** — University of Bremen, Germany
Multiscale Modelling of notched PMMA Polymers

P16 **Luca Bonfirraro** — Max Planck Institute for Solid State Research, Germany
Stochastic-DDCI: Accurate Magnetic Modeling in Large Active Spaces

P17 **Anne Brandmeier** — Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany
Training and application of machine-learned force fields for catalytically active nanoparticles

P18 **Marvin Breier** — University of Bremen, Germany
Generalizing the Judgement of Energy DIstribution (JEDI) Strain Analysis for Periodic Systems under Lattice Deformation

P19 **Michael Bühler** — University of Würzburg, Germany
Planar to Dewar isomerisation from azaborine to azaboranaphthalene under mechanistic, electronic, and substituent control

P20 **Joanna Bukojemska** — University of Warsaw, Poland
Solving Bethe Salpeter equation using tensor decomposition

P21 **Martin Dagleish** — Ludwig-Maximilians-Universität München, Germany
Investigating Higher-Order Excited-State Properties with Constraint-Based Orbital-Optimized Excited States (COOX)

P22 **Shubhajit Das** — Technische Universität Dresden, Germany
Capturing Activity Trends in Metal-Organic Framework Catalysis using Minimal Active Site Models

P23 **Shreya Das** — Heidelberg University, Germany
Unravelling Molecular Motor Coupling in Porous Molecular Scaffolds at Atomic Scale

P24 **Deepika Deepika** — Indian Institute of Technology Kanpur, India
Effects of Conterion on the Solvation Structure and Migration Kinetics of Hydroxide Ions in Concentrated Aqueous NaOH Solutions

P25 **Michael Deffner** — University of Hamburg, Germany
The Design of Problem-Specific Molecular Descriptors

P26 **Daniel Deißenbeck** — Heinrich Heine University, Düsseldorf, Germany
Reaction Discovery on Copper Surface Using Periodic Nanoreactor Molecular Dynamics with Machine-Learned Force Fields

P27 **Adrian Dempwolff** — University of Heidelberg, Germany
Recent Developments in the Algebraic Diagrammatic Construction Framework

P28 **Javier Camargo Díaz** — Freie Universität Berlin, Germany
Computational Studies of PFAS Detection, Adsorption, and Degradation

P29 **Samuel Dier** — University of Münster, Germany
Frozen-Density Embedding for Mixed-Reference Spin-Flip TDDFT

P30 **Linus Bjarne Dittmer** — University of Heidelberg, Germany
Improving the description of electron attachment and detachment in ADC at almost no cost via repartitioning

P31 **Werner Dobrautz** — Helmholtz Zentrum Dresden-Rossendorf, Germany
Improving Sample-Based Quantum Diagonalization for Strongly Correlated Electronic Structure Problems

P32 **Jacek Dobrzyniecki** — University of Warsaw, Poland
Ground state of the Hubbard model with spin-dependent linear potential

P33 **Jindra Dušek** — ETH Zürich, Switzerland
Incorporating rotational symmetry and quantisation into instanton theory

P34 **Diyan Unmu Dzujah** — TU Dresden, Germany
Mechanism of Thermal Defunctionalization of Phenyl and Methyl Groups on Graphene

P35 **Sebastian Ehlert** — Microsoft Research, AI for Science
Accurate and scalable exchange-correlation with deep learning

P36 **Jonathan Eifler** — University of Potsdam, Germany
A MACE-Based Machine Learning Potential for GNR Formation on Au (111)

P37 **Chen Chen Er** — Technische Universität Dresden & Helmholtz-Zentrum Dresden-Rossendorf, Germany
Modelling Confined High-entropy Materials

P38 **Matías Emanuel di Mauro Esteban** — MPI-FKF, Germany
Transcorrelating Solids: Solving the twin problems of slow basis-set and thermodynamic limit convergence of Quantum Chemistry methods at the same time.

P39 **Steffen Fauser** — CEA DAM-DIF, France
GW Approximation within the Projector Augmented-Wave Method

P40 **Noah Felis** — University of Bremen, Germany
Machine-Learning-Accelerated Structure Determination Combining Surface XRD and DFT

P41 **Anthuan Ferino-Pérez** — Saarland University, Germany
Complex-variable equation-of-motion coupled-cluster variants with triples corrections for the description of electronic decay processes

P42 **André Flacke** — Philipps-Universität Marburg, Germany
Finite Nuclear Size Effects in Parity-Violating Properties of Chiral Molecules

P43 **Tea Frey** — University of Zagreb, Croatia
Elastic Properties of Organic Crystals: A Periodic DFT Study

P44 **Rico Friedrich** — Technische Universität Dresden, Germany
Automated Corrections for Materials Design of Ionic Systems: AFLOW-CCE

P45 **Hans Georg Gallmetzer** — University of Vienna, Austria
Excited-state Dynamics of a Photoswitchable Vitamin D Receptor Agonist

P46 **Gaurab Ganguly** — University of Vienna, Austria
Mechanism-Guided Design of Metal-Based Molecular Solar-Thermal Batteries

P47 **Michael Gatt** — Universität Innsbruck, Austria
What Does It Take to Automate Multireference Electronic Spectroscopy for Large-Scale Astrochemical Explorations?

P48 **Sven Geller** — University of Cologne, Germany
Modelling the Impact of Structural and Electronic Disorder Effects on the Charge Transport Properties of Merocyanines

P49 **Leon Gerndt** — Humboldt-Universität zu Berlin, Germany
Multireference Studies of Strongly Correlated Transition Metal Complexes: When DFT Fails at the Geometry

P50 **Carl Leon Giard** — University of Münster, Germany
Piezochromism of tridentate platinum complexes for adaptive materials

P51 **Szabolcs Goger** — Universität des Saarlandes, Germany
Absorption and Response Properties of Molecules Coupled to Optical Quantum Cavities with Coupled Cluster Theory

P52 **Namrata Gohain** — Philipps University Marburg, Germany
On the Role of Substituent Arrangements versus Carbon Backbone towards Chirality Observables

P53 **Richard Gundermann** — University of Potsdam, Germany
Understanding Resonance-Enabled Polaritonic Control of Hydrogen Transfer Dynamics and Relaxation

P54 **Kota Hanasaki** — University of Zurich, Switzerland
Ab initio calculation of energy-dependent Hubbard parameters for DFT+U using real-time TDDFT

P55 **Jonas Hänseroth** — Technische Universität Ilmenau, Germany
Exploiting the Softening of Universal MLIPs for Data-Efficient Material-Specific Models

P56 **Redouan El Haouari** — Ruhr-Universität Bochum, Germany
Machine-Learning Potentials for redox chemistry in solution

P57 **Eric Heller** — University of Freiburg, Germany
Instanton theory for reaction rates far from equilibrium

P58 **Jenna Hendrix** — Carl von Ossietzky University of Oldenburg, Germany
Bridging the Gap: A Benchmark Study of Hyperfine Coupling Constants from EPR Spectroscopy to Qubit Decoherence Times

P59 **Jesper Holtmann** — Bielefeld University, Germany
Towards the construction of a global potential energy surface for H4+ through isotropic direction sampling

P60 **Hannes Hoppe** — ETH Zurich, Switzerland
Vibrationally Nonadiabatic Dynamics of (Floppy) Molecules in Liquid Environments

P61 **Mateja Hrast** — Jožef Stefan Institute, Slovenia & Technical University of Munich, Germany
Ab-initio Auger spectrum of the ultrafast dissociating $2p_{3/2}^{-1}\sigma^*$ resonance in HCl

P62 **Marc Ickler** — Heidelberg University, Germany
All-Electron Machine-Learned Orbital-Free Density Functional Theory for Periodic Systems

P63 **Richard Jacobi** — University of Vienna, Austria
Computational protocol for the prediction of energy transfer rates in DNA-liposome hybrids

P64 **Grzegorz Jaczewski** — University of Warsaw, Poland
Ab-initio relativistic embedding methods for strongly correlated systems

P65 **Natalia Goncharova** — University of Graz, Austria
Multimer Embedding for Predictions of Polymorph Stability

P66 **Robin Guttmann** — University of Graz, Austria
HB49rev and HB35revx10: New Highly Accurate Benchmark Sets for Hydrogen-Bonded Systems

Poster Session 2 (Tuesday, P67–P132)

P67 **Nicolas Jahn** — University of Potsdam, Germany
Nonadiabatic Molecular Dynamics Enables Direct Modeling of Plasmon-Driven Processes at the Single Molecule Level

P68 **Azin Mazloom Jalali** — Friedrich Schiller University Jena, Germany
Temperature-Dependent Collapse of Surface-Tethered PNIPAM at a Bioactive-Glass-Relevant Silica–Water Interface: A Dissipative Particle Dynamics Study

P69 **Kjell Janke** — Philipps-Universität Marburg, Germany
Quantum Electrodynamic Corrections in Molecules

P70 **Andy Jiang** — University of Georgia, USA
Higher Order Pair Natural Orbital-Based Coupled-Cluster Methods

P71 **Tomáš Jíra** — University of Chemistry and Technology, Prague, Czech Republic
Spin-Orbit Induced Dynamics in Atom-Ion Collisions

P72 **Jannis Leif Johann** — University of Vienna, Austria
Ultrafast Intersystem Crossing and Transient 2c–3e S–S Bond Formation in [(bpy)Pt(SPh)$_2$]

P73 **Michelle Kalinicenko** — Bielefeld University, Germany
Quantum chemical study of possible reaction pathways between IO and CH3OO

P74 **Daniel Kats** — Max Planck Institute for Solid State Research, Stuttgart, Germany
ElemCo.jl: A Julia Package for Electron Correlation Methods

P75 **Siyara Kehelwalathenne** — University of Leipzig, Germany
Probing Isotope Effects on Metal–H_2 Chemical Bonding in Cu-, Zn-, and Bimetallic Cu/Zn-HKUST-1 Metal–Organic Frameworks

P76 **Jamoliddin Khanifaev** — University of Bonn, Germany
Investigation of HCl-H2O electrolyte using AIMD simulations

P77 **Mira Kim** — Heinrich Heine University Düsseldorf, Germany
Analytical gradients and non-adiabatic couplings within the algebraic diagrammatic construction scheme

P78 **Nico Kißing** — University of Bremen, Germany
Emergence in Computational High-Pressure Chemistry: Equations of State of Molecular Clusters

P79 **Maximilian Peter-Paul Kovar** — TU Wien, Austria
Chemical Reaction Machine Learning: Data-Centric Shortcuts for Reaction Energetics and Transition State Geometries

P80 **Jennifer Kremper** — Heinrich-Heine University Düsseldorf, Germany
Photophysical Properties of a MR-TADF Emitter in Complex Environments

P81 **Alexander Kreuzkamp** — Heinrich Heine University, Düsseldorf, Germany
Computational investigation on red-light driven molecular motors enabled by triplet-triplet annihilation upconversion

P82 **Simon León Krug** — ETH Zurich, Switzerland
Internal conversions with non-adiabatic instanton theory

P83 **Timo Lebeda** — University of Cambridge, UK
The ultrananonlocal LAK meta-GGA: An efficient density functional for molecules, materials, and interfaces

P84 **Ka Hei Lee** — Helmholtz-Zentrum Berlin, Germany
Light-induced electron dynamics in open-shell systems via the restricted open-shell method

P85 **Katharina Leitmann** — Ruhr-Universität Bochum, Germany
Freezing the Unfreezable: para-Hydrogen Microsolvation Tunes CH5+ Fluxionality

P86 **Regina Lennarz** — Heinrich Heine University Düsseldorf, Germany
Simulating mechanochemical disassembly pathways of PdnL2n supramolecular architectures

P87 **Qianlin Li** — Freie Universität Berlin, Germany
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P99 **Elham Mazarei** — University of Potsdam, Germany
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P102 **Xincheng Miao** — University of Würzburg, Germany
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Diffusion Mechanisms of Oxygen in $CaMnO_3$ Perovskites and Related $CaMnO_{2.5}$ Ordered Oxygen Vacancy Structures

P104 **Mihnea Mlak-Marginean** — Marburg University, Germany
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P105 **Andreas Mölkner** — Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany
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P107 **Sara Karimi Nejad** — Leibniz University Hannover (LUH), Germany
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P109 **Grzegorz Niedzielski** — Jagiellonian University, Krakow, Poland
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P110 **Anastasiia Nihei** — Technische Universität Dresden & Helmholtz-Zentrum Dresden-Rossendorf, Germany
Non-van der Waals Heterostructures from an Integrated Computational Approach

P111 **Oscar Obel** — University of Heidelberg, Germany
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P112 **Aykut Öztürk** — Technische Universität Berlin, Germany
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P113 **Deepak Ojha** — Potsdam University, Germany
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P114 **Róża Okoń** — University of Warsaw & Wrocław University of Science and Technology, Poland
Capturing emergent behaviour in complex reaction networks through quantum chemistry-based kinetic modelling

P115 **Jan Christoph Ortlepp** — Heinrich Heine University Düsseldorf, Germany
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P116 **Tomáš Ovad** — University of Chemistry and Technology, Prague, Czechia
Electron attachment as a route to exotic reactivity: the case of sulfur heterocycles

P117 **Ram Dhari Pandey** — Nicolaus Copernicus University in Toruń, Poland
Heteroatom Doping for Enhanced Domain-Based Charge Transfer in Organic Dyes

P118 **Giovanni Parolin** — Heidelberg University, Germany
REMP up your ADC

P119 **Rémi Pasquier** — University of Regensburg, Germany
Optimized grids for the real-space resolution of the identity in low-scaling GW

P120 **Sandip Paul** — Indian Institute of Technology Kanpur, India
Simulating Chloroalkane-Methanol Binary Liquid Mixtures and Correlating with Femtosecond Thermal Lens Spectroscopy

P121 **Sebastian Pauly** — Heidelberg University, Germany
Understanding the Electronic Structure of a Radical-Functionalized Azobenzene MOST Candidate

P122 **Michael Peintinger** — Mulliken Center for Theoretical Chemistry, Germany
Development and benchmarking of vibe-qc, a molecular and periodic electronic-structure code targeting correlated methods for solids

P123 **Luca Nils Philipp** — University of Würzburg, Germany
High-Order Response Functions with Duschinsky Coupling and Finite Temperature: Application to Two-Dimensional Resonance Raman Spectroscopy

P124 **Fabian Pieck** — Leipzig University, Germany
Combining DFT, MLP, and Monte-Carlo Approaches to Model Area-Selective Deposition Processes

P125 **Annelene Plump** — University of Bremen, Germany
From Hydrostatic to Anisotropic Compression: The Development of two new Mechanochemical Simulation Methods

P126 **Pavel Pokhilko** — University of Vienna, Austria
SHARC meets Q-CHEM: interface for TDDFT dynamics with non-adiabatic and spin-orbit couplings for arbitrary states

P127 **Jan Polena** — University of Chemistry and Technology, Prague, Czech Republic
Can a solvated electron reach the reactant?

P128 **Christoph Li Popko** — Heidelberg University, Germany
Tensile Testing on an Atomistic Scale: Gold–Carbene Interfaces under Mechanical Strain

P129 **Johannes Lukas Post** — University of Düsseldorf, Germany
Spin–Orbit Coupling in TDDFT: From the Tamm–Dancoff to the Random Phase Approximation

P130 **Ulrich Pototschnig** — University of Hamburg, Germany
Electron Dynamics and Spin Selectivity in Chiral Molecules and Interfaces

P131 **Peter E. Hartmann** — University of Graz, Austria
Molecular Insights into Alkene Hydrogenation with Manganese(I) NacNac Catalysts

P132 **Fabio Kratzer** — University of Graz, Austria
Deciphering the Solvent Dependence of Hydrogen Generation from Seawater and Silicon Waste

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P133 **Anne-Marie Kelterer** — Graz University of Technology, Austria
Wavelength-Controlled Fragmentation of Mixed-Functionalized Norrish Type-I Acylgermanes

P134 **Pragya Pudasaini** — Humboldt-Universität zu Berlin, Germany
Probing Oxyl Character and Reactivity in a Biomimetic Non-Heme Oxoiron(IV) Complex

P135 **Vania Martins Ramos** — Universidade Federal de São Carlos, Brazil
Systematic Active-Space Construction for the Multiconfigurational Description of a Mn(I) Photosensitizer Candidate

P136 **Sheetal Ranaut** — Indian Institute of Technology Jammu, India
Machine Learning-Guided Molecular Insights into Metal-Catalyzed Asymmetric Reactions: A Case Study with a Pharmaceutically Relevant Version of Suzuki-Miyaura Coupling

P137 **Hamza Rappoun** — Paderborn University, Germany
Spatially Resolved Vibrational Dynamics of Water in Aqueous $Ba(SCN)_2$ Using Machine-Learned Potentials

P138 **Vyshna Ratheesh** — IWR, University of Heidelberg, Germany
Controlling Excited States of Anthracenes through Heteroatom Substitution

P139 **Sreejita Ray** — Freie Universität Berlin, Germany
Combinatorial Structure Search and First-Principles Stability Analysis of Borophene Phases on Ag(111)

P140 **Kassandra Rensinghoff** — Universität Münster, Germany
Vibrational Spectroscopy of Molecules in High Pressure Enviroments with GOSTSHYP

P141 **Charlotte Rickert** — MPI Solid State Research, Germany
The Aperiodic Defect Model: Treating Aperiodic Defects in Fully Periodic Systems with Wavefunction Methods

P142 **Anton Rikus** — University of Münster, Germany
How good are excited-state geometries from subsystem TDDFT?

P143 **Lukas Sebastian Rindt** — Heidelberg University, Germany
Re-purposing Optimization Trajectories: Do Hessians from Quasi-Newton Schemes Yield Reasonable Free Energies?

P144 **Christopher-Matthias Röper** — Universität des Saarlandes, Saarbrücken, Germany
A Perturbative Triples Approach for EOM-CC-(D)EA/(D)IP Treatments of Excited States in Strong Magnetic Fields

P145 **Marco Romanelli** — University of Vienna, Austria
Quantum-classical non-adiabatic dynamics in extended solid-state materials: a SHARC-VASP approach

P146 **Bibek Samal** — University of Regensburg, Germany
Perturbative Spin-Orbit Coupling in CP2K for Molecules, Bulk Materials, and Surfaces

P147 **Joana-Lysiane Schäfer** — Freie Universität Berlin, Germany
Girsanov path reweighting for core set Markov state models

P148 **Timo Scharfe** — University of Augsburg, Germany
FrayedEnds: Basis-Set-Free Quantum Chemistry

P149 **Jonathan Scherlitzki** — Freie Universität Berlin, Germany
A Novel Variational Quantum Trajectory Approach to Nuclear Quantum Dynamics

P150 **Julian Schiefer-Strauß** — University of Innsbruck, Austria
Calculation of Energy Potential Surfaces and Absorption Spectra of Rubidium-Noble Gas Mixtures

P151 **Thomas Schnappinger** — Stockholm University, Stockholm, Sweden
The Cavity-Born-Oppenheimer Framework: Electronic Reshaping and Multireference Effects under Vibrational Strong Coupling

P152 **Friederike Schneider** — Heidelberg University, Germany
Can Triple Excitations Restore Spin Symmetry of Excited States within the ADC Framework?

P153 **Leopold Seidler** — University of Bonn, Germany
Periodic g-xTB — Approaching DFT Accuracy for Periodic Systems at Semiempirical Cost

P154 **Faezeh Shabani** — TU Chemnitz, Germany
From Single α-Helices to Helical Bundles: Electronic Properties and Stability

P155 **Akhilesh Sharma** — Indian Institute of Technology Roorkee, India
Conversion of Light Alkanes into Value–Added Products Over Boron–Based Catalysts

P156 **Omid Shayestehpour** — Paderborn University, Germany
Prokyon – An Open-Source Library for On-the-Fly Machine Learning of Interatomic Potentials

P157 **Kristoffer Simula** — Max Planck Institute for Solid State Research, Germany
Transcorrelation in Solid-State Matter

P158 **Ramandeep Singh** — University of Münster, Germany
Pressure induced luminescence shift in coordination compounds

P159 **Natalia Skałecka** — University of Warsaw, Poland
Constructing systematically convergent Gaussian basis sets for solids

P160 **Stepan Sklenak** — J. Heyrovsky Institute of Physical Chemistry of the Czech Academy of Sciences, Prague, Czech Republic
DFT investigation of the effects of hydration on the reactivity of the low-valency distant binuclear vanadium species balanced in zeolites

P161 **Nathalie Smith** — University of Hamburg, Germany
Neural Network Potentials of 2D Nanoconfined Water: Efficient Training Set Construction and Calculation of Dielectric Properties

P162 **Richard Springborn** — Ruhr-Universität Bochum, Germany
Enhancing the Transferability of Machine Learning Potentials by Training on Atomic Properties

P163 **Ilija Srpak** — Max Planck Institute for Solid State Research, Stuttgart, Germany
A new Molecular Dynamics approach for treating spin-phonon effects in solids

P164 **Luca Steiner** — Freie Universität Berlin, Germany
Fluorine-Specific π-Stacking Accelerates Ring-Opening Copolymerisation

P165 **Tom Erik Steinkopf** — Martin-Luther-Universität Halle-Wittenberg, Germany
Configurational Entropy of Mixing as a Spatially Resolved, Time-Dependent Observable

P166 **Michael Stier** — University of Stuttgart, Germany
Reliable Redox-Potential Simulations of Proteins

P167 **Jesús Lucia Tamudo** — University of Vienna, Austria
Breaking Harmonics Walls: The Anharmonic Vibronic Coupling Approach in Trajectory Surface Hopping Simulations

P168 **Balaram Thakur** — Technische Universität Dresden, Helmholtz-Zentrum Dresden-Rossendorf, Germany
Coordination Corrected Enthalpies for the Thermodynamics of Ionic Materials

P169 **Franz Thiemann** — Leipzig University, Germany
Interactions in N-Heterocyclic Carbene Monolayers on Cu(111) and Au(111) from Periodic Energy Decomposition Analysis and NOCV

P170 **Vishwas Tiwari** — University of Paderborn, Germany
Atomistic Insights into Si(100) Surface Passivation by Water: ReaxFF Molecular Dynamics and Fine-Tuned Machine Learning Interatomic Potential Simulations.

P171 **Johannes Tölle** — University of Hamburg, Germany
Benchmarking Tensor Hypercontraction Techniques for Molecular Systems

P172 **Kai Töpfer** — Freie Universität Berlin, Germany
Machine-Learning Supported Reaction Mechanism Exploration of cis-trans Isomerisation in Retinal

P173 **Aleksandar Trajkovski** — University of Bonn, Germany
Implementations of Molecular Integral-Equation Solvation Models: From 6D-MOZ to RISM-SCF

P174 **Thuy Truong** — University of Augsburg, Germany
Excited-State Pair Natural Orbitals for Shallow System-Adapted Quantum Circuits

P175 **Iordanis Tsakontsis** — Bielefeld University, Germany
Accurate treatment of hyperfine coupling in the diabatic potential model of HI

P176 **Ritaj Tyagi** — University of Regensburg, Germany
Accelerating Large-Scale GW Calculations using Real-Space Resolution of Identity

P177 **Anna Ulrich** — Heidelberg University, Germany
Molecular switches and machines

P178 **Anna Luisa Upterworth** — Martin Luther University Halle-Wittenberg, Germany
Philicity at Play: Entropy, Enthalpy and Miscibility in Alkane–Perfluoroalkane Mixtures

P179 **Natália Lussari Vrech** — Universität Bremen, Germany
Low-Spin Co(III) Stability in Cobalamin Derivatives Under Hydrostatic Pressure: A Computational Investigation

P180 **Henry Wang** — Ruhr-Universität Bochum, Germany
Simulations of Tricalcium Silicate-Water Interfaces using High-Dimensional Neural Network Potentials

P181 **Nils Wegerich** — University of Frankfurt, Germany
Quantum Chemical Study of the Reactivity of a Platinum(II)nitrene with 1,4-Benzoquinone

P182 **Anna Marleen Weidlich** — Heidelberg University, Germany
Absorption spectra of N-heteroacenes

P183 **Jonas Weinhold** — Karlsruhe Institute of Technology, Germany
Spin-State Switching in Trinuclear Exchange-Coupled Systems

P184 **Jonas Weiser** — University of Augsburg, Germany
Extending an Automated Ensemble-Averaged Kinetics Framework for Reverse Intersystem Crossing in Flexible TADF Emitters

P185 **Rahel Weiß** — University of Bremen, Germany
Interactive JEDI Strain Analysis as a Pre-Screening Tool for Mechanophore Design

P186 **Christian Wiebeler** — University of Augsburg, Germany
Molecular Dynamics and Computational Spectroscopy for Systems in Soft Condensed Phase

P187 **Tillmann Lukas Wigger** — Heinrich Heine University Düsseldorf, Germany
Automated Reaction detection with different Bond Order Schemes

P188 **Severin Wittek** — Humboldt University of Berlin, Germany
Improving the scaling of AC0 with laplace-THC

P189 **Lukas Wittmann** — University of Bonn, Germany
A Minimally Empirical and Self-Consistent Implicit Solvation Model for Robust Quantum Chemistry in Solution

P190 **Konstantinos Zois** — University of Vienna, Austria
Simulating a bimetallic rotaxane with classical molecular dynamics

P191 **Carsten Zülch** — Philipps-Universität Marburg, Germany
Small molecular ions as probes for variations of fundamental constants

P192 **Hana Zupan** — Freie Universität Berlin, Germany
Grid-Based Description of Molecular Association

P193 **Christopher Zurek** — RWTH Aachen University, Germany
Accelerating Conformer Reranking with Machine Learning Interatomic Potentials

P194 **Tom-Luka Zwarg** — Heinrich-Heine University, Düsseldorf, Germany
Computational discovery of the initial formation processes of the solid-electrolyte interface and the role of novel additives

P195 **Jan Leodolter** — Graz University of Technology, Austria
Automated Assembly and Characterization of Coordination Cages

P196 **Tobias-Erik Monz** — University of Graz, Austria
Thermodynamic Properties of Various Polymorphs of 2-Thiobarbituric Acid via Quasi-Harmonic Calculations

P197 **Oskar Schweighofer** — University of Graz, Austria
Detailed Pathways of Chemical Reactions in Molecular Crystals Using MLFFs

Abstracts: Invited Talks

I1

Schrödinger in Graz

David Clary[1]

[1] *University of Oxford, United Kingdom*

2026 is the centenary of Schrödinger's great paper which introduced wave mechanics. It is also the 90th anniversary of Schrödinger's move from Oxford to Graz in 1936. Building on my recent biography *Schrödinger in Oxford*, I will describe Schrödinger's life, career and scientific contributions with particular emphasis on the difficult two years he spent in Graz [1].

References

[1] D. C. Clary, *Schrödinger in Oxford* (World Scientific Publishing, 2022).

I2

Learning Reaction Mechanisms from Molecular Dynamics: Isomerization in Retinal and p–Coumaric Acid

Bettina Keller[1]

[1] *Freie Universität Berlin, Germany*

Accurate reaction coordinates are essential for understanding chemical reactions, yet identifying them remains a longstanding challenge. While free–energy landscapes and minimum–energy paths are widely used for this purpose, they need not coincide with the pathways actually followed by reactive trajectories. The committor provides a rigorous, dynamics–based reaction coordinate because it directly measures the probability of reaching products before reactants. For molecular reactions, the committor is difficult to determine and even harder to represent in terms of physically interpretable molecular coordinates. Recent developments combining transition–path sampling with machine–learned committors now make it possible to characterize reactive trajectories directly and to identify the molecular motions that control their dynamics.
In this talk, I will present an analysis of the thermal *cis–trans* isomerizations of retinal and p–coumaric acid using "Artificial Intelligence for Molecular Mechanism Discovery" (AIMMD). In these apparently simple double–bond rotations one might expect the central torsion and the free–energy landscape to largely determine the reaction pathway. Instead, the transition–path ensembles reveal strongly curved, stepwise S–shaped pathways involving the coupled motion of four dihedral angles around the reactive bond. This S–shape is absent from the minimum–free–energy path: it arises from the dynamics of the short transition events combined with the mass asymmetry between heavy–atom and hydrogen–bearing dihedrals. These results highlight that reaction pathways cannot be inferred from the potential or free–energy surface alone but require an explicit description of the underlying dynamics and that committor–learning and transition–path techniques allow us to move beyond a static description of reaction pathways.

References

[1] K. Töpfer, G. Lazzeri, V. Ossanna, F. Renner, G. Lattanzi, R. Covino, B. G. Keller, *arXiv preprint arXiv:2604.24245* **2026**.

I3

Adsorbates at periodic metal and insulating surfaces: From structure to non-equilibrium dynamics

Peter Saalfrank[1]

[1] *University of Potsdam, Germany*

The theoretical modelling of atoms and molecules adsorbed at solid surfaces is key to the understanding of important physicochemical phenomena and applications, ranging from heterogeneous catalysis over surface femtochemistry and electrochemistry to astrochemistry, to name just a few. In this talk, I will review some of our work over the last decades on the structure, spectroscopy, and – mostly photoinduced – dynamics, of adsorbates at periodic metal and insulating surfaces. Underlying methods will be covered "on the fly". Specific examples include quantum size effects in thin metal films [1,2], femtosecond-laser driven, "hot-electron" mediated dynamics and reactions at metal surfaces [3,4,5], and, finally, the vibrational spectroscopy, dynamics and kinetics of small molecules at insulator surfaces, notably for CO on NaCl [6,7,8].

1. G. Materzanini, P. Saalfrank, and P.J.D. Lindan, Phys. Rev. B **63**, 235405 (2001).
2. P. Saalfrank, J.I. Juaristi, M. Alducin, M. Blanco-Rey, and R. Díez Muiño, J. Chem. Phys. **141**, 234702 (2014).
3. A. Tetenoire, Ch. Ehlert, J.I. Juaristi, P. Saalfrank, and M. Alducin, J. Phys. Chem. Lett. **13**, 8516 (2022).
4. S. Lindner, I. Lončarić, L. Vrček, J.I. Juaristi, M. Alducin, and P. Saalfrank, J. Phys. Chem. C **127**, 14756 (2023).
5. B. Mladineo, J.I. Juaristi, M. Alducin, P. Saalfrank, and I. Lončarić, J. Chem. Phys. **163**, 044103 (2025).
6. A.D. Boese and P. Saalfrank, J. Phys. Chem. C **120**, 12637 (2016).
7. S. Sinha, B. Mladineo, I. Lončarić, and P. Saalfrank, J. Phys. Chem. C **128**, 22117 (2024).
8. A. Choudhury, J.A. DeVine, S. Sinha, J.A. Lau, A. Kandratsenka, D. Schwarzer, P. Saalfrank, and A.M. Wodtke, Nature **612**, 691 (2022).

I4

How Atoms Interact Within Molecules

Alexandre Tkatchenko[1]

[1] *University of Luxembourg, Luxembourg*

Fundamental understanding of interatomic forces in molecules must emerge from quantum mechanics, yet widely used empirical force fields rely on simplified mechanistic approximations that often fail to capture the complexity of many-body systems. Here we employ recent developments in quantum field theory (QFT) for long-range electron correlation and machine learning force fields (MLFFs) to directly compute the depth and scatter of interatomic forces for molecular systems containing hundreds of atoms. We find that while the average interaction strength decays polynomially with interatomic separation, the interaction scatter remains robust and exhibits substantial anisotropy. Both QFT and MLFFs demonstrate that increasing the molecular size further amplifies this scatter and anisotropy – a phenomenon not considered in traditional textbook empirical models. These results provide new benchmarks for force models, shift the focus from interacting atoms to interacting "hotspots" that might determine the folding pathways of (bio)polymers, and rationalize why MLFFs are uniquely successful in capturing the nuances of complex molecular systems. Our findings offer a roadmap for the development of more accurate and quantum-aware molecular force fields.

I5

Status and Future Challenges for periodic Coupled-Cluster Simulations in Materials

Andreas Grüneis[1]

[1] *TU Wien, Austria*

Coupled-cluster (CC) theory is regarded as a gold-standard approach for correlated molecular systems, but its extension to materials poses significant conceptual and computational challenges. Recent advances are enabling increasingly realistic CC simulations of solids and surfaces, offering systematically improvable predictions of electronic and structural properties. In this talk, I will review progress in periodic CC theory, focusing on long-range correlation and the treatment of metals, applications to adsorption on metallic surfaces, and equation-of-motion CC calculations of charged excitations and band gaps. Particular attention will be given to finite-size effects and the thermodynamic limit, which remain central challenges for quantitative calculations of extended systems. I will also discuss how machine-learned force fields can extend CC-level accuracy to lattice dynamics and larger length and time scales. Finally, I will highlight open questions concerning computational scaling, finite-size and basis-set errors, higher-order correlation, and the reliability of approximations for large and polarizable systems. Addressing these challenges will be essential for establishing coupled-cluster theory as a broadly applicable predictive framework for materials.

References:
Masios et al., Phys. Rev. Lett. 131, 186401 (2023);
Carbone et al., Faraday Discuss. 254, 586–597 (2024);
Moerman et al., J. Chem. Theory Comput. 21, 1865–1878 (2025);
Moerman et al., Phys. Rev. B 111, L121202 (2025);
Schönbauer et al., J. Chem. Theory Comput. 22, 6906–6916 (2026);
Schäfer et al., Nat. Commun. 16, 9108 (2025).

I6

From Molecules to Materials: Effective Hamiltonians for Spin Dynamics and Magnetic Exchange

Leticia González[1]

[1] *University of Vienna, Austria*

Complex molecular and materials systems often involve multiple interacting electronic and spin degrees of freedom, making the connection between electronic structure and experimental observables challenging. While electronic-structure calculations provide access to the underlying states and interactions, their dynamics and collective behavior often become tractable only when this information is mapped onto suitable effective Hamiltonians. In this talk, I will illustrate how such Hamiltonians can be exploited to describe distinct manifestations of electronic and spin coupling, in molecules and materials. Specifically, I will discuss the photoinduced spin crossover of the open-shell Fe(III) complex $[Fe(qsal)_2]NCS$ and the spin interactions in triangular $Fe(III)_3O$-based metal–organic frameworks. In the first case, a linear vibronic coupling Hamiltonian parametrized from multistate RASSCF calculations enables nonadiabatic dynamics including spin–orbit coupling, providing insight into the coupled electronic and nuclear dynamics underlying ultrafast spin crossover. In the second, broken-symmetry DFT combined with spin-Hamiltonian reconstruction is used to unravel the origin and consequences of magnetic frustration. Finally, I will discuss ongoing efforts to extend nonadiabatic molecular dynamics from molecules to periodic materials.

I7

Long-range magnetic order in metal-free two-dimensional materials

Hongde Yu[1], Solveig M. Oelke[1], Thomas Heine[1]

[1] *TU Dresden, Germany*

Magnetism is typically connected with metal atoms, and long-range magnetic ordering with spin-orbit coupling. We present a new concept to generate strong ferro- and antiferromagnetic coupling in metal-free materials based on radical polyaromatic building blocks such as heterotriangulenes, which are arranged in regular planar honeycomb-kagome lattices.
We will share our theoretical considerations for the design of metal-free magnetism in 2D systems. Following basic considerations (Lieb theorem or Ovchinnikov rule), systems made of pure carbon lattices such as coupled triangulenes will give antiferromagnetic ground states. We will present two routes to achieve strong ferromagnetic couplings. In the first one, we will utilize heterotriangulanes. Due to the presence of foreign atoms, the Ovchinnikov rule is violated and ferromagnetic coupling is possible. We observed cases with coupling constants above 50 meV. A second route is to construct heterostructured lattices made of two different building units, so that the spins reside in the same graphene-like sublattice and the Ovchinnikov rule holds. We will present systems with Curie temperatures above 500K.
Finally, we present a strategy for the stabilization and synthesis of metal-free magnetic systems based on a switching strategy between zwitterion and biradical.

1. H. Yu, J. Sun, T. Heine, J. Chem. Theory Comput. 19 (2023) 3486-3497
2. H. Yu, T. Heine, J. Am. Chem. Soc. 145 (2023) 19303-19311
3. H. Yu, T. Heine, Sci. Adv. 10 (2024) eadq7954
4. H. Yu, Y. Jing, T. Heine, Acc. Chem. Res. 58 (2025) 61-72
5. H. Yu, T. Heine, J. Am. Chem. Soc. 148 (2026) 13822-13833
6. S. M. Oelke, H. Yu, T. Heine, chemrxiv, https://doi.org/10.26434/chemrxiv.15005836/v1

I8

Relativistic GW for solids and molecules

Vibin Abraham[1], Jacob Adamski[1], Gaurav Harsha[1], Pavel Pokhilko[1], Dominika Zgid[1,2]

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[2] *Physics Department, University of Michigan, Ann Arbor, MI, 48109, USA*

The fully self-consistent GW (scGW) method with an iterative solution of the Dyson equation provides a consistent approach for describing the ground and excited states without any dependence on the mean-field reference. In this work, I will discuss a relativistic version of scGW for molecules and solids containing heavy elements using the exact two-component (X2C) Coulomb approximation. We benchmarked the SOC-81 data set containing closed shell heavy elements for the first ionization potential using the fully self-consistent GW as well as one-shot GW. The self-consistent GW provides superior results compared to G0W0 with PBE reference while removing the starting point dependence. The photoelectron spectra obtained at the X2C level demonstrate very good agreement with the experimental spectra. We also observe that scGW provides very good estimation of ionization potential for the inner d-shell orbitals. Additionally, using the total energy, we investigate the equilibrium bond length and harmonic frequencies of a few halogen dimers using scGW. Overall, our findings demonstrate the applicability of the fully self-consistent GW method for accurate ionization potential, photoelectron spectra, and total energies in finite systems with heavy elements with a reasonable computational scaling. Finally, I will discuss X2C-1e scGW calculations for solids such as semiconductors/metals, conventional insulators, and topological insulators. Here we will analyze conventional properties such as band structure, equilibrium lattice constant and bulk modulus in addition to discussing the zeros of Green's functions and leading a novel analysis applicable to discovering properties of novel topological materials.

References

[1] Chia-Nan Yeh, Avijit Shee, Qiming Sun, Emanuel Gull, Dominika Zgid, Relativistic self-consistent : Exact two-component formalism with one-electron approximation for solids, PRB, 106, 085121

[2] Vibin Abraham, Gaurav Harsha, Dominika Zgid, Relativistic Fully Self-Consistent GW for Molecules: Total Energies and Ionization Potentials, J. Chem. Theory Comput. 2024, 20, 11, 4579–4590

[3] Harsha Gaurav, Abraham Vibin, Dominika Zgid, Challenges with relativistic GW calculations in solids and molecules, 10.1039/D4FD00043A (Paper) Faraday Discuss., 2024

I9

Let them learn: AI models that master materials physics

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Machine learning is transforming the way we perform simulations to predict and understand the properties of materials. Traditional "physics-informed" models build symmetry, smoothness, and other physical priors into the mathematical structure of the model to guide learning. In this talk, I'll explore how much physics we really need to include when teaching machines the quantum behavior of matter. I'll contrast models constrained by physical assumptions with emerging, unconstrained approaches that learn physical relationships directly from data. These models can reach, and sometimes exceed, the accuracy and efficiency of their physics-based counterparts, though they require some care to avoid unphysical results. I'll illustrate these ideas using PET-MAD, a lightweight, data-driven model that learns across the periodic table, providing accurate predictions of the microscopic properties of materials that include a quantification of the model uncertainties, both against the reference electronic-structure calculations, and against experiments.

I10

Molecule-2D heterostructures - still a challenge for computational chemistry

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Low-dimensional materials have garnered significant attention due to their exceptional physical and chemical properties. Heterostructures consisting of such materials with specially designed molecules allows the finetuning of properties as a result of charge transfer, both via physisorption or covalent functionalization. We investigated two different types of 2D materials, graphene with its semimetallic behaviour and MoS2 as a semiconductor with strongly bound excitons. The interaction of these 2D materials with various electron acceptor and donor molecules as suitable n- and p-type dopants was studied. As a first outcome, graphite and bilayer graphene were investigated with the strong electron-accepting FeCl3 as an intercalant. Both the electronic and magnetic properties can be adjusted by different stacking sequences and by applied strain [1]. Furthermore, the intercalation of halogens in graphitic materials supports the formation of polyhalogen anions in confined space due to charge transfer. Additionally, chemical modification of the acceptor and donor molecules and their arrangements are investigated with respect to their doping affinity of graphene. The same acceptors and donors are also used for charge transfer to and from MoS2, where both single and double side functionalization was investigated [2]. With the careful selection of donors and acceptors on either side, an increase of the doping to MoS2 could be achieved. Double side covalent functionalization of MoS2 can control the electronic and the optical band gaps [3]. Depending on the choice of the chemical groups and their coverage, the electronic properties can be tuned from semiconducting to metallic and the optical band gap can be shifted by up to 1 eV. For all these calculations, a careful selection of the DFT functional applied, supercell size and computational parameters is necessary. Additionally, to get reliable results for electronic and optical band gaps, the GW method and the BSE approach has to be applied on top of the Kohn-Sham DFT results.

[1] J. Dai, S. Yadav, B. Paulus, C. 2023, 9(4), 95
[2] S. Ray, B. Paulus, Nanomaterials 2025, 15(3), 193
[3] K. Wang, M. Kapitzke, L. Green, B. Paulus, J. Mater. Chem. C 2022, 10, 6009

I11

Accelerated Materials Discovery with Machine-Learning and DFT

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Lead-free double perovskites with the composition A2BB'X6 have emerged as a promising new class of materials for photovoltaic applications as alternative to the lead- containing halide perovskites. They are stable for a wide range of chemical compositions on the different sublattices, which allows tuning of desired properties. However, efficient exploration of this enormous chemical space requires innovative machine-learning and high-throughput methods. In a first example we considered only two elements in different ratios on both sublattices B and B'. By using a Minimum Redundancy Maximum Relevance (MRMR) feature selection algorithm and Gaussian Process Regression (GPR) we show that important photophysical parameters such as recombination rates and Stokes shifts can be predicted from a large data set of ground state properties, which was generated by high-throughput DFT calculations [1]. Next, we allowed many different elements on all sublattices to create so-called high entropy compositions with increased thermodynamic stability due to the gain of configurational entropy. A Sobol sequence is used to generate a minimal set of representative combinations capable of describing the broader compositional space. By combination with a Trust Region Bayesian Optimization (TuRBO), subsequent iterations are efficiently guided, reducing the computational load from thousands of calculations to mere hundreds. This streamlined approach allows us to identify compositions for well-behaved semiconductors with optimized band gap and effective electron and hole masses, providing a powerful pathway for accelerating the discovery of high-performance, lead-free double perovskites.

[1] M.S. Günthert, L. Lüer, O. Stroyuk, O. Raievska, C. Kupfer, A. Osvet, B. Meyer, C.J. Brabec, Digital Discovery 5 (2026) 1215–1227.

Abstracts: Contributed Talks

C1

Wave functions are mapped in intermolecular potential energy surfaces of π-stacked aggregates

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$\pi-\pi$ interactions are of utmost importance for life- and material-science but their nature is still controversially debated. The existing discrepancies are raionalized by (i) Demonstrating that $\pi-\pi$ interactions reflect the wave function properties of repulsive interactions in a particularly pronounced manner (ii) Showing that the resulting fluctuations in repulsive interactions lead to the counterintuitive behavior of repulsive and attractive components in the interaction energy.
The wave function character of repulsive interactions is rationalized by partitioning the exchange-repulsion contribution to the interaction energy between two closed shell systems into molecular orbital pair contributions to the exchange (MOPCE) energy [1,2]. This reveals that the repulsion in π-stacked dimers is governed by repulsion of the π-orbitals on the respective systems. For constant interplanar distances MOPCE terms are essentially proportional to the squared overlap integral of the respective orbitals, which causes that the exchange-repulsion interactions are mapping spatial oscillations of the interacting wave functions. Our model resembles frontier orbital theory and allows to reproduce energetically favorable arrangements of π-stacked systems such as benzene- or pentacene-dimers [3] as well as donor-acceptor systems such as benzene and para-benzoquinone.
Furthermore, we explain why the importance of attractive and repulsive contributions to the interaction of two molecules frequently leads to controversal debates. Repulsive interactions commonly decrease exponentially with the distance of interacting systems while attractive interactions behave smoother e.g. as a power of the inverse distance. This causes that arrangements with reduced repulsion will give rise to a decreased equilibrium distances for which the repulsive interactions are generally increased. This repulsion paradoxon: less repulsion gives more repulsion! is shown to be relevant for various $\pi-\pi$ interactions and halogen bonding.

[1] J. Henrichsmeyer, M. Thelen, M. Bröckel, M. Fadel, S. Behnle, M. Sekkal-Rahal, and R. F. Fink, ChemPhysChem, 2023, e202300097.
[2] J. Henrichsmeyer, M. Thelen, and R. F. Fink, ChemPhysChem, 2024, e202400887.
[3] M. Thelen, J. F. Henrichsmeyer, A. Buchwald, and R. F. Fink, J. Am. Chem. Soc. 2026, 148, 16151.

C2

Reformulation of intermolecular perturbation theory using imaginary time

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It is shown that perturbation theory for interacting molecules within the framework of the interaction picture in quantum mechanics can be reformulated through integrals over products of imaginary time cross correlation functions, providing a natural way for its factorization. Thus in second order an alternative expression for the London dispersion energy contribution is obtained in which the standard Casimir-Polder integration over products of susceptibilities at imaginary frequencies is replaced with integration over products of cross correlation functions at imaginary frequencies [1]. Thanks to a relation of the latter to the Green's function of the unperturbed Hamiltonian in principle they are accessible through, e.g., Green's function or diffusion quantum Monte Carlo, numerical split operator imaginary time propagation, or other numerical and analytical path integral methods. In some cases like that of the harmonic oscillator they could even be determined analytically. Furthermore, susceptibilities and cross correlation functions can be related through a Fourier cosine transform. Besides a derivation and a discussion of the revamped second-order contributions extensions of the imaginary time approach to higher orders of perturbation theory will be presented.

[1] G. Jansen, "Factorization of static perturbation theory for weakly coupled systems using imaginary time", Phys. Rev. A 113, L030201 (2026).

C3

Excited-state dynamics using the SHARC4 package

S. Mai[1], S. Mausenberger[1], S. Polonius[1], T. Piteša[2], D. Vörös[1], L. Grünewald[1], L. González[1]

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Nonadiabatic dynamics simulations in the excited electronic states are nowadays essential for research in spectroscopy, photochemistry and -biology, and materials science. One of the most widely used nonadiabatic dynamics methods is trajectory surface hopping, which is the core method implemented in the SHARC (Surface Hopping including ARbitrary Couplings) package. The recent updates to versions 4.0 and 4.1 include a wide array of new methodological options. Most importantly, the new hierarchical electronic structure interface framework [1] enables multiscale modeling of extended systems via QM/MM and LVC/MM [2], the description of multi-chromophoric systems [3], the sampling of excited-state free energy surfaces [4], automated machine learning training [1], the modeling of the excitation process with realistic laser pulses [5], and many other possibilities. In my talk, I will provide an overview of the SHARC4 package and its methods and show corresponding applications.

[1] S. Mausenberger, S. Polonius, S. Mai, L. González: Efficient, Hierarchical, and Object-Oriented Electronic Structure Interfaces for Direct Nonadiabatic Dynamics Simulations, J. Chem. Theory Comput. 21, 8994–9008 (2025), DOI: 10.1021/acs.jctc.5c00878
[2] S. Polonius, O. Zhuravel, B. Bachmair, S. Mai: LVC/MM: A Hybrid Linear Vibronic Coupling/Molecular Mechanics Model with Distributed Multipole Electrostatic Embedding for Highly Efficient Surface Hopping Dynamics in Solution, J. Chem. Theory Comput. 19, 7171–7186 (2023), DOI: 10.1021/acs.jctc.3c00805
[3] T. Piteša, S. Mai, L. González: Efficient Excitonic Configuration Interaction for Large-Scale Multichromophoric Systems Using the Resolution-of-Identity Approximation, J. Phys. Chem. Lett. 16, 2800–2807 (2025), DOI: 10.1021/acs.jpclett.5c00065
[4] D. Vörös, H. G. Gallmetzer, J. Dietschreit, S. Mai, L. González: Umbrella Sampling for Excited States using a Semiempirical Method, J. Am. Chem. Soc. Au 6, 3547–3569 (2026), DOI: 10.1021/jacsau.6c00302
[5] L. Grünewald, L. van Dam, S. Mai: An Excitation Strategy for the Initial Condition Generation for Surface Hopping Trajectories using Electron-only Dynamics including Explicit Laser Pulses, J. Chem. Theory Comput. 21, 12741–12754 (2025), DOI: 10.1021/acs.jctc.5c01390

C4

From Biased Trajectories to Local Mobility: Residence Times as a Route to Diffusivity

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A molecular trajectory contains a detailed record of motion, but turning that record into a position-dependent diffusivity is far from straightforward. In heterogeneous systems, observed motion reflects both local mobility and free-energy-driven drift, which must be disentangled when the dynamics are reduced to a single transport coordinate. Established methods either alter the dynamics through harmonic confinement or introduce choices involving lag times, spatial discretization, regularization, parametrization, or the long-time limits of noisy correlation functions.
I will present a residence-time approach for determining position-dependent diffusivities directly from biased molecular trajectories [1]. Once an adaptive bias has approximately removed the free-energy gradient along the transport coordinate, local diffusivities can be inferred from mean first-exit times out of finite spatial intervals. The estimator requires no additional restrained simulations, kinetic model fitting, or correlation-function extrapolation. Because residence-time observations are strongly correlated, blocking analysis provides statistically consistent uncertainty estimates.
The method is assessed for systems of increasing complexity: oxygen diffusion in a liquid slab, water permeation through a fluid phospholipid bilayer, and transport of water and volatile organic compounds through a highly ordered skin-barrier model. The latter is particularly demanding because slow structural relaxation and long-lived membrane asymmetries complicate both free-energy and diffusivity calculations [2,3]. In the liquid slab, the inferred diffusivities recover independent bulk values within statistical uncertainty. In the membrane systems, they undergo a more stringent test through propagator-level validation: the diffusivity profiles are combined with the free-energy landscape to predict time-dependent probability distributions, which are compared directly with molecular trajectories.
These results show how residence-time statistics can turn enhanced-sampling trajectories into spatially resolved descriptions of molecular mobility. The talk will examine the assumptions behind this reduction, where it succeeds, and how its validity can be tested rather than presumed.

References

[1] R. Thomas, P. R. Prabhakar, M. von Domaros, *arXiv* (2026), DOI: 10/q59x (Under review in *J. Chem. Theory Comput.*).
[2] R. Thomas, P. R. Prabhakar, D. J. Tobias, M. von Domaros, *ACS ES&T Air* **3**, 580–589 (2026), DOI: 10/hbpnjd.
[3] R. Thomas, P. R. Prabhakar, D. J. Tobias, M. von Domaros, *J. Phys. Chem. B* **129**, 1784–1794 (2025), DOI: 10/g9wqtk.

C5

A Multifidelity Monte Carlo Approach for Simulating the Diffusion Coefficient of Water

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Self-diffusion coefficients computed from molecular dynamics simulations are known to suffer from systematic finite-size effects. To mitigate this dependence, additive correction terms have been proposed. [1,2] More recently, the OrthoBoXY approach [3] introduced tetragonal simulation boxes with specific aspect ratios for which the correction term vanishes.
In this contribution, we present an alternative strategy: a multifidelity Monte Carlo framework for the computation of diffusion coefficients of water that explicitly exploits, rather than corrects for, the size dependence. [4] Multifidelity methods, originally developed in the field of uncertainty quantification, combine computational models that evaluate the same quantity of interest—in this case the diffusion coefficient—at different levels of accuracy and computational cost. [5] In our framework, the model hierarchy is defined by the size of the simulation boxes. Water is represented by a rigid three-point model, and uncertainties in the non-bonded force-field parameters, namely the Lennard-Jones parameters and partial charges, are propagated through the simulations.
To assess the capabilities of the proposed framework, we conduct a large-scale study involving six computational models, three parameter calibrations, cubic and tetragonal simulation box geometries, and two sets of perturbed force-field parameters. Our results demonstrate that computational speed-ups of up to one order of magnitude can be achieved when successive models exhibit only small differences in their correlations with the high-fidelity model. Furthermore, nearly all combinations of high- and low-fidelity models outperform simulations relying exclusively on a single high-fidelity model, highlighting the robustness and efficiency of the multifidelity approach.

References

[1] B. Dünweg and K. Kremer, J. Chem. Phys., 99, 6983–6997 (1993).
[2] I.-C. Yeh and G. Hummer, J. Phys. Chem. B, 108, 15873–15879 (2004).
[3] J. Busch and D. Paschek, J. Phys. Chem. B, 127, 7983–7987 (2023).
[4] T. Frömbgen, A. Kuhn, J. Dölz, and B. Kirchner, J. Chem. Phys., 164, 024504 (2026).
[5] B. Peherstorfer, K. Willcox, and M. Gunzburger, SIAM Rev., 60, 550–59 (2018).

C6

Beyond Verlet: Unlocking a 2x Speedup in Molecular Dynamics with Novel Time Integrators

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Molecular dynamics (MD) simulations constitute a central pillar of computational chemistry. While the methods for force computation have advanced significantly in recent decades, the underlying time integration algorithm is still mostly the same – namely, the Verlet integrator in its various (equivalent) formulations. Previous attempts to improve integration, such as those by Yoshida or Suzuki, have not translated into better efficiency for practical MD simulations. Here, we present a family of novel integration algorithms which we have optimized to substantially enhance the performance of MD simulations. Our algorithms are explicit 8-stage Runge–Kutta–Nyström methods that are both symmetric (i.e. time-reversible) and symplectic, ensuring excellent long-term stability and energy conservation. The performance gains are significant: Compared to a MD simulation with a Verlet time step of dt=0.5 fs, our methods deliver either a 2.5-fold increase in computational speed at the same accuracy, or a 150-fold increase in accuracy (measured in terms of energy conservation) at the same computational cost. Even compared to a Verlet time step of dt=1.0 fs, our approach still achieves a two-fold speedup at the same accuracy, or a six-fold increase in accuracy using the same resources. As a time integration algorithm, our approach is independent of the underlying computational model, and thus compatible with all flavors of MD, including ab initio MD (AIMD), force field MD, and machine learned interatomic potentials (MLIPs). Geometric constraints and modern thermostats (such as NHC) can easily be supported, albeit not yet implemented. The speedup directly translates to savings in computational time and energy consumption, making a significant contribution to a more environmentally friendly computational chemistry. Given that MD simulations are estimated to consume around 2 terawatt-hours per year, our methods could potentially save half of this energy. An initial implementation will soon be publicly available in the open-source simulation package LAMMPS.

C7

Dipole-Renormalized Reservoirs for Adaptive Open-QM/MM Simulations of Liquid Water

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Open-boundary QM/MM simulations allow a quantum subsystem to exchange molecules and energy with a surrounding classical reservoir. This provides a natural framework for studying condensed-phase systems, but it also raises a central question: how physically compatible must the classical reservoir be with the quantum region, especially when the QM subsystem is small and interface effects become important?
Here, we address this question for liquid water by constructing a dipole-renormalized molecular reservoir for adaptive open-QM/MM simulations. Starting from a flexible Amber-compatible three-site water model, we optimize the reservoir against a full ab initio liquid-water trajectory. The key target is the molecular dipole distribution extracted from maximally localized Wannier centers, which transfers low-order quantum electronic information into the classical model. The optimization adjusts atomic charges, bonded parameters, and Lennard-Jones interactions while preserving the simple three-site Amber functional form.
The optimized reservoir is tested in CP2K open-QM/MM simulations for different QM-region sizes. For a large QM region containing approximately 440 water molecules, the energy-ratio diagnostic confirms that the boundary contribution is small and the system satisfies the Grand Canonical condition. In this regime, the dipole-renormalized reservoir reproduces the particle-number distribution, structural radial distribution functions, Wannier-resolved RDFs, and molecular dipole distribution of the corresponding ab initio subsystem. For a smaller QM region containing approximately 190 water molecules, where the surface-to-volume ratio is less favorable and the interface becomes more influential, the optimized reservoir still maintains substantially improved molecular consistency compared with a non-renormalized flexible TIP3P reservoir. An additional extreme test with approximately 50 QM water molecules defines the qualitative compatibility limit of the present dipole-matched three-site model.

C8

Description Is Not Understanding: From Molecular Trajectories to Atomistic Mechanisms

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Recent advances in machine-learned interatomic potentials (MLIPs) have fundamentally changed atomistic simulation. Molecular dynamics (MD) trajectories with near ab initio accuracy can now be generated on time scales that allow statistically converged sampling of rare events and direct determination of kinetic observables. As a consequence, the computational bottleneck has shifted from generating trajectories to understanding them. Understanding, however, requires more than describing representative trajectories or visualizing rare events. Atomistic mechanisms cannot be observed directly - they are scientific hypotheses that must be inferred from molecular dynamics and tested quantitatively. The central question therefore becomes: When does a proposed atomistic mechanism correctly describe the underlying dynamics? In this talk I will argue that atomistic mechanisms should be formulated as physically motivated coarse-grainings of molecular dynamics. Instead of identifying abstract metastable states solely through statistical learning, I will construct state spaces from chemically meaningful descriptors, such as lattice-site occupancies, local coordination environments, or defect configurations. Transition probabilities sampled from long MD trajectories then define Markov state models whose degree of Markovianity provides an objective measure of the validity of the proposed mechanism. Within this framework, the Chapman–Kolmogorov test becomes a quantitative criterion for testing mechanistic hypotheses rather than merely validating kinetic models. Using lithium diffusion in lithium silicides and sulfur-vacancy diffusion in monolayer MoS_2 as examples, I will demonstrate that different transport processes become Markovian only for different physically motivated state spaces. These state spaces reveal the elementary transport mechanisms and allow competing mechanistic hypotheses to be compared quantitatively.[1,2] Finally, I will discuss the construction of accurate MLIPs. I will demonstrate that fine-tuning pretrained universal foundation models provides a robust and efficient strategy for obtaining ab initio-quality force fields largely independent of the underlying model architecture.[3] Moreover, I will show that a single fine-tuned model can accurately describe an entire class of compounds, opening the door to transferable, high-fidelity simulations across chemically related materials.

[1]Qaisrani . . . , Dreßler, JCTC, 2026 22 (11), 5373-5387 https://doi.org/10.1021/acs.jctc.5c02035
[2]Flötotto, . . . , Dreßler, . Small 2026 22(20), e10679 https://doi.org/10.1002/smll.202510679
[3]Hänseroth, . . . ,Dreßler. JCPL, 2026. https://doi.org/10.1021/acs.jpclett.5c03801.

C9

Excited-state methods based on state-averaged long-range CASSCF short-range DFT

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In the present work we propose two distinct state-averaging (SA)-based methodologies for the calculation of excited states, in conjunction with the long-range complete active space self-consistent field (CASSCF) short-range density functional theory (DFT) approach (CAS-srDFT).1 The state-specific density ansatz, termed SA-CAS-srDFT, initially determines the variational parameters of an approximate srDFT functional that operates with state-averaged densities. Subsequent to convergence, the CAS-srDFT energies of each state are computed from the state-specific one- and two-body densities. The second approach is termed configuration interaction (CI)-srDFT, for which a first-order correction is added to the approximate SA density CAS-srDFT functionals. Unlike the state-specific density approach SA-CAS-srDFT, diagonalisation of the first-order corrected effective Hamiltonian CI matrix yields orthonormal CI solutions for every state. In both approaches, the total one-body and on-top pair density (OTPD) was employed for the final energy evaluation. It was observed that the CI-srDFT approach gives physically correct potential curves for ethylene, in contrast to SA-CAS-srDFT. Moreover, the CI-srDFT approach demonstrates a reduced dependence of excitation energies on the number of states in the average when compared to the SA-CAS-srDFT method. The accuracy of the various CAS-srDFT methods was investigated for 139 singlet excitation energies of 28 typical organic chromophores. The two excited-state approaches in conjunction with multiconfiguration pair-density functional theory (MC-PDFT) were also employed in the benchmark study for comparing the accuracy with CAS-srDFT. It was found that CI-srDFT methods are more accurate than their SA counterparts, giving a mean absolute error of just 0.17 eV when using the sr-ctPBE functional. The accuracy of the new SA-based CAS-srDFT methods was observed to be impressive for organic molecules; however, this was not found to be transferable when investigating excited states of transition-metal complexes. In fact, none of the CASSCF-DFT excited-state methods introduced in this study, and also MC-PDFT, were found to provide a consistent improvement of CASSCF excitation energies.

1. B. Helmich-Paris, E. R. Kjellgren, H. J. Aa. Jensen, Phys. Chem. Chem. Phys. 2025, 27, 15331.

C10

Reaction Discovery in Periodic Systems

J. Meisner[1], D. Deißenbeck[1], S. Ralf[1], W. A. Kopp, [1], P. Meier[1]

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Recent developments of efficient computational methods make it possible to discover chemical reaction networks starting from basic principles and elucidate novel reaction mechanisms that might otherwise have been overlooked. In this talk, the improvements of ab initio nanoreactor molecular dynamics (NMD) [1] are demonstrated. [2,3] The extension of NMD to periodic systems also allows the investigation of surface reactions and porous media. Among others, an application of periodic NMD simulations to construct a complex reaction network for zeolite catalysis is given, which elucidates the reaction mechanism of the unwanted side reactions in the Selective Catalytic Reduction of N2O formation.[4]

[1]L.-P. Wang et al., Nat. Chem. 2014 6, 1044-1048.
[2]J. A. Meissner, J. Meisner, J. Chem. Theory Comput. 2025 1, 218-229.
[3]P. Kuboth et al., J. Chem. Inf. Model. 2025 65, 7, 3127–3136.
[4]D. Deißenbeck et al., Angewandte Chemie 2026, 65(6), p.e14074.

C11

Quantum chemical calculations on surface adsorption

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For an accurate description of surface reactions, one needs (a) a reliable structure of the surface under reaction conditions, (b) good geometries including the effects of long-range interactions of adsorbed species with the surface, (c) precise adsorption energies, and (d) vibrational frequencies which enable a comparison with experiment. To tackle this, we start from surface structures obtained in VASP calculations, and automatically cut embedded cluster models of different size and shape. Energies are calculated by DFT to investigate cluster size effects and for small clusters by MP2 and CCSD(T) to obtain higher accuracy. In many cases, we are interested in characteristic vibrational frequencies of the adsorbed molecules. Often, we scale DFT calculated harmonic frequencies with a scaling factor obtained for the gas-phase molecule. For benchmark systems, we determine the frequencies directly from ab initio calculations at CCSD(T) level including anharmonic corrections. Results are shown for CO adsorbed on α-Al2O3(0001) [1] and on different low-index CeO2 surfaces [2,3]. Influences of surface termination, coverage, and accuracy of the quantum chemical calculations will be discussed.

[1] S. Gojare, S. Chen, J. Chen, Z. Yu, J. Vázquez Quesada, P. N. Pleßow, K. Fink, Y. Wang; Adsorption of CO on α-Al2O3(0001): A Combined Experimental and Computational Study. ChemPhysChem, 2025, 26, e202401134. https://doi.org/10.1002/cphc.202401134
[2] J. Vázquez Quesada, S. Bernart, F. Studt, Y. Wang, K. Fink; CO adsorption on CeO2(111): A CCSD(T) benchmark study using an embedded-cluster model. J. Chem. Phys. 2024, 161, 224707. https://doi.org/10.1063/5.0231189
[3] J. Vázquez Quesada; The weakly bound CO molecule adsorbed on the low-index CeO2 surfaces: A case for a CCSD(T) benchmark study using an embedded-cluster model. J. Chem. Phys. 2026, 164, 024706. https://doi.org/10.1063/5.0292423

C12

Water splitting at two-dimensional carbon-based materials

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Two-dimensional materials often exhibit unique electronic properties that are promising for applications in electronic devices, and they are well-suited for catalysis due to their high surface areas. In particular, using sunlight or other renewable sources to photo- or electrocatalyze the water splitting reaction at these materials may provide a significant contribution to a sustainable energy infrastructure.
Here, we present density functional theory (DFT) investigations on two example materials that have been used to catalyze the hydrogen evolution reaction (HER). The first group are MXenes, specifically Ti3C2 and V2C [1]. They show some HER activity on their own, but can also be used as support materials for rhodium atoms, which leads to HER rates that rival those of platinum catalysts. Our calculations suggest that the Rh atoms are partially oxidized, and that the degree of hydroxylation strongly affects the hydrogen adsorption energy and, consequently, the HER activity. Adsorption switches from strongly exothermic to endothermic with increasing hydroxyl coverage, providing an intermediate region in which the hydrogen binding hits the sweet spot for efficient catalysis.
The second example are covalent organic frameworks (COFs), which consist of molecular building blocks which are connected in two-dimensional networks. Varying the building blocks and their connectivity, for example by considering different constitutional isomers of imine-linked COFs, leads to a large variation in HER activity [2]. Our calculations revealed that hydrogen adsorption energies of the two isomers are rather similar, but solvent effects and differences in the charge distribution upon hydrogen adsorption may play a large role for the observed activity differences. [3]

References:
[1] A. S. Zverev, C. Penschke, L. Cancellara, S. Reinicke, C. Günter, S. Rüstig, N. Deepak, S. Kogikoski Jr, P. Saalfrank, and I. Bald, Small 22, e10349 (2026).
[2] J. Yang, S. Ghosh, J. Roeser, A. Acharjya, C. Penschke, Y. Tsutsui, J. Rabeah, T. Wang, S. Y. Djoko Tameu, M.-Y. Ye, J. Grüneberg, S. Li, C. Li, R. Schomäcker, R. Van De Krol, S. Seki, P. Saalfrank, and A. Thomas, Nat. Commun. 13, 6317 (2022).
[3] F. Gottwald, C. Penschke, and P. Saalfrank, Phys. Chem. Chem. Phys. 26, 21821–21831 (2024).

C13

Revisiting the optical band gaps of Co_3O_4 using molecular methods

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In recent years, the antiferromagnetic material Co_3O_4 has been extensively studied for its experimentally observed optical band gaps [1–3]. Previous computational studies [4,5] of these properties have mainly relied on periodic approaches with DFT or Green function techniques. In this work, we present an additional novel perspective on the optical band gaps of Co_3O_4 by using molecular methods together with an electrostatic embedding approach [6,7].
Our protocol combines TD-DFT and multireference methods, such as SA-CASSCF/NEVPT2 [8] and MR-EOM-CC [9], yielding results in close agreement with experimental observations. Based on these results, a detailed analysis of the band gaps' origin is performed, linking the excited-state properties to the antiferromagnetic ground state of the material. Particular emphasis is placed on exploring the influence of both "ionic" and "neutral" antiferromagnetic states [10] inherent in the material, which can be specifically addressed using multireference methods. Our results highlight the significant impact of these states on the optical properties and their role in influencing the observed band gaps. This study contributes to a deeper understanding of the optical behavior of Co_3O_4 and emphasizes the importance of explicitly considering the antiferromagnetic ground states in comprehensive analyses of such materials.

References

[1] A. Cherkashin, F. Vilesov, *Sov. Phys. Solid State* **11**, 1068 (1969).
[2] K. J. Kim, Y. R. Park, *Solid State Commun.* **127**, 25–28 (2003).
[3] C.-M. Jiang et al., *J. Phys. Chem. C* **118**, 22774–22784 (2014).
[4] V. Singh et al., *J. Chem. Theory Comput.* **11**, 64–72 (2015).
[5] J. Chen, X. Wu, A. Selloni, *Phys. Rev. B* **83**, 245204 (2011).
[6] A. Dittmer, R. Izsák, F. Neese, D. Manganas, *Inorg. Chem.* **58**, 9303–9315 (2019).
[7] A. Dittmer et al., *Phys. Chem. Chem. Phys.* **27**, 17225–17244 (2025).
[8] C. Angeli et al., *J. Chem. Phys.* **114**, 10252–10264 (2001).
[9] M. Nooijen et al., *J. Chem. Phys.* **140**, 081102 (2014).
[10] R. H. Summerville, R. Hoffmann, *J. Am. Chem. Soc.* **98**, 7240–7254 (1976).

C14

Engineering Hybrid Two-Dimensional TMDC/Perovskite Heterostructures from First Principles

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The integration of transition-metal dichalcogenide (TMDC) monolayers with thin perovskites nanosheets offers a versatile platform for engineering the electronic and excitonic properties of van der Waals heterostructures. First-principles simulations play a key role in understanding the microscopic mechanisms governing interfacial interactions and in guiding the design of new hybrid materials.
In this contribution, we present a series of density functional theory (DFT) studies of TMDC-based heterostructures performed in close collaboration with experimental groups. Structural optimization and electronic structure calculations reveal how the atomic arrangement, dielectric environment, and charge-compensating species determine the band alignment and interlayer coupling. For $WS_2/(PEA)_2PbI_4$, the calculations predict an unconventional band alignment where the organic spacer suppresses electron transfer while enabling efficient hole transfer through a valence-band cascade. Extending this approach to $MoSe_2$/2D-perovskite heterostructures demonstrates how the relative alignment of electronic and excitonic states controls the competition between charge transfer and energy transfer, providing design principles for tailoring excitation pathways. Simulations of $WSe_2/(BA)_2PbI_4$ further identify hybridized interfacial electronic states responsible for tunable spin-dependent interlayer exciton emission observed experimentally. Finally, recent first-principles investigations of the emerging $WSe_2/[Ca_2Nb_3O_{10}]^-$ heterostructure reveal weak interfacial coupling but a strong sensitivity of the electronic structure to the charge-compensating environment of the oxide nanosheet, highlighting the importance of defects and local chemistry in modifying excitonic properties.
Together, these studies demonstrate how atomistic simulations provide predictive insight into the structure–property relationships of hybrid TMDC heterostructures and establish computational guidelines for designing next-generation optoelectronic, spintronic, and quantum photonic materials.

C15

Photodynamics of multi-mode multi-state molecular systems coupled to a dissipative environment

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Ultrafast processes in condensed phase photoexcited molecular systems involve the transition from a coherent dynamical regime – where a precise phase relation exists between different wave packet components – to incoherent "classical-like" dynamics. The decoherence process is driven the dissipation due to the surrounding molecular environment. From the computational viewpoint, modelling the dynamics of nonadiabatic vibronic quantum systems interacting with fluctuating environments becomes especially challenging when the system' s high dimensionality precludes the calculation of its eigenstates. To overcome this limitation, a novel eigenstate-free formalism is introduced. This approach represents the open quantum system as a mixture of high-dimensional, time-dependent wave packets, governed by coupled Schrödinger equations [1], while the environment is modeled using a multi-component quantum master equation. A computationally efficient implementation of this formalism employs the variational Gaussian/multiconfigurational time-dependent Hartree (G-MCTDH) ansatz for the wave packets and propagates the environment dynamics using hierarchical equations truncated at the first or second level [2]. The methodology is validated through simulations of the dynamics in vibrationally excited adsorbates [2], multichromophoric aggregates, and symmetry-breaking donor-acceptor systems [3,4], explicitly incorporating multiple vibrational modes and accounting for the effects of the residual bath in full dimensionality. These results demonstrate the potential of the approach as a powerful quantum dynamical method for modeling complex system-bath interactions involving numerous degrees of freedom across multiple time scales.

[1] D. Picconi, I. Burghardt, J. Chem. Phys., 2019, 150, 224109. https://doi.org/10.1063/1.5099983
[2] D. Picconi, J. Chem. Phys., 2024, 161, 164108. https://doi.org/10.1063/5.0233708
[3] A. Aster, B. Bornhof, N. Sakai, S. Matile, E. Vauthey, J. Phys. Chem. Lett., 2021, 12, 1052. https://doi.org/10.1021/acs.jpclett.0c03641
[4] D. Picconi, J. Chem. Phys., 2022, 156, 184105. https://doi.org/10.1063/5.0089887

C16

A simple way to deal with decoherence in trajectory surface hopping

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Tully's fewest switches surface hopping is a leading computational method for excited-state dynamics in molecules and materials. It unfortunately lacks a proper description of decoherence, which has lead to numerous "decoherence correction" schemes. However, the by far most commonly used "energy-based correction" is not guaranteed to improve accuracy - in fact, it often even harms more than it helps. The problem is that it relies on a parameter that assumes a given energy scale, and therefore only works for systems that are similar to those the method was originally tested on.
As a simple way to avoid this problem, I suggest to restrict decoherence to regions of low nonadiabaticity measured by the Massey parameter [1]. The advantage of this criterion is that the threshold is dimensionless, so the same value is suitable for a variety of systems with vastly different size and absolute energy scales, which I will demonstrate with a suite of benchmark systems including conical intersection system, condensed-phase models, photosynthetic complexes, and molecules with spin-orbit coupling. I will also show how the method helps describe charge conduction in periodic molecular crystals [2]. The method adds minimal computational effort and the results are relevant for any surface-hopping simulations of excited-state dynamics in complex systems.

[1] J.E. Runeson, J. Chem. Phys. 163, 154105 (2025).
[2] J.E. Runeson, T.J.G. Drayton, and D.E. Manolopoulos, J. Chem. Phys. 161, 144102 (2024).

C17

Modelling nonadiabatic dynamics of excitonic and plasmonic photoswitchable systems

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Nonadiabatic dynamics determine the fate of photoexcitation in complex excitonic and plasmonic systems. While the former feature the presence of multiple chromophores in close proximity to each other, the latter exhibit coupling of molecules and metal nanoparticles. Understanding photoinduced dynamics of these systems requires nonadiabatic treatment involving multiple excited states and numerous nuclear degrees of freedom, thus posing a challenge for modelling. In this contribution, we report on surface hopping simulations using semiempirical quantum chemical methods for excitonic and plasmonic systems featuring azobenzene—a prototypical molecular photoswitch.

First, we consider multichromophoric azobenzene systems. We model their photoinduced, nonadiabatic dynamics using an on-the-fly surface hopping approach combined with the semiempirical rAM1/FOMO-CI method [reparameterized Austin Model 1 / floating occupation molecular orbital – configuration interaction]. A transition density matrix analysis is used to characterize exciton (de)localization and evolution. In particular, we focus on exciton localization timescales, excited state lifetimes, and isomerization quantum yields in the aggregated state [1,2]. We also study how the inclusion of double excitations in addition to singles into the semiempirical CI surface hopping simulations affect exciton dynamics of multiazobenzenes [3].

Second, we report on nonadiabatic surface hopping TD-DFTB [time-dependent density functional tight binding] dynamics of a plasmonic gold nanocluster–azobenzene system exhibiting hundreds of electronically excited states [4]. Specifically, we study excited-state relaxation after molecular and plasmonic excitations, providing direct insight into elementary steps relevant to plasmonic chemistry.

1. Titov, E. J. Phys. Chem. C 2023, 127, 13678.
2. Titov, E. ACS Omega 2024, 9, 8520.
3. Titov, E. J. Phys. Chem. Lett. 2024, 15, 7482.
4. Jahn, N; Titov, E. in preparation 2026.

C18

Quantifying photoinduced electron transfer in RNA and DNA

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Photoinduced electron transfer (PET) is one of the most fundamental phenomena responsible for the conversion of light into chemical reactions. It is also one the most common photophysical processes occurring in UV-excited nucleic acids. It leads to the population of long-lived excited charge transfer (CT) states, characterized by quantum yields strongly dependent on sequence. However, the exact origins of this dependency remained largely obscure until recently. In this talk, I will demonstrate how quantum chemical calculations can support the discovery of new types of photoredox reaction types and prebiotically plausible pathways to RNA and DNA nucleosides under the conditions of early Earth [1,2]. I will also discuss how nucleobase sequence, stacking overlap and weak interactions modulate electronic couplings and energies of key CT states [3, 4]. These discoveries of productive photoredox reactions and nonenzymatic self-repair pathways in RNA and DNA allowed us to propose an alternative DNA nucleobase, which can substantially enhance the photostability of DNA oligomers [5, 6]. Finally, I will show our recent developments of nonadiabatic transition state theory which enable accurate prediction of photoinduced electron transfer timescales in organic chromophores [7].

1. Xu J., Chmela V., Green N.J., Russel D.A., Janicki M.J., Góra R.W., Szabla R., Bond A.D., Sutherland J.D., Nature, 582 (2020) 60.
2. Janicki M., Szabla R., Angew. Chem. Int. Ed., 64 (2025) e202413498.
3. Kufner C.L., Crucilla S., Ding D., Stadlbauer P., Šponer J., Szostak , Sasselov D.D. and Szabla R., Chem. Sci., 15 (2024) 2158 .
4. Szkaradek K.E., Stadlbauer P., Sponer J., Góra, R. Szabla, Chem. Commun., 56 (2020) 201.
5. Szabla R., Zdrowowicz M., Spisz P., Green N.J., Stadlbauer P., Kruse H., Šponer J. and Rak J., Nat. Commun., 12 (2021) 3018.
6. Szabla R., Kruse H., Stadlbauer P., Šponer, Sobolewski A.L., Chem. Sci., 9 (2018) 3131.
7. Szabla, R.*, Barboza, C.A., J. Phys. Chem. Lett., 17 (2026) 2057–2064.

C19

Topological Photophysics in Molecular Aggregates: The Role of Charge-Transfer States

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Topological band theory links global properties of periodic wave functions to robust boundary and interface phenomena. Transferring this concept to molecular excitonic materials is appealing because molecular packing controls excitonic couplings and could therefore provide a chemical route to localizing and directing optical excitations. Real molecular aggregates, however, are not simple Frenkel-exciton lattices: nearby charge-transfer (CT) configurations can modify the excitonic band structure and generate localized states that resemble topological edge modes. We investigate this interplay in perylene-bisimide aggregates using symbolic configuration interaction (SymbolicCI), a fragment-based approach that constructs ab initio aggregate Hamiltonians in a chemically interpretable basis of Frenkel and CT configurations [1,2]. These atomistic Hamiltonians are used to test periodic hybrid Frenkel–CT lattice models. Exact Feshbach elimination shows that the reduced Frenkel problem is generally energy dependent rather than a static Su–Schrieffer–Heeger Hamiltonian. Candidate topological phases are therefore evaluated using isolated gaps and inversion indicators of the explicit multiband spectrum, adiabatic continuity to the controlled SSH limit, and separate finite-chain boundary diagnostics. We find that a broad range of apparent SSH inversions predicted by static downfolding collapses to a narrower set of inversion-nontrivial multiband gaps when the CT sector is restored. Charge transfer plays a dual role: differential CT dressing can reverse the effective dimerization in the idealized model, but CT-rich boundary states can also imitate topological localization. In the microscopic PBI candidate, destructive interference suppresses the nearest-neighbour splitting to the resolution limit of the mapping, where longer-range couplings invalidate a two-bond SSH description. The resulting states differ in excitonic character, localization, oscillator strength, and expected field response. These results define the conditions under which SSH-like topology can be meaningfully assigned in molecular excitonic materials.

[1] A. Singh and M. I. S. Röhr, J. Chem. Theory Comput. 20, 8624–8633 (2024).
[2] J. E. Adelsperger, C. de Graaf, and M. I. S. Röhr, J. Chem. Theory Comput. 22, 4201–4217 (2026).

C20

Orbital-free DFT is becoming practical

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In 1964, Hohenberg and Kohn proved that the ground state of an interacting electron system is completely determined by its electron density. In principle, this result promises a dramatic simplification of electronic structure theory: instead of solving for a many-electron wave function in a 3N-dimensional space, one could obtain the ground state by minimizing an energy functional of the three-dimensional electron density. In practice, this vision has remained unrealized, because the key ingredient—the kinetic-energy functional of the density—is unknown for real molecular systems. Modern Kohn-Sham density functional theory circumvents this obstacle by introducing auxiliary orbitals, but at the price of cubic scaling with system size.

In this talk I will show that the missing functional can be learned to sufficient accuracy for increasingly complex chemistry using rotation equivariant machine learning models. After reaching chemical accuracy relative to KS-GGA functionals on relaxed geometries, I will give an update on speedups and show latest results on non-equilibrium geometries, open shell systems and for much of the periodic table.

These results suggest that machine learning may finally enable a practical realization of the Hohenberg–Kohn program, opening an orbital-free density functional theory route to chemically accurate electronic-structure calculations for large systems, complementing both linear-scaling density functional theory and machine-learned interatomic potentials.

C21

Self-Consistent Fine-Tuning of Machine Learning Interatomic Potentials by Targeted Exploration of Configuration Space

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Universal machine learning interatomic potentials (uMLIPs) have shown an astonishing ability of describing almost arbitrary chemical systems reasonably. They are, however, not accurate enough for the calculation of quantities like reaction rate constants that require a very precise representation of the potential energy surface. Fortunately, uMLIPs can be fine-tuned for a system of interest, by training them on system-specific ab initio reference data. The generation of such reference data is nontrivial since it requires a broad sampling of the relevant configuration space, often out of reach for direct ab initio molecular dynamics (AIMD) [1]. Self-consistent fine-tuning is a solution for this: long MD samplings are done with the uMLIP, a relevant subset of sampled structures is recalculated with the ab initio method, and this is iterated with the successively fine-tuned MLIP to cure the slightly incorrect configurational sampling of the uMLIP [2].
In this contribution, I show how self-consistent fine-tuning of uMLIPs can be combined with umbrella samplings along complex reaction coordinates with the Caracal program package [3]. This enables the black-box setup of highly accurate targeted MLIPs for different chemical reaction mechanisms in arbitrary environments. I present the application of this method on a benchmark study of small proton exchange reactions in the gas phase [4], and then continue with real-world applications - based on actual ab initio data - for larger gas phase systems and hydrogen diffusions on different metal surfaces. In all examples, ring polymer molecular dynamics (RPMD) is used to consider nuclear quantum effects. Finally, an outlook is given on how to generalize the method to arbitrary chemical systems and reaction mechanisms.

[1] J. Steffen, A. Alibakshi, J. Chem. Phys. 161, 184116 (2024).
[2] J. Steffen, J. Phys. Chem. C, 129, 13513 (2025).
[3] J. Steffen, J. Chem. Theory Comput., 19, 5334 (2023).
[4] J. Steffen, submitted (DOI: 10.26434/chemrxiv.15002012/v1).

C22

Implementing Reduced Density Matrix Functional Theory within a Kohn–Sham like Framework

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Density functional theory (DFT) has been one of the central pillars of electronic structure theory for decades. Its remarkable balance between accuracy and computational efficiency has made it the method of choice across chemistry, materials science, and condensed matter physics. Despite its tremendous success, however, DFT faces well-known limitations, particularly in the description of strongly correlated systems, where static and dynamic correlation cannot be treated on equal footing.
Reduced density matrix functional theory (RDMFT) offers a promising alternative by replacing the electronic density with the one-body reduced density matrix as the fundamental variable. While this richer descriptor naturally incorporates key aspects of electronic correlation, the theoretical framework of RDMFT has historically lacked some of the foundational structures that have made Kohn–Sham DFT so successful.
I will present a reformulation of RDMFT built from first principles, introducing a fictitious partially interacting reference system analogous to the Kohn–Sham construction. The resulting effective Hamiltonian provides a rigorous foundation for developing the theory and, crucially, enables the construction of an adiabatic connection within the RDMFT framework. We explore the properties of this adiabatic connection and demonstrate how it can be used to analyze correlation effects and guide functional development.
By establishing an adiabatic connection for RDMFT, we recover one of the most powerful conceptual tools of modern electronic structure theory in a density-matrix setting. This creates a systematic pathway for constructing and benchmarking new functionals and brings RDMFT significantly closer to becoming a practical and broadly applicable successor to conventional density functional theory.

C23

Beyond Potential Energy Surfaces: A Quantum Geometric View of Molecular Electronic Structure

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Quantum geometry has become a central language in modern condensed matter physics. Its central quantity, the quantum geometric tensor, characterizes how the wavefunction varies with changes in external parameters, thereby providing a unified description of the coupling between a quantum system and its environment. This perspective has proven instrumental in understanding phenomena ranging from anomalous Hall conductance to the Aharonov–Bohm effect. Molecular quantum chemistry, by contrast, is still formulated primarily in terms of energies, gradients, and potential energy surfaces. Yet the same quantum geometric structures arise naturally when electronic states are viewed as functions of nuclear coordinates, electromagnetic fields, or other external parameters. In this contribution, I will highlight that quantum geometry provides a complementary language for molecular electronic-structure theory, revealing that a variety of seemingly distinct molecular response properties are, in fact, manifestations of the same underlying quantum geometric tensor. Whenever observables depend on derivatives of the electronic wavefunction, geometric contributions naturally emerge. Examples include corrections beyond the Born–Oppenheimer approximation, magnetic response phenomena, and several forms of vibrational spectroscopy.

Recent methodological advances have made these quantities accessible within established quantum chemical frameworks. Linear response current-density functional theory now enables the evaluation of the full molecular quantum geometric tensor for a broad range of density functional approximations, extending its application to regimes such as strong magnetic fields. Here, the Berry curvature enters the nuclear equations of motion as a Lorentz-like force, enabling quantitative descriptions of molecular dynamics and rovibrational spectroscopy. These results demonstrate that molecular motion is governed not only by the potential energy surface, but also by the geometry of the underlying electronic wavefunction itself. Taken together, these developments establish quantum geometry as an organizing language for electronic response, nuclear motion, spectroscopy, and external perturbations. The same perspective naturally extends beyond isolated electronic states: degenerate state manifolds require non-Abelian geometric structures, opening routes towards unified, gauge-covariant descriptions of coupled electronic, spin, and nuclear dynamics. Molecular quantum chemistry may therefore benefit substantially from adopting a geometric viewpoint that has already transformed condensed matter theory.

C24

Theoretically Designed Electro- and Photoactivated Earth-Abundant Metal-Based Catalysts for Dihydrogen Generation from Water

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Dihydrogen plays a crucial role in the daunting challenge of replacing fossil fuel-based economy with renewable, clean, and low-cost energy economy. Water splitting by electro- and photo-activated transition metal catalysts has emerged as a viable solution for generation of alternative fuel, H2 gas. However, there are still significant challenges that need to overcome in order to have a commercially viable and low-cost catalyst. The crucial features an efficient and long-lasting catalytic device need to possess are a) unidirectional and kinetically fast multielectron transfer from photosensitizer to the catalytic core, b) robust ligand framework that can sustain different redox states formed in the catalytic cycle, and c) high operational rates by the catalytic core for dihydrogen generation from water. In this regard, several novel Earth-abundant Co-, Ni-, and Cu-based mono- and multimetallic electro-/photocatalysts have been designed via detailed study with Density Functional Theory (DFT). These catalysts can display features like electronic coupling between different metal centers, biomimetic proton relay pathways, and proton-coupled electron transfers to promote their catalytic activity. Moreover, a set of much-needed guiding principles have been provided that can be used by an experimental chemist as a toolbox for synthesis of novel H2 generation catalysts while minimizing the decomposition of the systems.

C25

Watching Excitons Move: Real-Time and Linear-Response TDDFT for Periodic Systems in TURBOMOLE

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[2]*Indian Institute of Science, Bengaluru, India*

The optical response of materials, from excitons in atomically thin semiconductors to laser-driven currents behind petahertz electronics, unfolds on femto- to attosecond timescales. Plane-wave codes dominate this field, yet local Gaussian bases offer a compelling alternative: all-electron accuracy, natural treatment of core excitations, seamless transition from molecules to crystals, and no artificial replication of low-dimensional systems. This talk presents the extension of TURBOMOLE's riper module into a full time-dependent spectroscopy engine for periodic systems of any dimensionality.

At its core is k-resolved real-time TDDFT: the time-dependent Kohn–Sham equations are propagated at every point of the Brillouin zone mesh using unitary Magnus and enforced-time-reversal-symmetry schemes, with the absorption spectrum extracted from the induced macroscopic current. Velocity and hybrid gauges are available, and gauge invariance is verified through the Thomas–Reiche–Kuhn sum rule. Crucially, the propagation now includes exact exchange, so global hybrid and range-separated functionals capture the excitonic effects that semilocal functionals miss, at the moderate cost enabled by density fitting and continuous fast multipole methods. Periodic linear-response TDDFT within the same framework provides an independent cross-check, and agreement between the two routes is demonstrated for chains, monolayers, and bulk crystals, culminating in the excitonic absorption of two-dimensional hexagonal boron nitride and bulk silicon.

Because real materials are rarely perfect crystals, the framework connects to embedding: real-time TDDFT embedding theory evolves a molecular region inside a frozen periodic environment, giving access to chromophores at surfaces and in explicit periodic solvent boxes at near-molecular cost, with solvatochromic shifts reproduced to within 0.1 eV of full-system references.

One code and one input thus carry a simulation from an isolated dye to a dye embedded in a crystal to the crystal itself. The talk concludes with the road ahead: strong-field dynamics and high-harmonic generation in solids with hybrid functionals, toward predictive attosecond materials science.

C26

Reshaping the research landscape for the description of excited states in solids: the tool set of the CP2K-Newton-X program package

Anna Hehn[1]

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The plethora of photochemical processes that need to be unraveled when aiming for a comprehensive description of excited states in solids becomes apparent when considering the process of upconversion: Converting two low energy photons to a high energy one is of vital importance for both catalytical as well as bio-medical applications, holding the promise to enhance both sensing, phototherapy, and theranostics as well as photocatalysis under low solar irradiance. The challenge of overcoming the systematically too low quantum yields demands an atomistic understanding of the full range of relevant photochemical mechanisms, emphasizing the need for an advanced quantum-mechanical tool set for the description of excited states within time-dependent density functional theory implying periodic boundary conditions. This presentation summarizes first steps towards setting up such a versatile tool set within the CP2K-Newton-X interface [1, 2], with recent developments enabling to go from static absorption and emission spectroscopy to non-adiabatic trajectory surface hopping beyond the classical path approximation [3,4].

[1] M. Iannuzzi et al., J. Phys. Chem. 130, 1237 (2026).
[2] J.-R. Vogt, J. Wilhelm, A. Hehn, J. Chem. Phys. 162, 082502 (2025).
[3] J.-R. Vogt, M. Schulz, R. S. Mattos, M. Barbatti, M. Persico, G. Granucci, J. Hutter, A. Hehn, J. Chem. Theory Comput. 21, 10474 (2025).
[4] M. Barbatti et al., Phys. Chem. Chem. Phys. 28, 15751 (2026).

C27

Spin–orbit Coupling of TDDFT Amplitudes: Left or Right?

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The need for a fast and easy procedure to include spin–orbit coupling (SOC) effects in calculations of transition metal compounds led Wang and Ziegler to devise an approximate relativistic TDDFT formalism. [1] It employs scalar relativistic auxiliary many-electron wave functions (AMEWs), constructed from the eigenvectors in Casida's formulation of TDDFT, to calculate SOC matrix elements (SOCMEs). Similar approaches to compute interstate SOCMEs from AMEWs were subsequently developed by several groups including our own. [2] One problem that arises in the framework of full linear response TDDFT and that has been addressed by a few groups only is the appropriate choice of amplitudes employed for constructing the AMEWs: When hybrid functionals are used, the eigenvalue equation for determining the amplitudes is non-Hermitian and hence left and right eigenvectors are not equal. Matrix elements of real symmetric operators (such as the electric dipole operator μ) connecting an excited state with the singlet electronic ground state are formed with the right eigenvector whereas the left eigenvector is employed for matrix elements of antisymmetric imaginary operators (such as the angular momentum operator L). It therefore seems appropriate to use the left eigenvector of a triplet state for computing SOCMEs with the singlet ground state. [3] As discussed in this contribution, this choice entails complications, however, when phosphorescence rates are to be determined in the framework of quasi-degenerate perturbation theory. Moreover, there is no clear prescription for SOCMEs between excited-state AMEWs, which are needed for both, intersystem crossing and phosphorescence rates. Extensive benchmarking against reference data from multi-reference configuration interaction wavefunctions show that electric transition dipole moments are much more sensitive with respect to the choice of AMEWs than SOCMEs. [4] The Spoiler program developed in our laboratory therefore employs renormalized right TDDFT amplitudes in conjunction with quasi-degenerate perturbation theory, but offers users the freedom of choosing the amplitudes otherwise.

References

1. F.Wang, T. Ziegler, J.Chem.Phys. 2005, 123, 154102.
2. F. Franco de Cavalho etal., 2014; X. Gao etal., 2017; F. Dinkelbach etal. 2017; B. de Souza etal., 2019.
3. P.Himmelsbach, C. Holzer, J.Chem.Phys. 2024,161, 244105.
4 .F.Meitza, M. Kleinschmidt, C.M. Marian, in preparation 2026.

C28

Super-Resolution for Real-Time Electronic Structure Theory in Complex Systems

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Calculating excited state spectra of large, complex systems is often prohibitively expensive with standard frequency-domain methods such as the Casida equations, the Bethe-Salpeter Equation, or Equation-of-Motion Coupled Cluster. Real-time methods provide an alternative, as all modes are excited simultaneously. However, long simulation times are required to resolve narrow spectral features with traditional Fourier signal analysis, significantly limiting system size, and any information about the molecular orbitals involved in each transition is lost. Super-resolution methods such as Compressed Sensing promise high-resolution spectra from much shorter signals but assume the spectrum to be sparse, an assumption which breaks down in larger systems where sharp features are embedded in a quasi-continuum of smaller nearby peaks. To overcome this, we combine recent, highly noise-tolerant super-resolution techniques with a physically motivated separation of transition dipoles into bright and quasi-continuum contributions in an integrated, self-validating workflow. Our approach extends prior reconstruction schemes [1, 2, 3] with substantially improved robustness, while also recovering transition-level molecular orbital insight, promising to become an alternative to frequency-domain methods, particularly for large systems or hybrid functionals. We demonstrate it on systems containing several hundred heavy atoms, achieving up to 20-fold speedups while maintaining spectral accuracy even for signals dominated by large continua.

[1] M. Kick et al., Nat. Commun. 15, 8001 (2024)
[2] A. Bruner et al., JCTC 12, 3741 (2016)
[3] X. Andrade et al., PNAS 109, 13928 (2012)

C29

The Subtleties of Calculating Non-Covalent Interactions with Wavefunction Based Methods

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Non-covalent interactions are ubiquitous within the natural world, driving many biological, chemical and physical phenomena including protein folding, catalysis, gas storage, stacking of two-dimensional materials and molecular liquid properties. However, quantifying these interactions is highly complex from both a computational and experimental standpoint.

Calculations are problematic due to slowly convergent correlation energy with respect to basis set size, small magnitude of the interaction energies and the requirement for delicate error cancellation between approximations made within the component molecules and the whole. These issues were further exacerbated when two state of the art methods, coupled cluster with single, double and perturbative triple excitations (CCSD(T)) and fixed-node diffusion Monte Carlo (FN-DMC), were found to disagree in the quantification of interaction energies for many dimers.[1, 2] These issues have not only raised questions about the reliability of quantum chemistry for the calculation of non-covalent interactions, but also called into question the trustworthiness of the methods themselves.

While these state-of-the-art quantum chemical methods are exact in principle, a plethora of approximations are required to ensure computational tractability. While each approximation may, in and of itself, result in only a small introduction of error, the combination of these small errors could possibly lead to a significant discrepancy between the methodologies.

Establishing platinum standard non-covalent interaction benchmarks is imperative for understanding our calculations and their limitations. In this work, rigorous and systematic evaluation of the various approximations invoked in coupled cluster calculations is carried out. The effect of these approximations are quantified, critically assessed and compared to FN-DMC results.

[1] Al-Hamdani et al., Nat. Commun., 12, 3927 (2021).
[2] Shi et al., J. Chem. Phys., 162, 144107 (2025).

Peyerimhoff Award Abstracts

A1

Development of Low-Order Scaling Multicomponent Methods

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Nuclear quantum effects (NQEs) are essential for accurately describing many chemical systems, particularly when light nuclei such as protons are involved. [1] However, their explicit quantum-mechanical treatment remains computationally demanding, especially for large and complex molecular systems.
To address this challenge, efficient low-scaling multicomponent quantum-chemical methods were developed within the Nuclear-Electronic Orbital (NEO) framework. [2] The developed methodology spans wave-function-based, density-based, and hybrid approaches, with a focus on improving their computational efficiency, scalability, and applicability to complex molecular systems. By combining integral-direct algorithms with density fitting and local approximations, the computational scaling and memory requirements were substantially reduced, enabling multicomponent calculations for systems with multiple quantum nuclei at previously inaccessible sizes. [3]
The resulting methods were applied to a broad range of systems, including protonated water clusters, DNA strands, hydrogen-bonded complexes, and molecular systems investigated experimentally in helium nanodroplets. They enable the investigation of proton affinities, structural changes induced by NQEs, vibrational hydrogen-bond shifts, and proton dynamics, and demonstrate cases where an explicit multicomponent treatment is essential to reproduce experimental observations. Together, these developments significantly expand the scope and applicability of multicomponent quantum chemistry and provide a foundation for the routine simulation of NQEs in complex molecular systems.

[1] Markland, T., Ceriotti, M. Nuclear quantum effects enter the mainstream. Nat Rev Chem 2, 0109 (2018) DOI: 10.1038/s41570-017-0109.
[2] Pavošević, F., Culpitt, T., Hammes-Schiffer, S. Multicomponent Quantum Chemistry: Integrating Electronic and Nuclear Quantum Effects via the Nuclear-Electronic Orbital Method. Chem Rev 120 (9), 4222-4253 (2020) DOI: 10.1021/acs.chemrev.9b00798.
[3] Hasecke, L. Development of Low-Order Scaling Multicomponent Methods. University of Göttingen (2026) DOI: 10.53846/goediss-11783.

A2

Quantum Information Theoretical Approach to Functional Theories

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Density functional approximations often struggle with strong static correlation, whereas systematically improvable wave function methods become increasingly costly as system size and multireference character grow. One-particle reduced density matrix functional theory (RDMFT) offers a complementary route by using the one-particle reduced density matrix (1RDM) as its basic variable. This richer description, however, raises the one-body N-representability problem: which 1RDMs arise from valid N-fermion states? Characterizing this domain is central both to the foundations of RDMFT and to the construction of reliable approximations. In this talk, we develop a unified geometric framework for functional theories in which the scope, which is specified by the basic observables and the fixed part of the Hamiltonian, determines the reduced variable, universal functional, and representability domain. For spin-1/2 fermions at fixed total spin, we use this framework to identify the orbital 1RDM with the moment map for the action of the corresponding unitary group on the many-fermion state space. The admissible ordered spectra form a moment polytope, yielding symmetry-adapted constraints for pure and ensemble one-body N-representability. We then extend this construction to ensembles of low-lying excited states. For a chosen weighting of the low-lying energies, one obtains a restricted domain of admissible orbital 1RDMs, whose ordered spectra form a convex polytope. Its facets provide weight-dependent, mixed-state generalizations of Pauli's exclusion principle. We discuss their structure and implications for excited state RDMFT. Finally, we introduce an ensemble Hartree-Fock theory for excited states as a tractable mean-field counterpart and derive the associated functional approximation. Together, these results establish symmetry, convex geometry, and N-representability as a common foundation for functional theories of ground and excited states.

Abstracts: Posters

P1

Davydov Splitting Without a Davydov Pair in Perylene Red Microcrystals with a Large Unit Cell

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Davydov splitting (DS) is a very common observable in molecular spectroscopy, and the dimer relation DS = $2V$ is often used to read the intermolecular Coulomb coupling directly off the measured splitting. We show that this relation can break down in crystals with more than two molecules per unit cell, using the Perylene Red crystal as an example. This perylene diimide crystallizes in the $Pca2_1$ space group with eight molecules per unit cell, which is quite unusual for organic crystals. We model its polarized absorption with a Frenkel–Holstein Hamiltonian. Excitation energies and transition densities are calculated by TDDFT methods with a hybrid exchange functional. Coulomb couplings are computed from Löwdin transition charges, which reproduce full TDDFT couplings to within 2% while the common dipole–dipole approximation overestimates the couplings. Lattice sums over periodic images account for intercell interactions.

The calculated spectra match the measured polarization-resolved absorption, including an apparent splitting of 520 cm^{-1} between the a- and b-polarized bands (610 cm^{-1} in experiment). A symmetry analysis of the Hamiltonian at $k = 0$ reveals the origin of this splitting. The eight monomers form two tetramer pools related by near-exact exchange symmetry. Tetramer eigenstates mix pairwise into plus/minus combinations and form four Davydov pairs. Projecting the full eigenstates onto these pair states shows that the two observed bands belong to two different pairs, i.e., the b-polarized band is the minus state of one pair and the a-polarized band the plus state of another. The apparent splitting therefore matches no Coulomb matrix element, and applying DS = $2V$ here yields a coupling with no physical meaning. The same analysis identifies the lowest bright state as J-like, which increases the spectral overlap for energy transfer. Additionally, Kinetic Monte Carlo simulations of Förster hopping based on these couplings give a density-independent diffusion coefficient of 3.3–3.5 nm^2/ps, in agreement with time-resolved measurements. Such subcluster symmetries are possible in any crystal with many molecules per unit cell, so the observed number of Davydov components alone is not sufficient to assign couplings.

P2

Generative Neural Networks to sample PIMC configurations

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To describe the quantum-mechanical nature and properties of systems with complex electronic structures, Path Integral Molecular Dynamics (PIMD) and related methods, such as Path Integral Monte Carlo (PIMC), are widely used. However, these methods suffer from high computational costs and slow convergence rates.
To address these limitations, we introduce a generative neural network, specifically a Variational Autoencoder (VAE), to improve sampling efficiency. The objective is to map high-dimensional imaginary-time configurations into a low-dimensional latent space while preserving the essential statistical information and structural patterns. This latent space captures the underlying distribution of the PIMC samples, allowing the generation of new physically plausible PIMC configurations through latent-space sampling.
To ensure translational and rotational equivariance, so-called E(n)-Equivariant Graph Convolutional Layers (EGCL) were incorporated into the VAE architecture. Both the encoder and decoder consist of combinations of EGCLs, additional neural network layers, and activation functions.
Another significant advantage of the proposed generative approach is that new configurations can be generated in parallel. Once the latent space has been learned by the encoder, multiple samples can be generated independently, allowing the efficient generation of large batches of PIMC configurations and reducing the overall computational cost, compared to conventional PIMC approaches.
Furthermore, we show that the generated samples reproduce the structural thermodynamic properties of reference PIMC simulations while reducing the associated sampling overhead [1]. These findings highlight the potential of generative approaches as efficient accelerators for quantum statistical simulations of systems with complex electronic structure.

References
[1] M. Hütter, M. Ončák, J. Chem. Theory Comput. 2025, 21, 4397.

P3

Prospective Parity-Violation NMR Experiments: A Relativistic DFT Study of Chiral Platinum Complexes

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The experimental detection of parity violation (PV) in molecules remains challenging [1]. In NMR spectroscopy, PV induces splittings between enantiomers below 1 mHz [2], far smaller than the typical resolution of over 0.5 Hz. An indirect strategy, therefore, measures diastereomeric splittings under fast exchange at different enantiomeric excesses and extrapolates them to the racemic limit, using PV-insensitive light nuclei as internal comagnetometers to reduce systematic effects [3]. This approach requires soluble, conformationally well-defined molecules with a sensitive heavy nucleus, large PV shifts, favorable signal-to-noise and linewidth characteristics, and controlled resolvable interactions with a chiral environment. To evaluate suitable candidates, relativistic quantum-chemical calculations are performed on soluble chiral platinum complexes synthesized in the laboratory of Jeanne Crassous, correlating their predicted heavy-element-enhanced PV shifts with experimentally relevant linewidths. To support the design of a PV-NMR experiment, the required molecular properties are calculated using a quasi-relativistic two-component approach [4]. Chemical shifts, indirect nuclear spin–spin couplings, PV-NMR shifts, and relaxation mechanisms associated with chemical-shift anisotropy, nuclear spin - molecular rotation, and nuclear electric quadrupole interactions are considered, while the performance of exchange–correlation functionals, solvent effects, and conformational averaging is systematically assessed. This comprehensive relativistic framework aims to identify promising chiral platinum complexes for future molecular PV-NMR experiments under realistic experimental conditions.

References

[1] R. Berger and J. Stohner, WIREs Comput. Mol. Sci. (2019), 9, e1396.
[2] (a) A. L. Barra, J. B. Robert, and L. Wiesenfeld, Phys. Lett. A (1986), 115, 443–447; (b) A. L. Barra, J. B. Robert, and L. Wiesenfeld, Europhys. Lett. (1988), 5, 217–222.
[3] (a) J. Eills et al., Phys. Rev. A (2017), 96, 042119; (b) E. Van Dyke et al., Phys. Chem. Chem. Phys. (2025), 27, 6092–6103.
[4] (a) C. van Wüllen, Z. Phys. Chem.(2010), 224, 413–426; (b) K. Gaul and R. Berger, J. Chem. Phys. (2020), 152, 044101; (c) S. A. Brück, N. Sahu, K. Gaul, and R. Berger, J. Chem. Phys. (2023), 158, 194109; (d) M. T. Colombo Jofré et al., J. Chem. Phys. (2022), 157, 064103.

P4

Quantifying and Eliminating Self-Interaction Error in Density Functionals: A Recipe for Traditional and Machine-Learning DFT

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The self-interaction error (SIE) is one of the key challenges for density functional theory, severely affecting reaction energies, quality of self-consistent densities, and dissociation limits. Constrained density functional theory (CDFT) allows for reconstructing the self-interaction free density and energy for both regular and also more recent deep-learning density functionals, like Skala. For different dissociation curves of charged molecules, we show that CDFT can recover the physically correct behavior. The CDFT approach restores the dissociation for both GGA and hybrid functionals. The Skala functional, while generally showing reduced SIE like a hybrid, improves significantly with CDFT and can match accurate CCSD(T) values for most systems. While deep learning based functionals address part of the SIE through parameterization, fundamentally they still retain SIE. CDFT offers a practical way to overcome these limitations and unlock more accurate and robust descriptions of SIE prone systems.

P5

Adsorption-Induced Structural and Optical Changes of a thioindigo-type photoswitch on Au(111)

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Peri-anthracenethioindigo (PAT) is an all-red-light-responsive indigoid photoswitch whose extended π-conjugation enables switching in the visible and near-infrared spectral region [1]. A distinctive feature of PAT is the pronounced structural difference between its bowl-shaped E-isomer and step-like Z-isomer in solution [1,2]. Isomerization therefore requires substantial geometric flattening of the molecule and was previously found to proceed exclusively on the triplet manifold [2]. These unique structural and photophysical properties make PAT a promising building block for dye-sensitized functional materials and optically responsive surfaces. However, practical applications require immobilization on surfaces, where molecule-surface interactions may significantly alter both the geometric and electronic structure of the molecule, raising the question whether the characteristic switching behavior of PAT can be retained upon adsorption on metallic substrates.
Here, we employ density functional theory (DFT) and time-dependent DFT to investigate the structural and optical properties of PAT adsorbed on Au(111). We analyze the adsorption of the E- and Z-isomers as well as twisted intermediate structures relevant to the switching pathway. Our calculations reveal pronounced surface-induced flattening of the molecule, substantially reducing the geometric differences between the two isomeric forms. Exciton analyses further shows how adsorption modifies the nature and energetic position of the low-energy optical excitations. Overall, the results provide molecular-level insights suggesting that PAT retains its ability to be excited by all-red light upon adsorption on the gold surface. Although adsorption leads to a flatter molecular geometry, the significant adsorption energy is expected to increase the effective switching barrier, indicating that switching on the surface may be less facile than in solution [2].

[1] L. Köttner, E. Ciekalski, H. Dube, Angew. Chem. Int. Ed. 2023 62, e202312955 DOI:10.1002/anie.202312955
[2] M. Hartinger, M. Herm, C. Schüßlbauer, L. Köttner, D. Guldi, H. Dube, C. Müller, Angew. Chem. Int. Ed. 2025, 64, e202510626 DOI: 10.1002/anie.202510626

P6

Molecules in Quantum Solids: Protonated Methane in para-Hydrogen Matrices

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Protonated methane (CH_5^+) is a non-classical carbocation characterized by three-center two-electron (3c − 2e) bonding. It is a key reactive intermediate in superacid environments and interstellar space. Its exceptionally flat potential energy surface enables highly correlated, large-amplitude motions among all constituent atoms, facilitating rapid interconversion across all 120 equivalent isomers. Consequently, CH_5^+ exhibits a remarkably complex vibration-rotation-tunneling infrared spectrum. Despite extensive experimental and theoretical efforts, stabilizing and fully characterizing this highly fluxional molecule and its isotopologues remains an ongoing challenge. In this work, using quantum path integral dynamics at CCSD(T) accuracy, we stabilized the structures of CH_5^+ and its isotopologues by embedding them in a *para*-hydrogen (p–H_2) quantum solid at 4 K. We observe that p–H_2 matrices stabilize distinct isotopomers that are rarely observed in the gas phase, thereby allowing the computation of isotopomer-specific infrared spectra at coupled cluster accuracy. These p–H_2 matrices produce more well-structured spectra compared to the gas phase at 4 K, and we are able to assign them using our dynamic mode-decomposition analysis of vibrational spectra.

P7

Overcoming Variational Collapse in Excited-State Simulations: A Benchmark of a Optimally Tuned Constraint-Based Orbital-Optimized Excited-State Approach

M. Asbach, J. Kussmann, C. Ochsenfeld (University of Munich, LMU/Germany)

The precise simulation of excited-state (ES) properties, particularly with regard to charge-transfer phenomena and complex spectroscopic parameters, remains a fundamental challenge within the field of computational chemistry. Whilst time-dependent density functional theory (TD-DFT) within the Tamm-Dancoff approximation (TDA) is widely utilised, even the application of system-specific tuning merely yields moderate accuracy and still struggles to perfectly capture all spectral features. In order to address this limitation, the optimally tuned constraint-based orbital-optimised excited-state (OT-COOX) method is introduced as a promising, computationally efficient alternative for modelling complex spectra.

The present study aims to systematically analyse the OT-COOX method, building upon these theoretical advantages. The framework is further expanded through the application of non-orthogonal configuration interaction (NOCI-COOX) to ensure proper spin symmetry and to capture multi-state couplings. When evaluated alongside multiple extant theoretical investigations, the overarching methodology yields illustrative results that are of a highly satisfactory nature. This combined framework has been demonstrated to exhibit the capacity to model complex electronic spectra and adequately describe CT transitions, thereby circumventing the fundamental limitations of conventional TD-DFT.

P8

Neutral electronic excitations from an iterative solution of the Bethe-Salpeter equation

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Green's function methods provide an attractive alternative to traditional post-Hartree-Fock wavefunction methods and to density functional theory (DFT), as they admit systematically improvable approximations and can offer a favorable balance between accuracy and computational cost for both molecular systems and solids. In practice, however, Green's function methods may be computationally demanding, leading to the use of a wide variety of computational protocols and approximations.
While one-particle Green's functions provide access to phenomena such as ionization and electron attachment, the study of neutral electronic excitations requires a two-particle formulation. A standard framework for this purpose is provided by the Bethe-Salpeter equation.
In practical implementations, the BSE is often cast into a Casida-like eigenvalue problem. Starting from the Casida-based BSE implementation of Wen et al. [1], we developed an alternative solver that avoids this reformulation and instead solves the BSE iteratively at Matsubara frequencies. The resulting response is then analytically continued to the real-frequency axis using the minimal pole representation method [2], which constructs a compact pole expansion from the Matsubara-frequency data. The extracted pole positions correspond to the neutral electronic excitation energies. We apply this approach using both static (frequency-independent) and dynamic interaction kernels. Results for a selection of small molecular systems are presented and compared with those obtained using the Casida formulation of the BSE, coupled-cluster singles (CCS), and coupled-cluster singles and doubles (CCSD).
References:
[1] WEN, Ming; HARSHA, Gaurav; ZGID, Dominika. Dynamically Corrected Bethe-Salpeter Equation Solver for Self-consistent GW Reference on the Matsubara Frequency Axis. arXiv preprint arXiv:2604.22187, 2026.
[2] ZHANG, Lei; GULL, Emanuel. Minimal pole representation and controlled analytic continuation of Matsubara response functions. Physical Review B, 2024, 110.3: 035154.

P9

Optimizing Input vs Correcting Output in Semiempirical Machine Learning

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Modifying semiempirical electronic-structure methods with machine learning (ML) provides a way to improve their accuracy while maintaining their low computational cost. Two different strategies are commonly used. In molecule-specific parameter learning, referred to as Parameter-ML here, an ML model predicts corrections to selected semiempirical parameters before the target property is calculated. In delta machine learning (Delta-ML), the semiempirical parametrization remains unchanged and an ML model instead learns the residual error of the calculated property. We systematically compare these strategies and investigate whether they benefit from a preceding reparameterization. Using OM2 and 6095 C7H10O2 isomers, we study atomization enthalpy as a precision-limited case and the HOMO–LUMO gap as a bias-dominated case, with traditional reparameterization and Direct ML as reference approaches. For Parameter-ML, parameter sensitivity alone does not identify the best learning target, adding a second parameter does not improve on the best single-parameter model, and a smaller molecule-specific fitting error does not necessarily lead to better downstream ML performance. Delta-ML requires fewer reference data than Direct ML. For atomization enthalpy, Parameter-ML and Delta-ML reach similar errors whereas prior reparameterization strongly improves Parameter-ML for the HOMO–LUMO gap. Transfer calculations further show an advantage of the OM2-based approaches for atomization enthalpy outside the fixed C7H10O2 composition, but not for the HOMO–LUMO gap.

P10

Beyond van der Waals: High-Throughput Prediction of 2D Materials from Non-Layered Crystals

T. Barnowsky (TU Dresden / DE & HZDR / DE), R. Friedrich (TU Dresden / DE & HZDR / DE)

The search for novel two-dimensional (2D) materials has become a central theme in materials and theoretical chemistry, driven by the exceptionally rich landscape of properties that emerge upon dimensionality reduction [1]. Considerable progress has been made for van der Waals (vdW) layered solids, where exfoliation is comparatively well understood. However, existing computational strategies remain largely constrained to this subclass of materials and provide limited predictive power beyond it [2]. In contrast, recent experimental findings indicate that structurally stable 2D sheets can also be obtained from non-layered, non-vdW bulk crystals [3,4], pointing to the need for more general theoretical descriptions of exfoliability and crystal cleavage.
In this work, we present a universal computational framework for identifying cleavage pathways and predicting exfoliable 2D sheets directly from bulk crystal structures [5]. At the core of this approach is the eXfoliation and Cleavage Potential (XCP), a descriptor designed to efficiently capture weak out-of-plane bonding motifs across chemically diverse solids. This enables high-throughput screening of large materials spaces beyond the limitations of layered crystal chemistry. Using this framework, we identify several thousand previously unrecognized candidate 2D materials embedded within non-layered bulk system, thereby providing a unified theoretical route for expanding the landscape of low-dimensional matter beyond conventional vdW systems.

[1] M. N. Gjerding et al., 2D Mater. 8, 044002 (2021)
[2] N. Mounet et al., Nat. Nanotechnol. 13, 246 (2018)
[3] A. P. Balan et al., Mater. Today 58, 164 (2022)
[4] H. Kaur and J. N. Coleman, Adv. Mater. 34, 2202164 (2022)
[5] T. Barnowsky, C. Timm, and R. Friedrich, Nat. Commun., accepted (2026)

P11

Exploiting Hard-Core Boson Structure in Quantum Chemistry

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The hard-core boson (HCB) formalism restricts electronic configurations to paired electrons, allowing the electronic structure problem to be expressed directly in terms of electron-pair creation and annihilation operators [1]. This reduces the effective Hilbert space while preserving important electron-correlation effects, offering a computationally efficient approximation for many chemical systems. Here, we explore the HCB formalism in two complementary applications. First, we employ the separable pair ansatz (SPA) [1], which approximates the many-electron wavefunction as a product of independently correlated electron pairs and naturally admits an HCB representation. Systematic benchmarks demonstrate a consistent approximation across different molecular systems and correlation regimes, with favorable classical scaling and extremely shallow quantum circuits [2]. Second, we exploit the HCB structure for basis-set-free orbital refinement. Building on a fully variational approach in which the many-body wavefunction and its underlying spatial orbitals are refined iteratively [3], we reformulate the energy functional and orbital-update equations directly in the HCB representation. This considerably simplifies the orbital update, replacing the general four-index two-particle reduced density matrix with a two-index HCB pair-density matrix. We apply the resulting refinement to both FCI and SPA calculations. Together, these applications demonstrate how the HCB formalism can be exploited at both the many-body and orbital-refinement levels to reduce the complexity of correlated electronic-structure calculations.

References:
[1] J. S. Kottmann and A. Aspuru-Guzik, Phys. Rev. A 105, 032449 (2022).
[2] L. Barta and J. S. Kottmann, arXiv:2606.26393 (2026).
[3] F. Langkabel, S. Knecht, and J. S. Kottmann, APL Comput. Phys. 2, 026106 (2026).

P12

Cutting Active Spaces Beyond the Strong-Orthogonality Condition via Excitonic Renormalization

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Fragmentation approaches provide a route to extending accurate electronic-structure methods to large systems. In multi-reference electronic structure theory, however, most fragmentation frameworks rely on the strong-orthogonality condition between fragments. In general, methods such as localized active space state interaction (LASSI) and excitonic configuration interaction (ECI) therefore require an impractically large amount of states per fragment for weakly interacting systems, prohibiting their applicability to various chemically relevant problems such as defect interaction and small molecule activation.
The recently introduced excitonic renormalization (XR) framework overcomes this limitation by enabling a modular separation of local and non-local correlation effects. Within XR, fragment interactions are recovered ab-initio from inexpensive integrals and strictly local transition densities.
Based on XR, I present a systematically convergent series expansion that starts from the strong-orthogonality limit and progressively incorporates inter-fragment coupling. Each order relaxes the orthogonality constraint while preserving the modular structure of the XR Hamiltonian. An efficient algorithm for compact optimization of fragment information is also introduced.
The approach is demonstrated for a molecular dimer with pronounced multi-reference character, illustrating systematic convergence beyond the strong-orthogonality regime. Hence, XR fills the gap of (multi-reference) methods efficiently making use of sparse Hamiltonians in an explicit manner beyond the strong-orthogonality condition.

P13

Analytic Gradients and Geometry Optimization for Orbital-Optimized Pair Coupled Cluster Doubles

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We introduce a reusable geometry-optimization engine in PyBEST for analytic, gradient-driven molecular structure optimization, with particular emphasis on orbital-optimized pair coupled- cluster doubles (OOpCCD/AP1roG). The engine interfaces PyBEST with the geomeTRIC optimizer, combining analytic electronic-structure gradients from PyBEST with the translation–rotation–internal coordinate (TRIC) framework and robust convergence machinery. Specifically, we present the first implementation of analytic OOpCCD nuclear gradients within a Lagrangian formalism. The approach is generally applicable to seniority- zero wavefunctions with orbital optimization and response one- and two-particle reduced density matrices. Owing to the seniority-zero structure of pCCD and orbital stationarity, the gradient equations are compact, reduce the storage of the two-particle reduced density matrix, and avoid finite-difference differentiation of wavefunction parameters. Validation on representative closed-shell systems shows that the PyBEST–geomeTRIC workflow converges robustly and reproduces reference equilibrium geometries and energies within tight tolerances. OOpCCD structural parameters deviate by about 0.02 Å from CCSD(F12c)(T*) and about 0.01 Å from MP2 reference bond lengths, while bond-angle deviations are generally below 1.

P14

Simulating Polymer Scission in Ultrasound Baths using Molecular Dynamics

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Ultrasound has been known for many decades as a way to induce chemical reactions. The reactivity of ultrasound is based on so-called cavitation events, which locally generate high pressures of several hundred atmospheres and temperatures of up to 5,000 °C. While ultrasound treatment can initiate many reactions, the field of polymer degradation deserves special mention. Here, the ultrasonic properties are capable of splitting polymer chains in their center up to a limiting polymer size, depending on the sonication and reaction conditions.[1] Simulations using Brownian dynamics (BD) were able to replicate these ultrasonic effects and simulate the chain degradation processes at rates similar to those observed in the experiment, as well as predict the scission at the center of the polymer chain.[2] Although BD are ideally suited for simulating such large systems, considerable approximations are made to achieve these results, such as a rough coarse-graining and highly simplified energy terms. In our research we attempted an all atom molecular dynamics (MD) approach simulating these reactions for a model polymer system. This is important since, even though BD are great in predicting macroscopic properties, the specific chain scission mechanisms remains unclear due to the approximations used. Although certain approximations are also used in MD simulations, force fields nevertheless provide significantly more information about the scission process at the atomic level, while further investigations at the quantum mechanical (QM) level are required to determine the exact cleavage mechanism, including a local strain analysis using the JEDI (Judgment of Energy Distribution) model. Furthermore, there is a poor understanding of how the direction of the ultrasonic shock wave affects the polymer. This is of interest, as an ultrasonic bath represents a highly random environment in which, theoretically, ultrasonic shock waves can impact the polymer from any possible angle. By investigating these conditions, we hope to understand how ultrasonic treatment can lead to reorientation and elongation before chain scission occurs.

1. Schaefer, Mark, et al. Macromolecules 49.5 (2016): 1630-1636.
2. Lorenzo, Turetta, and Lattuada Marco. Ind. Eng. Chem. Res. 60.29 (2021): 10539-10550.

P15

Multiscale Modelling of notched PMMA Polymers

Florian Bischoff (University of Bremen), Alexander Dellwisch (University of Bremen), Tim Neudecker (University of Bremen)

Finite Element Method (FEM) Simulations are used to simulate a wide variety of different materials and processes, like car crashes[1], airplanes[2] or even crystal behavior[3]. The material models used in these simulations are most often derived from experimental data[4]. This leads to a host of different problems, like high dependence on the quality of these experiments and the cost and time investment needed to perform them. In this project we aim to replace this experimental approach by deriving the parameters required in FEM via Molecular Dynamics (MD) simulations. These are used to determine material values, such as the elastic modulus or Poisson's ratio, and generate stress-strain curves through the deformation of bulk polymers[5]. In this first part of the project, bulk polymer systems with defects are set up, where, on two opposite sides of the simulation box, elliptic notches of varying width and height are incorporated. Strain-stress curves and elastic moduli are taken from these simulations to model similarly shaped samples on the FEM level. The effects of the notch size on the notch factor, notch tip stress and the stress distribution in the cross section between the two notches are investigated.

[1] Usama Idrees, Sajjad Ahmad, Imtiaz Alam Shah, Muhammad Talha, Rehman Shehzad, Muhammad Amjad, Seyed Saeid Rahiamin Koloor, Journal of Engineering Research 2023, 11, 100007.
[2] M. Roellig, L. Schubert, U. Lieske, B. Boehme, B. Frankenstein and N. Meyendorf, 11th International Thermal, Mechanical & Multi-Physics Simulation, and Experiments in Microelectronics and Microsystems (EuroSimE) 2010, 1-9.
[3] A. Ma, F. Roters, D. Raabe, Computational Materials Science 2007, 39 ,91-95.
[4] D. Huri, T. Mankovits, 2018 IOP Conf. Ser.: Mater. Sci. Eng. 2018, 393, 012018.
[5] Sourabh Kumar, Baris Demir, Alexander Dellwisch, Lucio Colombi Ciacchi, Tim Neudecker, Macromolecules 2023; 56 (21), 8438–8447.

P16

Stochastic-DDCI: Accurate Magnetic Modeling in Large Active Spaces

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The simulation of magnetic properties in strongly correlated systems remains a central challenge in electronic structure theory. The Difference-Dedicated Configuration Interaction (DDCI)[1] method is widely regarded as a gold standard for computing magnetic exchange couplings, but its applicability is limited to small magnetic systems due to the steep growth of the configuration-interaction space with the number of correlated electrons. Here we introduce a stochastic formulation of DDCI[2] based on full configuration interaction quantum Monte Carlo and the generalized active space framework, which largely alleviates the computational bottleneck of conventional DDCI. This development extends DDCI methodologies to substantially larger active spaces and opens the door to the study of more complex magnetic systems.

[1] Miralles J. Castell O., Caballol R., Malrieu J.-P., Chem. Phys, 1993, 172, 33-43.
[2] Bonfirraro L., Weser O., Calzado C. J., Vincent R., Li Manni G., JCTC, 2026, 22, 6572–6583.

P17

Training and application of machine-learned force fields for catalytically active nanoparticles

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Small metal nanoparticles are frequently subjects of catalytic studies and have shown exceptional catalytic activity. To be able to understand the catalytic behavior and predict the reactivity of small particle based catalyst, theoretical methods describing the dynamic properties of metal clusters are of interest. One particular challenge is to be able to scan a wide range of reaction conditions and catalyst compositions to optimize catalytic performance.
Recent developments in Machine Learning (ML) technology have enabled the construction of so-called foundation models. The aim of a foundation models is to develop an interatomic potential, that is able to predict the total energy and atomic forces for any arbitrary chemical system. Finetuning on configurations generated for a specific type of system using a foundation model gives the opportunity to refine these models. This makes it possible to increase the accuracy and predictive power of the model for a wide range of elements and structures and makes machine-learned foundation models the method of choice for molecular dynamics (MD) simulations of catalytically active metal clusters.
This poster presents a study on workflow optimization and training dataset construction for the finetuning process of a Message Passing Atomic Cluster Expansion (MACE) foundation model. We aim to develop a reliable methodology to finetune the MACE-MH-1 model for different cluster sizes and elemental compositions for transition metal clusters.

[1] Yuan, E. C.-Y.; Liu, Y.; Chen, J.; Zhong, P.; Raja, S.; Kreiman, T.; Vargas, S.; Xu, W.; Head-Gordon, M.; Yang, C.; Blau, S. M.; Cheng, B.; Krishnapriyan, A.; Head-Gordon, T. Foundation models for atomistic simulation of chemistry and materials. Nature Reviews Chemistry 2026, 10, 212–230.
[2] Batatia, I.; Lin, C.; Hart, J.; Kasoar, E.; Elena, A. M.; Norwood, S. W.; Wolf, T.; Csányi, G. Cross Learning between Electronic Structure Theories for Unifying Molecular, Surface, and Inorganic Crystal Foundation Force Fields. 2025.

P18

Generalizing the Judgement of Energy DIstribution (JEDI) Strain Analysis for Periodic Systems under Lattice Deformation

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Mechanical deformation, such as stretching or compression, can alter the geometric, energetic, and spectroscopic properties of materials and induce chemical transformations. Understanding how mechanical strain is accommodated within a material is therefore essential for rationalizing its response to deformation. The Judgement of Energy DIstribution (JEDI) analysis quantifies strain energy contributions associated with individual bonds, angles, and dihedral angles within the harmonic approximation. JEDI can be used with a variety of electronic structure programs for molecules, supramolecular clusters, and two- or three-dimensional periodic systems through the Atomic Simulation Environment (ASE). Here, we present a generalization of JEDI that enables the analysis of strain energy distributions induced by lattice deformation in periodic systems. The extension employs an augmented Hessian that explicitly incorporates the lattice vectors and their coupling to the scaled atomic coordinates. This allows the strain energy associated with a lattice deformation to be decomposed into contributions from individual redundant internal coordinates. Combined with a color-coded three-dimensional visualization, the generalized JEDI approach enables intuitive analysis of strain energy distributions in materials ranging from covalent periodic materials to molecular crystals with intermolecular interactions. The method can be applied to arbitrary lattice deformations and systems containing structural defects, substantially broadening the scope of JEDI and providing a quantitative framework for relating lattice deformation to its energetic response at the most fundamental level.

P19

Planar to Dewar isomerisation from azaborine to azaboranaphthalene under mechanistic, electronic, and substituent control

M. Bühler (University of Würzburg/DE), M. I. S. Röhr (University of Würzburg/DE)

BN incorporation offers a promising strategy to tune photoswitches for molecular solar thermal energy storage (MOST) [1,2]. Here, we examine the planar to Dewar valence isomerisation of 4a,8a-azaboranaphthalene, a π-extended BN-doped analogue of azaborine, and compare its behaviour with related carbon and azaborine systems. Using DFT, TD-DFT, nudged elastic band calculations and pathway refinements at the state-averaged XMS-CASPT2 level, we show that BN doping significantly reshapes the ground-state reaction pathway [3]. Relative to the carbon analogue the pathway becomes distinctly asymmetric through the formation of a metastable intermediate stabilized by a transient BN interaction. The transition structure exhibits strong similarity to an S0/S1 conical intersection, consistent with an efficient nonradiative relaxation channel [4]. In addition, systematic substituent screening identifies routes to red-shift excitation energies, enhance oscillator strengths, and improve Dewar formation, highlighting BN-doped azaborine frameworks as tunable candidates for MOST applications.

[1] A. Brough, A. N. Lamm, S. Y. Liu and H. F. Bettinger, Angew. Chem. Int. Ed., 2012, 51, 10880–10883.
[2] K. Edel, X. Yang, J. S. A. Ishibashi, A. N. Lamm, C. Maichle-Mössmer, Z. X. Giustra, S. Y. Liu and H. F. Bettinger, Angew. Chem. Int. Ed., 2018, 57, 5296–5300.
[3] M. Bühler and M. I. S. Röhr, Phys. Chem. Chem. Phys., 2026, DOI: 10.1039/D6CP01750A.
[4] F. Sturm, M. Bühler, C. Strapper, J. S. Schneider, H. Helten, I. Fischer and M. I. S. Röhr, Phys. Chem. Chem. Phys., 2024, 26, 7363–7370.

P20

Solving Bethe Salpeter equation using tensor decomposition

Joanna Bukojemska, Dominika Zgid, (University of Warsaw)

Optical spectroscopy, including absorption and photoluminescence techniques, is one of the most widely used methods for investigating the electronic structure of materials. However, a full understanding of optical spectra and the assignment of spectral features require advanced theoretical modeling of neutral excitations. While the single-particle Green's function formalism is sufficient for describing N $\pm$ 1 processes, such as ionization and electron attachment, neutral excitations necessitate an extension to two-particle Green's functions. Such a description is naturally provided by the Bethe–Salpeter equation (BSE), which relates the two-particle response function to a two-particle interaction kernel. In our study, we focus particularly on electron–hole correlations. Historically, applications of the BSE have relied on a variety of uncontrolled approximations. Although these approximations reduce computational cost, they also introduce errors that are difficult to quantify systematically. Our goal is to use the Matsubara Green's function formalism and employ modern applied mathematics techniques, such as tensor decomposition methods, to develop efficient implementations of the Bethe–Salpeter equation while systematically removing uncontrolled approximations. We believe that employing only well-justified approximations—and eliminating those introduced solely to reduce computational cost that can cause significant errors, such as the Tamm–Dancoff approximation (TDA)—will yield more accurate results. Additionally, we aim to avoid unnecessary simplifications of the BSE, such as the static kernel approximation, which renders the study of phenomena like double excitations impossible. Furthermore, the Matsubara Green's function formalism allows us to introduce self-consistent improvements without loss of information, as optical spectra can still be obtained by analytical continuation from the imaginary axis to the real axis. Moreover, tensor decomposition techniques, such as tensor hypercontraction (THC) and density-fitted resolution of the identity (RI-DF), can reduce the computational cost of implementing the BSE from $O(N^6)$ down to $O(N^4)$. The resulting formalism will then be applied to investigate optical excitations in both molecular and periodic systems.

P21

Investigating Higher-Order Excited-State Properties with Constraint-Based Orbital-Optimized Excited States (COOX)

A. Martin R. J. Dagleish (LMU Munich/DE), B. Dr. Jörg Kußmann (LMU Munich/DE), Z. Prof. Christian Ochsenfeld (LMU Munich/DE)

Higher-order excited-state properties such as nuclear Hessians and vibronic spectra are sensitive to level of theory and have significant computational cost. Conventional response-based approaches suffer from root crossings, degenerate excited states, and the need for state-specific relaxed reference orbitals. The Constraint-based Orbital-Optimized eXcited-state (COOX) method [1] promises to circumvent these issues by obtaining the excited state as a saddle point of the ground-state density functional under a constraint potential constructed from, e.g., (s)TDA amplitudes, yielding variationally optimized, state-specific orbitals within a single-reference framework, potentially at the cost of ground-state properties. Note that COOX was subsequently extended to multi-reference systems as NOCI-COOX [2], to include static-correlation effects.
A critical practical aspect is the necessity of Fermi-smearing fractional occupation number (FON) MO-CPSCF theory for certain COOX states. However, since the partially occupied space is typically small, the FON overhead is negligible.
First illustrative examples include the computation of vibronic spectra using the state-specific COOX ground- and excited-state geometries and Hessians utilizing symmetric finite-difference evaluations, requiring only independent single-point COOX calculations at displaced geometries. Franck–Condon and Herzberg–Teller absorption and fluorescence band shapes are obtained with full Duschinsky mixing and zero-point corrections, facilitated by Eckart alignment to remove translation and rotation impurities. Utilizing the time-dependent thermal autocorrelation function approach derived from De Souza's formulation [3] efficiently yields temperature-dependent absorption and fluorescence profiles, which circumvent the explicit summation. This approach is extensible in a straightforward manner to other static molecular properties for electric and magnetic perturbations.

[1] J. Kussmann, Y. Lemke, A. Weinbrenner, C. Ochsenfeld, J. Chem. Theory Comput. 20, 8461–8473 (2024).
[2] Y. Lemke, J. Kussmann, C. Ochsenfeld, J. Chem. Theory Comput. 21, 10193–10211 (2025).
[3] B. de Souza, F. Neese, R. Izsák, J. Chem. Phys. 148, 034104 (2018).

P22

Capturing Activity Trends in Metal-Organic Framework Catalysis using Minimal Active Site Models

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Metal-organic frameworks (MOFs) offer immense potential as catalysts owing to their modular and tunable structures, yet their vast combinatorial design space makes discovery challenging. Although realistic models of MOF active sites are effective for detailed mechanistic investigations, their complexity and system-specific nature hinder transferable, predictive catalyst design. Here, we demonstrate that activity trends in MOF catalysis can be captured using representative minimal active site models (MASMs) that retain only the local metal coordination environment, significantly reducing computational cost.[1] Using quantum chemical data from these MASMs, we derive regression models [2] that map catalytic activity onto easily computable descriptor variables. These descriptors enable rapid prediction and comparison of realistic multinuclear active sites across diverse MOF families represented by more than ten thousand experimentally reported crystal structures. Our framework rationalizes the exceptional activity of experimentally reported catalysts, identifies specific activity-limiting constraints in these active sites, highlights avenues to overcome them, and predicts candidate materials that surpass the performance of the state-of-the-art catalysts.[1] The present approach lays a robust foundation and a starting point for establishing a powerful theoretical framework for predicting and optimizing active sites in crystalline and glassy MOFs for a wide range of chemical transformations.

References:
[1] Das, S.; Heine, T. ChemRxiv, 2026, doi: 10.26434/chemrxiv.15006082/v1
[2] Laplaza. R.; Das, S.; Wodrich M. D.; Corminboeuf, C. Nat. Protoc. 2022, 17, 2550-2569.

P23

Unravelling Molecular Motor Coupling in Porous Molecular Scaffolds at Atomic Scale

Shreya Das, Saeed Amirjalayer, (Heidelberg University)

Molecular machines and motors are external stimuli such as light into controlled motion at the nanoscale. The motion of individual molecular motors has been extensively studied. However, less in known about how several motors operate when placed close to each other. In solution, these interactions occur randomly, making them difficult to study systematically. Metal Organic Frameworks (MOFs) provide a controlled platform for tailoring the interactions between molecular motors due to their building block, which allows the distance and geometry between motors to be precisely adjusted.
Motivated by this, we investigated MOFs featuring organic linkers containing light-driven molecular motors. We examine how variations in the intermolecular interactions between these motors shape the energy landscape. We analyze the impact on the relative stability of the two ground-state conformations and on corresponding motor dynamics along the unidirectional rotation. Moreover, the influence on optical properties is revealed enabling comparison with experiment measurements.
Three scaffolds with different linker length were compared. For each scaffold, configurations with 0 to 100 percent of motors in the metastable state are generated using an ab-initio parametrized force field. Resulting energy distributions reveal how scaffold geometry influences energetic stability. As metastable concentration increases, these distributions shift and overlap, indicating strong intermolecular interactions and modulation of the local potential energy landscape. To investigate this coupling, molecular dynamics simulations at different temperatures reveal how neighboring linkers constrain or facilitate one another during rotation within the periodic framework and how this influences resulting motor dynamics. TD-DFT calculations on dimers of adjacent linkers extracted from the lowest energy structures at each concentration enable to link local inter-motor coupling to variation in the UV/Vis absorption spectra.
Overall, coupling between molecular motors is the key factor linking local motion to structural and optical responses. Understanding these inter-motor couplings provides access to molecular-based design concepts for developing functional materials based on light-responsive molecular motors.

Reference
1. Kolodzeiski, E., and Amirjalayer, S. (2021). Collective structural properties of embedded molecular motors in functionalized metal organic frameworks. Physical Chemistry Chemical Physics, 23(8), 4728 to 4735. doi 10.1039/d0cp06263d

P24

Effects of Conterion on the Solvation Structure and Migration Kinetics of Hydroxide Ions in Concentrated Aqueous NaOH Solutions

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Hydroxide ion (OH^-) transport in aqueous solutions is primarily governed by the Grotthuss mechanism, in which charge migration occurs through proton transfer along hydrogen bond networks rather than through conventional vehicular diffusion. Despite its fundamental importance, the influence of counterions on this transport process remains unclear. In this study, we employ reactive molecular dynamics simulations, i.e., ReaxFF, over a broad range of NaOH concentrations to systematically examine how increasing ion concentration alters solvation structure and OH^- transport dynamics. Our results reveal the presence of two distinct OH^- populations with markedly different transport characteristics. Water-solvated OH^- ions exhibit rapid proton-transfer dynamics, whereas Na^--coordinated OH^- species, $[OH^- \ldots Na^+]$, show substantially hindered transport, characterized by proton-transfer free energy barriers that are higher and significantly slower transfer kinetics. As the NaOH concentration increases, progressive disruption of the hydrogen-bond network, together with enhanced ion pairing, leads to a pronounced reduction in OH^- diffusion coefficients. These findings provide a molecular-level explanation for the anomalously low OH^- mobility observed experimentally in concentrated alkaline solutions. Overall, this work elucidates the microscopic origins of concentration-dependent OH^- transport and identifies counterion coordination as a critical factor controlling proton-transfer efficiency in alkaline electrochemical environments.

P25

The Design of Problem-Specific Molecular Descriptors

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Molecular descriptors encode topological, structural, and sometimes electronic information of a molecule into a feature vector that is used as input for machine learning algorithms. The choice of these descriptors is decisive for model performance, and designing problem-tailored descriptors can often be fruitful. We discuss several approaches and practical considerations, along with comparative studies and implications for active learning workflows, with the aim of providing insights for researchers developing effective molecular representations.

P26

Reaction Discovery on Copper Surface Using Periodic Nanoreactor Molecular Dynamics with Machine-Learned Force Fields

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Understanding reaction mechanisms on transition-metal surfaces is essential for optimizing heterogeneous catalysis. However, relying on chemical intuition can overlook unconventional and hidden reaction pathways. While nanoreactor molecular dynamics (NMD) simulations [1] combined with periodic boundary conditions [2] and density functional theory (DFT) offer agnostic pathway discovery, their broad application is limited by high computational costs. Machine-learned force fields (MLFFs) can accelerate these simulations, but they currently lack the accuracy and reliability needed for the highly distorted, reactive structures sampled during NMD. In this work, we developed an iterative workflow in which a many-body atomic cluster expansion model (MACE) [3] is iteratively fine-tuned using structures generated during periodic NMD simulations. Here, the hydrodeoxygenation of glycerol on a copper surface was investigated. Different acceleration strategies were employed to enhance the sampling of adsorption and reaction events involving glycerol and molecular hydrogen on the copper surface. The NMD simulations successfully reproduced key intermediates, such as dihydroxyacetone and hydroxyacetone, and discovered the main reaction pathway to 1,2-propanediol [4], effectively validating our approach. Furthermore, extraction of reactants and products from reactive trajectories followed by MLFF-based pathway refinement, free-energy corrections based on phonon calculations, and DFT single-point re-evaluation revealed a feasible, previously overlooked side-reaction mechanism. The proposed workflow establishes a practical framework for the automated, reaction-agnostic mechanistic exploration of heterogeneous catalytic reactions at near-DFT accuracy.

References

[1] L.-P. Wang, A. Titov, R. McGibbon et al., *Nat. Chem.* **6**, 1044–1048 (2014).
[2] D. Deißenbeck, P. Meier, W. A. Kopp et al., *Angew. Chem. Int. Ed.* **2026**, *65*(6), e14074.
[3] I. Batatia, D. P. Kovacs, G. Simm et al., *Adv. Neural Inf. Process. Syst.* **35**, 11423–11436 (2022).
[4] S. Gupta, A. Rajan, E. Fako et al., *J. Phys. Chem. C* **2025**, *129*(13), 6262–6274.

P27

Recent Developments in the Algebraic Diagrammatic Construction Framework

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The hierarchy of algebraic diagrammatic construction (ADC) schemes are reliable, accurate, hermitian, and size-consistent perturbation-theoretical ab-initio methods for the description of ionized, electron-attached and electronically excited states. While ADC originates from Green's function or propagator theory, more recent developments instead utilize the intermediate state representation (ISR) formalism, which gives access to state and transition properties of arbitrary operators.

We present recent theoretical developments as well as their implementation in adcc, an open-source Python/C++ module available on PyPI and Conda. The combination of Python with C++ enables a rapid development cycle, while the C++ backend guarantees a high computational performance. Currently, adcc is interfaced to the popular open-source PySCF and Psi4 programs.

Moreover, we present a highly parallel ADC(2) implementation based on a quadrature-based decomposition of the electron repulsion integrals. Compared to conventional formulations, Q-ADC features a reduction of both arithmetic scaling and memory footprint.

P28

Computational Studies of PFAS Detection, Adsorption, and Degradation

Javier Camargo Díaz (FU Berlin), Tobias Reitz (FU Berlin), Beate Paulus (FU Berlin)

Per- and polyfluoroalkyl substances (PFAS) are persistent pollutants which remediation presents three challenges: detecting them at subpicomolar concentrations, removing them from water, and destroying them into useful products. We approach all three with quantum-chemical calculations, focusing on the interactions between PFAS and materials. For detection, we model a fluorinated 2D polymer (2DF1) that quenches fluorescence upon selective PFAS interaction [1]. Periodic models reproduce the experimental band gap and a stacking-dependent narrowing consistent with agglomeration-induced quenching, while binding energies from GFN2-xTB/CREST conformer sampling of finite-size models, refined with hybrid DFT (TPSSh-D3/def2-TZVP), reproduce the selectivity for PFOA and trace it to fluorine-specific, hydrophobic, and electrostatic contributions rather than to a single descriptor.

While 2DF1 detects PFAS selectively, it is itself a fluorine-containing material, a drawback for remediation. As a fluorine-free alternative, Rainer Haag's group synthesised a quaternised polyethylenimine (qPEI) polymer for PFAS adsorption [2]. To understand the PFAS bindings modes we develop a minimal model of their functionalised monomers and quantify the competition between deprotonated and netrual trifluoroacetic acid (TFA), a problematic ultrashort-chain PFAS, and water and counter-ions for the binding sites. This rationalises what drives TFA uptake against water and points to functionalisations favouring PFAS over solvent molecules.

Finally, for degradation, we turn to PFAS bound at Lewis-acidic sites of a defect-bearing material [3]. Modelling representative edge and in-plane defect motifs, we find this interaction substantially lowers the energetic cost of C–F and C–C cleavage relative to the freestanding PFAS, pointing to defect chemistry as an activation route toward remediation.

Taken together, the three cases trace PFAS–material chemistry from recognition through retention to bond activation, translating an interaction-level picture into design guidance for remediation materials.

[1] Ghanbari, Reza, et al. "Two-dimensional fluorinated nanomaterials for fast and selective detection of PFAS at subpicomolar concentrations." Matter 9.7 (2026).

[2] Meier, Patrick Arkadiusz, et al. "qPEI Thin Films as Adsorbers of Different PFAS in Aqueous Media." Macromolecular Chemistry and Physics 227.13 (2026).

[3] Yang, Nanyang, et al. "Solvent-free nonthermal destruction of PFAS chemicals and PFAS in sediment by piezoelectric ball milling." Environmental Science & Technology Letters 10.2 (2023).

P29

Frozen-Density Embedding for Mixed-Reference Spin-Flip TDDFT

S. Dier (University of Münster/DE), J. Neugebauer (University of Münster/DE)

We present the first combination of mixed-reference spin-flip TDDFT (MRSF-TDDFT) with QM/QM embedding, implemented in the Serenity program. Spin-flip TDDFT overcomes several well-known limitations of conventional TDDFT, such as the description of states with multireference character and of conical intersections involving the ground state, but its response states suffer from severe spin contamination. The mixed-reference formulation largely removes this contamination while retaining these advantages, which makes it well suited to computational photochemistry. While MRSF-TDDFT has already been combined with QM/MM embedding, a QM/QM approach has, to the best of our knowledge, not been reported so far.

As QM/QM embedding we employ uncoupled frozen-density embedding (FDEu), which makes the study of photochemistry with MRSF-TDDFT in complex environments feasible. The environment enters through an embedding potential in the ground-state SCF, so that the triplet reference orbitals carry the environmental effect, while the spin-flip response is evaluated on these orbitals.

We validate the method against supersystem calculations and apply it to the relaxed scan geometries of the maleic to fumaric acid isomerization, each surrounded by several explicit water molecules. We also report wall times, demonstrating the efficiency gain of the embedded description over supersystem calculations.

We plan to continue by implementing analytical gradients for MRSF-TDDFT in Serenity and for the embedded method, and to explore the inclusion of the environment response in the excitation step.

P30

Improving the description of electron attachment and detachment in ADC at almost no cost via repartitioning

L. B. Dittmer (University of Heidelberg/DE), A. Dreuw (University of Heidelberg/DE)

The algebraic diagrammatic construction (ADC) for the electron propagator is a well-established perturbation theoretical method for the computation of ionisation potentials (IP) and electron affinity (EA). Its second-order variant EA-ADC(2) strikes a favourable balance between accuracy and computational cost ($O(N^4)$), though a third-order description of electron detachment processes is usually necessary for satisfactory performance. So far, reducing the incurred error has always been achieved by increasing the perturbative order, which necessarily increases the computational complexity. Here, we present the first implementation of a repartitioned IP/EA-ADC scheme in second and third order, which switches out the standard Møller-Plesset choice of Hamiltonian partitioning with the size-consistent Brillouin-Wigner (BWs) partitioning. This choice can be interpreted as the partial inclusion of (n+2)-order terms into the IP/EA-ADC(n) matrix. IP/EA-BWs-ADC introduces only minimal computational overhead compared to standard IP/EA-ADC while significantly improving the description of both electron attachment and detachment processes.

P31

Improving Sample-Based Quantum Diagonalization for Strongly Correlated Electronic Structure Problems

E. Ricci (Dresden University of Technology/DE), A. R. Khamadja (University of Constantine / DZ), W. Dobrautz (Helmholtz Zentrum Dresden-Rossendorf/DE)

Accurately describing strongly correlated electronic systems remains one of the central challenges in electronic structure theory. Recent hybrid quantum–classical approaches, including Quantum Selected Configuration Interaction and Sample-Based Quantum Diagonalization, address this challenge by using a quantum device to sample electronic configurations from an approximate many-body state. The sampled Slater determinants define a compact, problem-adapted subspace in which the Hamiltonian is diagonalized classically to obtain ground-state properties. This framework shifts the role of the quantum computer from full energy evaluation to targeted configuration-space exploration, making it particularly attractive for near-term quantum devices.

In this talk, I discuss two complementary strategies for improving the efficiency and accuracy of such sampling-based diagonalization schemes. First, we employ a similarity-transformed electronic Hamiltonian that concentrates the ground-state weight into a smaller determinant subspace. This reduces the number of configurations required for an accurate classical diagonalization, but introduces a non-Hermitian effective Hamiltonian. While non-Hermiticity is often problematic for quantum algorithms, we show that it can still be exploited in a sampling-based setting by using the transformed Hamiltonian to initialize a Unitary Cluster Jastrow ansatz with physically informed single and double amplitudes. To mitigate quantum sampling errors, we further apply classical post-processing that projects measured configurations onto the correct particle-number sector.

Second, we address a key sampling bottleneck: approximate quantum states can become strongly overconcentrated on a small number of dominant configurations, especially the Hartree–Fock determinant. This leads to inefficient coupon-collector-like scaling, where already discovered determinants are repeatedly resampled while rare but important configurations remain difficult to access. To overcome this limitation, we investigate improved UCJ-based state preparation strategies. The cluster operators are constructed from a double factorization of coupled cluster T2 amplitudes and decomposed into eigenvalue-resolved components, each of which is independently time-evolved as a cluster generator. This provides a multi-state sampling perspective that broadens access to relevant configuration space, reduces sampling redundancy, and improves the reconstruction of compact ground-state subspaces for strongly correlated molecular systems.

P32

Ground state of the Hubbard model with spin-dependent linear potential

J. Dobrzyniecki (University of Warsaw/PL), T. Busch (Okinawa Institute of Science and Technology/JP)

We investigate the competition between attractive spin-spin interactions and spin-separating external forces in the ground state of a one-dimensional Fermi-Hubbard model. We consider a lattice with open boundary conditions, subject to a linear external potential whose gradient is opposite for the two spin components, so that each spin species sees a potential minimum at a different end of the lattice. Using density-matrix renormalization group (DMRG) simulations, we map the ground-state density distributions and the number of doubly occupied sites as a function of the potential gradient β and interaction strength. We identify three distinct regimes separated by critical threshold gradients: (i) a small-β regime where fermion pairing remains robust against the external potential; (ii) an intermediate-β phase-separated regime characterized by a staircase-like decrease in the doublon number, corresponding to the successive, one-by-one breaking of bound pairs; and (iii) a large-β regime where the two spin components are completely spatially separated. We complement the numerical results with a phenomenological model and a local-density approximation analysis, from which we derive closed-form analytical estimates for these critical threshold values. We also verify that the staircase structure persists under additional harmonic confinement. Our results are directly testable in cold-atom experiments, and demonstrate that a spin-dependent linear potential enables precise, integer-level control of the number of bound fermion pairs.

P33

Incorporating rotational symmetry and quantisation into instanton theory

J. Dušek (ETHZ), J. E. Lawrence (NYU), J. O. Richardson (ETHZ)

Properly describing rotational symmetry of molecules is key for accurate simulations of quantum molecular dynamics. While Ring-Polymer Instanton Theory (RPI) correctly captures zero-point energy and tunnelling effects, the treatment of rotational partition functions in this framework has so far only been done classically. Our goal is to incorporate rotational quantisation of states into RPI from first principles in order to to describe energy splittings between molecular rotational states (a spectroscopically observable quantity) and gas-phase reactions more accurately (i. e. beyond TST and standard RPI). Recently, we have extended RPI by introducing perturbative corrections (PC) [1, 2], which further improve its accuracy by describing anharmonic vibrational effects. We will describe further extensions of the perturbative approach to treat other anharmonic effects such as rotation-vibration coupling

[1] Jindra Dušek, Joseph E. Lawrence, Jeremy O. Richardson, J. Chem. Phys., 2026, 164, 024104
[2] Joseph E. Lawrence, Jindra Dušek, Jeremy O. Richardson, J. Chem. Phys., 2023, 159,014111

P34

Mechanism of Thermal Defunctionalization of Phenyl and Methyl Groups on Graphene

Diyan Unmu Dzujah (TU Dresden/DE), Thomas Brumme (TU Dresden/DE), Thomas Heine (TU Dresden/DE)

Covalent functionalization allows modification of the π-electrons of graphene, inducing local pyramidalization and distortion of the C network. In this theoretical study, we investigate the preferred configurations of phenyl and methyl functionalization on graphene and their corresponding influence through sp^2-to-sp^3 hybridization on the structural, electronic, and magnetic properties using density-functional theory. Among the investigated configurations, the cis-1,4 arrangement is the most stable and preserves the diamagnetic state. In contrast, adsorption of phenyl or methyl groups with unequal functionalization of the A and B sublattices produces uncompensated π states with ferrimagnetic ordering. The activation barriers for thermal defunctionalization are evaluated for single-adsorbate and dimer removal pathways using climbing-image nudged elastic band calculations. The calculated barriers are 1.87 and 2.07 eV for single methyl and phenyl removal, respectively, and increase to 3.00 and 3.50 eV for the corresponding dimer pathways. These results indicate a preference for sequential rather than collective removal. The single-phenyl barrier of 2.07 eV agrees with the activation energy estimated from Arrhenius analysis of the experimental Raman ID/IG ratio, further supporting this mechanism in phenyl-functionalized graphene.

P35

Accurate and scalable exchange-correlation with deep learning

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Density Functional Theory (DFT) underpins much of modern computational chemistry and materials science. Yet, the reliability of DFT-derived predictions of experimentally measurable properties remains fundamentally limited by the need to approximate the unknown exchange-correlation (XC) functional. The traditional paradigm for improving accuracy has relied on increasingly elaborate hand-crafted functional forms. This approach has led to a longstanding trade-off between computational efficiency and accuracy, which remains insufficient for reliable predictive modelling of laboratory experiments. Here we introduce Skala, a deep learning-based XC functional that surpasses state-of-the-art hybrid functionals in accuracy across the main-group chemistry benchmark set GMTKN55 with an error of 2.8 kcal/mol, while retaining the lower computational cost characteristic of semi-local DFT. This demonstrated departure from the historical trade-off between accuracy and efficiency is enabled by learning non-local representations of electronic structure directly from data, bypassing the need for increasingly costly hand-engineered features. Leveraging an unprecedented volume of high-accuracy reference data from wavefunction-based methods, we establish that modern deep learning enables systematically improvable neural exchange-correlation models as training datasets expand, positioning first-principles simulations to become progressively more predictive.

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A MACE-Based Machine Learning Potential for GNR Formation on Au (111)

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Our work is motivated by the interaction between light and matter. As a model system, we investigate the formation of graphene nanoribbons (GNRs) on Au(111).
GNR formation proceeds in two steps. First, 10,10'-dibromo-9,9'-bianthracene (DBBA) is converted into polyanthrylene via Ullmann coupling. Subsequently, polyanthrylene undergoes cyclodehydrogenation to form GNRs. We focus on the second reaction, which experiments have shown to be promoted by both charge injection and temperature, effects that can be induced by laser irradiation of the Au(111) surface.
We investigate several reaction mechanisms proposed by Björk et al. [1], Blankenburg et al. [2], and Ma et al. [3]. Our aim is to re-evaluate the minimum-energy pathways using the more accurate r2SCAN functional instead of PBE and to identify the relevant intermediates and transition states.
Finally, we compare the predicted reaction pathways and barriers with experimental observations, including the bright edge features reported by Hoffmann-Vogel and the zipping-up mechanism observed by Björk et al. As an outlook, the developed potentials can be applied to large-scale molecular dynamics simulations to study GNR growth and light-driven surface reactions on experimentally relevant time scales.
For this purpose, we developed two MACE machine-learning potentials trained on PBE and r2SCAN reference data. These potentials enable efficient Nudged Elastic Band (NEB) calculations of complete reaction pathways that would be computationally demanding with conventional ab initio methods.
As an outlook, the developed MACE potentials could be used to investigate experimentally observed phenomena, such as the bright edge features reported by Hoffmann-Vogel and the zipping-up mechanism proposed by Björk et al. Furthermore, they enable large-scale molecular dynamics simulations on nanosecond timescales, providing access to GNR growth processes under experimentally relevant conditions and, ultimately, to the investigation of light-driven surface reactions on metal surfaces.

[1] S. Blankenburg et al., Acs Nano 6, 2020 (2012).
[2] C. Ma et al., Chemical Science 12, 15637 (2021).
[3] C. Ma et. Nature Communications 8, 14815 (2017).

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Modelling Confined High-entropy Materials

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High-entropy materials (HEMs) are single-phase multi-component disordered systems with unique electronic and thermal properties that are promising for applications in the energy and electronics sectors. Yet, how this entropy-driven stabilization behaves at reduced dimensionality considering finite temperature behavior remains unclear. A detailed understanding of their surfaces and 2D variants is thus crucial for future applications. Here, we develop a general approach to model and understand the surfaces and 2D variants of these HEMs at finite temperature [1]. At the heart of our approach is the mapping of the bulk-to-surface relation based on a spectral method through ordered cell expansion [2] within the AFLOW framework [3,4]. Predictive descriptors including the entropy forming ability (EFA) [5] and disordered enthalpy-entropy descriptor (DEED) [6] are crucial to assess synthesizability. We applied our methodology to four experimentally realized bulk high-entropy carbides with reported favorable synthesizability [5,6] — MoNbTaVWC5, HfNbTaTiZrC5, HfNbTaZrC4, and HfTaTiZrC4. Our results show that the entropy-stabilization content of their 2D variants is strongly thickness dependent, decreasing from bulk to intermediate slab thicknesses due to 3D symmetry breaking. A Boltzmann statistical analysis of ensemble energetics further reveals that surface terminations deviate from ideal random mixing and are strongly composition-dependent. Critical insights into the system-wide properties such as the density of states are also provided. Overall, our approach enables a systematic route for modelling low-dimensional high-entropy materials and their surfaces at finite temperature.

[1] C.-C. Er, T. Barnowsky, R. Friedrich, manuscript submitted 2026.
[2] K. Yang., C. Oses, S. Curtarolo, Chem. Mater. 2016, 28, 6484.
[3] C. Oses, M. Esters, D. Hickes, et al., Comput. Mater. Sci. 2023, 217, 111889.
[4] M. Esters, C. Oses, S. Divilov, et al., Comput. Mater. Sci. 2023, 216, 111808.
[5] P. Sarker, T. Harrington, C. Toher, et al., Nat. Commun. 2018, 9, 4980.
[6] S. Divilov, H. Eckert, D. Hicks, et al., Nature 2024, 625, 66.

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Transcorrelating Solids: Solving the twin problems of slow basis-set and thermodynamic limit convergence of Quantum Chemistry methods at the same time.

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The accuracy of correlated electronic structure calculations of periodic and extended systems performed in finite simulation cells is compromised by two main issues: slow basis-set convergence, driven by the electron–electron Coulomb cusp at short interelectronic distances, and finite-size errors arising from Brillouin-zone discretization and the missing long-wavelength correlations in the thermodynamic limit (TDL). In small-gap, highly polarizable systems, the latter is compounded further by the infrared divergence in finite-order many-body perturbation theory, which can only be cured by an infinite resummation of particle-hole ring diagrams (RPA).

We present a transcorrelated (TC) framework that addresses both problems on an equal footing: the long-range transcorrelated approach. An exact similarity transformation of the Hamiltonian by a Jastrow factorization, performed in the TDL, yields a non-Hermitian effective Hamiltonian in which the bare Coulomb operator is dressed by two- and three-body effective potentials. Enforcing the Kato cusp condition at short range and the exact Gaskell/RPA asymptotic behavior of the optimal Jastrow correlator at long range regularizes the Coulomb singularity and screens the long-wavelength tail simultaneously, accelerating basis-set convergence and curing the IR divergence without diagrammatic resummation.

Using the uniform electron gas as a model system, we show within the long-range TC method that the transcorrelated mean-field reference energy converges to the TDL orders of magnitude faster than Hartree–Fock, with the associated effective Madelung constant decaying as $1/N$ (vs. $1/N^{1/3}$ conventionally) for long-range correlators. We further demonstrate that TC-MP2 yields a finite correlation contribution that converges rapidly to the TDL, in good agreement with quantum Monte Carlo benchmarks, while basis-set convergence accelerates from $1/M$ to $1/M^{5/3}$. These results establish long-range transcorrelation as a promising strategy for the efficient treatment of ab-initio periodic systems, extendable even to highly polarizable metals.

References

[1] H. Luo, A. Alavi, JCTC 14, 1403–1411 (2018).
[2] M. Gell-Mann, K. A. Brueckner, Phys. Rev. 106, 364 (1957).
[3] E. Christlmaier et al., JCP 159, 014113 (2023).
[4] H. Luo et al., Phys. Rev. B 113, 165146 (2026).
[5] G. G. Spink et al., Phys. Rev. B 88, 085121 (2013).

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GW Approximation within the Projector Augmented-Wave Method

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The GW approximation describes the electronic self-energy through the product of the single-particle Green's function (G) and the screened Coulomb interaction (W). It enables to compute quasiparticle correction to the DFT energies, in order to calculate accurate band structures and excitation properties.
The Projector augmented-wave (PAW) approach [1] combines the efficiency of pseudopotentials with the accuracy of all-electron methods. It relates the pseudo wavefunction to the all-electron wavefunction through a linear transformation that restores the all-electron properties.
The combination of the GW and PAW approaches is a conceptually and numerically challenging task. In the past, several attempts to this problem have been discussed [2,3,4]. In this work, we reexamine this issue in great detail, in particular the different possible choices in the computation of the oscillator strength matrix elements, the treatment of the core contribution, the inclusion or exclusion of semicore states, the numerical integration scheme, the completeness of the various basis sets involved, the limit in the sum over high energy bands in the correlation self-energy, the potential occurrence of cross terms between the plane wave and onsite basis sets, and the different implementations in the ABINIT and VASP electronic structure codes.

References

1. P. Blöchl, Phys. Rev. B 1994, 50 (24), 17953
2. B. Arnaud, M. Alouani, Phys. Rev. B 2000, 62 (7), 4464
3. M. Shishkin, G. Kresse, Phys. Rev. B 2006, 74 (3), 035101
4. J. Klimeš, M. Kaltak, G. Kresse, Phys. Rev. B 2014, 90 (7), 075125

P40

Machine-Learning-Accelerated Structure Determination Combining Surface XRD and DFT

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The determination of atomic structures represents a high-dimensional global optimization problem, requiring a large number of first-principles evaluations of the energy. Machine-learning-accelerated algorithms can significantly decrease the number of these calculations, making global optimizations considerably more efficient. An example is the Global Optimization with First-Principles Energy Expressions (GOFEE) algorithm, which uses an on-the-fly trained Gaussian Process Regression (GPR) surrogate potential.[1] While optimizations with GOFEE reliably yield the thermodynamically most stable structure, the structure of experimental samples is governed by the kinetics of their preparation, which can stabilize higher-energy phases that a purely energetic search does not identify. This limitation can be addressed by including the match to experimental data in a combined optimization approach. Such an approach has already been used successfully to identify nanoparticle and bulk crystal structures from Pair Distribution Function (PDF) data.[2] We present a method that extends the combined optimization approach to surfaces by using Surface X-Ray Diffraction (SXRD) data. SXRD probes the atomic structure of a surface by measuring Crystal Truncation Rods (CTRs), the continuous rods of intensity that arise along the surface normal when a crystal is truncated. In a structure search, the match to the experimental CTRs and the DFT energy are mixed into a combined fitness value, which is minimized globally. The implementation builds on the Atomic Simulation Environment (ASE), making it compatible with a wide range of electronic structure codes and with global optimization algorithms other than GOFEE, which is employed here.
The combined global optimization approach is validated on a simulated Pt surface and then applied to an experimental dataset of a graphene monolayer adsorbed on Ni(111). It identifies two co-existing surface phases and reaches agreement with the experimental CTR data comparable to the existing structural model.[3] This establishes the combined global optimization as a versatile tool for complex surface characterization problems.

References

[1] M. K. Bisbo, B. Hammer, Phys. Rev. Lett. 2020, 124, 086102.
[2] M. Kløve, S. Sommer, B. B. Iversen, B. Hammer, W. Dononelli, Adv. Mater. 2023, 35, 2208220.
[3] R. Rameshan, V. Vonk, D. Franz, et al., Sci. Rep. 2018, 8, 2662.

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Complex-variable equation-of-motion coupled-cluster variants with triples corrections for the description of electronic decay processes

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Electronic resonances are metastable unbound states that govern many electron-driven decay processes and play a central role in modern spectroscopy. Their theoretical description remains challenging because they are embedded in the electronic continuum. Non-Hermitian methods combined with complex-variable techniques, such as complex scaling (CS) and complex basis functions (CBF), provide an efficient framework to study this metastable states by analytically continuing the Hamiltonian into the complex plane [1,2]. Accurate treatment of electron correlation is equally important, making the equation-of-motion coupled-cluster (EOM-CC) formalism a particularly suitable approach. In this contribution, we present novel complex-variable EOM-CC methods with triples corrections [3,4] and extend them to the description of electronic resonances. The combination of EOM-CC methods with the CBF approach enables the inclusion of triples contributions in the treatment of a broad range of metastable states. The non-Hermitian SCF implementation was carried out within CFOUR [5], while the EOM-CC developments were implemented in QCumbre [6]. The obtained results allow us to assess the importance of the inclusion of triples in the study of electronic decay processes. Furthermore, these methods improve the description of previously studied metastable states such as the core-ionized states present in double Auger decay or temporary anions resulting from the electron attachment to Rydberg states of organic molecules like CO and N2, where the triples contribution play a crucial role.

References

1. N. Moiseyev, Non-Hermitian Quantum Mechanics, Cambridge, University Press, Cambridge, UK, (2011)
2. C. W. McCurdy, and T. N. Rescigno, PRL, 41, 1364 (1978).
3. M.-P. Kitsaras, et. al, JCP, 160, 094112 (2024)M.-P. Kitsaras, et. al, JCP, 160, 094112 (2024)
4. M.-P. Kitsaras, F. Hampe, L. Reimund, and S. Stopkowicz, JCTC, 21, 10177 (2025)M.-P. Kitsaras, et. al, JCTC, 21, 10177 (2025)
5. D. A. Matthews, L. Cheng, M. E. Harding, F. Lipparini, S. Stopkowicz, T.-C. Jagau, P.G. Szalay, J. Gauss, J. F. Stanton, JCP, 152, 214108 (2020)D. A. Matthews, et. al, JCP, 152, 214108 (2020)
6. F. Hampe, S. Stopkowicz, N. Groß, M.-P. Kitsaras, L. Grazioli, S. Blaschke, L. Monzel, U. P. Yergün, C.-M. Röper, QCUMBRE, https://www.qcumbre.org/ F. Hampe, et. al, QCUMBRE, https://www.qcumbre.org/

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Finite Nuclear Size Effects in Parity-Violating Properties of Chiral Molecules

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The weak interaction is one of the fundamental interactions in nature alongside gravity, electromagnetism and the strong interaction. Despite being rather weak in the low-energy regime, it is responsible for phenomena such as beta-decay of nuclei. Furthermore, parity is not conserved in the weak interaction, which means that this interaction behaves differently for left- and right-handed (e.g. chiral) systems. Parity violation (PV) lifts the degeneracy of enantiomers, resulting in a small energy difference between them. The PV contribution depends very sensitively on the electronic wavefunction at the nucleus. Therefore, especially for systems containing heavy elements, the nucleus should be treated as a finite nuclear density distribution, described, for example, by a Gaussian nuclear model, rather than as a point like object. The poster will provide insight into the effects of introducing a finite nucleus on the corresponding behavior of the parity-violating contribution of the weak interaction. We will discuss the identification of PV effects that are sensitive to such nuclear-size effects. Such effects could potentially be investigated in processes involving changes in the nuclear radius, including decays of heavy nuclei. Furthermore, they may be relevant to high-precision laser spectroscopy for investigating nuclear properties across the nuclear chart. This opens up further possibilities for predicting molecular properties based on changes in nuclear radii.

R. Berger, J. Stohner, WIREs Comput Mol Sci 2019, 9, e1396.
L. Visscher, K.G. Dyall, At. Data Nucl. Data Tables, 1997, 67, 207-224.
D. Andrae, Chapter 4 in Relativistic Electronic Structure Theory - Fundamentals, (Ed.: P. Schwerdtfeger), Elsevier, 2002.
E. van Lenthe, E. J. Baerends, J. G. Snijders, J. Chem. Phys. 1993, 99, 4597-4610.
C. van Wüllen, J. Chem. Phys. 1998, 109, 392-399.

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Elastic Properties of Organic Crystals: A Periodic DFT Study

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Understanding and controlling of mechanical properties such as elasticity and plasticity are crucial for the application of molecular organic crystals as functional materials in the fields of electronics, pharmaceuticals and medicinal chemistry. The flexibility of the crystal is influenced by numerous factors, primarily molecular structure, intermolecular interactions and crystal packing. This research focuses on a series of halogenated benzene-based crystals containing different types of linkers (e.g. ester, azo, etc.), in which small molecular modifications resulted in distinct mechanical properties.2 The impact of halogen substituents and polymorphism on mechanical response is tested through a series of experiments such as three point bending tests and Raman spectroscopy of bent crystals. These results are compared with an extensive computational analysis based on periodic DFT methods performed in CRYSTAL23. Young's moduli were calculated on the optimized crystal systems.3 Interaction energies of chosen dimers within the crystal structure were computed on several different levels of theory and compared with directional elastic moduli. The impact of small deformations within the unit cell upon bending of the crystal is investigated in detail by critical points QTAIM analysis of non-bonded interactions between molecules. Theoretical Raman spectra have been calculated and compared with experimental Raman spectra of undeformed and deformed crystals. A detailed theoretical overview such as this could improve existing models describing elastic bending of crystalline organic systems for the purpose of understanding and predicting the mechanical properties of molecular crystals.

1. D. P. Karothu, J. Mahmoud Halabi, E. Ahmed, R. Ferreira, P. R. Spackman, M. A. Spackman, P. Naumov, Angew. Chem. Int. Ed. 61(10) (2022) 1433-7851.
2. S. Saha, G. R. Desiraju, Chem. Commun., 54 (2018) 6348-6351.
3. E. Kiely, R. Zwane, R. Fox, A. M. Reilly, S. Guerin, CrystEngComm, 23(34) (2021) 5697–5710.

P44

Automated Corrections for Materials Design of Ionic Systems: AFLOW-CCE

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Materials databases such as AFLOW [1] leverage ab initio calculations for autonomous materials design. The predictive power critically relies on accurate formation enthalpies — quantifying the thermodynamic stability of a system. For ionic materials such as oxides and nitrides, standard DFT leads to errors of several hundred meV/atom [2,3]. We have recently developed the "coordination corrected enthalpies" (CCE) method yielding highly accurate room temperature formation enthalpies with mean absolute errors down to 27 meV/atom [3]. Here, we introduce AFLOW-CCE [4,5] — our implementation of CCE into the AFLOW framework. It provides a tool where users can input a structure file and receive the CCE corrections, or even the CCE formation enthalpies if pre-calculated LDA, PBE or SCAN values are provided. The implementation features a command line tool, a web interface, and a Python environment.

[1] S. Curtarolo et al., Comput. Mater. Sci. 2012, 58, 218.
[2] V. Stevanović et al., Phys. Rev. B 2012, 85, 115104.
[3] R. Friedrich et al., npj Comput. Mater. 2019, 5, 59.
[4] R. Friedrich et al., Phys. Rev. Mater. 2021, 5, 043803.
[5] R. Friedrich & S. Curtarolo, J. Chem. Phys. 2024, 160, 042501.

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Excited-state Dynamics of a Photoswitchable Vitamin D Receptor Agonist

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The vitamin D receptor (VDR) plays a key role in regulating epidermal homeostasis and inflammation, making its activation a primary therapeutic strategy for treating psoriasis [1]. However, systemic administration can cause adverse calcium metabolism issues, highlighting the need for targeted activation [2, 3]. Photopharmacology addresses this issue by using light-responsive molecular photoswitches, such as S-PhotoVDRM, to control drug activity in a reversible manner with high spatial and temporal resolution [4]. Although azobenzene derivatives have been widely studied [5], the atomistic mechanisms of these photoswitchable ligands in biological environments are not well understood. To address this issue, we have investigated the azobenzene-based vitamin D receptor (VDR) photoswitch, PhotoVDRM, using a multiscale computational approach that combines classical molecular dynamics and QM/MM calculations with nonadiabatic dynamics [5]. Although the trans isomer is thermodynamically favoured, the cis form forms specific interactions with key VDR activation residues. Both isomers undergo efficient photoisomerisation, with the cis isomer exhibiting faster dynamics and higher conversion. Importantly, binding-pocket confinement forces the reaction to proceed predominantly via a pedal-like mechanism characterised by internal azo motion and constrained phenyl arms, with the cis isomer also utilising a rotational pathway.

Literature:
[1] Penna, G. J.Bone Miner. Res. 2007, 22, V69–V73.
[2] Martin, A. J. Dermatol. Clin. 2025, 43, 1–9.
[3] Jin, Z. Bioorg. Med. Chem. 2025, 119, 118067.
[4] Kobauri, P. Angew. Chem., Int. Ed. 2023, 62, e202300681.
[5] Jerca, F. A. Nat. Rev. Chem. 2022, 6, 51–69.
[6] Rovira, X. ACS Cent. Sci. 2025, 11, 2340–2352.
[7] Warshel A, J. Mol. Biol. 1976, 103, 227-249.
[8] Field M. J, J. Comput. Chem. 1990, 11, 700.

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Mechanism-Guided Design of Metal-Based Molecular Solar-Thermal Batteries

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Molecular solar-thermal (MOST) systems store sunlight as metastable chemical energy and release it on demand as heat. Metal-based switches are attractive—the (fulvalene)$Ru_2(CO)_4$ system remains the only organometallic photoswitch that both harvests and thermally releases solar energy—yet efficient candidates are scarce, because visible-light absorption, high energy density, and long-lived but releasable photoproducts are intrinsically coupled and often antagonistic. Here I present a mechanism-guided framework that turns these constraints into predictive design rules, establishing two design gates that every viable photoswitch must pass. I first resolve why one of the field's earliest proposals—the earth-abundant Fe analogue advanced to replace precious Ru—fails [1]. The decisive factor is not the magnitude of spin-orbit coupling, but the Marcus regime governing intersystem crossing (ISC). State-of-the-art multireference (XMS-CASPT2) and TD-DFT calculations, combined with Marcus theory, show that access to the reactive triplet manifold is controlled by the singlet-triplet crossing seam. Ru lies in the crossover regime, where ISC is barrierless and ultrafast, whereas Fe falls in the inverted regime, trapped behind a high barrier despite strong spin-orbit coupling. This spin-gated ISC defines the Photo Gate [1,2]. A complementary Storage-Stability Gate governs the charged state: raising stored energy density erodes the thermal back-reaction barrier, so energy density and shelf life trade off, and metal and ligand identity set where a system sits in this window [2]. Screening 21 bimetallic combinations across both gates collapses the chemical space to two qualifying systems: the Ru_2 benchmark and (Fv)OsW$(CO)_5$, a newly predicted heterobimetallic switch that halves the precious-metal loading of the benchmark [2]. These mechanistic design rules also provide a foundation for ligand engineering to further optimize visible-light absorption, energy storage, and release kinetics in next-generation MOST systems.

References

[1] G. Ganguly, L. González, *J. Am. Chem. Soc.* **147**, 41855–41866 (2025).
[2] G. Ganguly, L. González, *ChemRxiv* (2026), DOI: 10.26434/chemrxiv.15001174.

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What Does It Take to Automate Multireference Electronic Spectroscopy for Large-Scale Astrochemical Explorations?

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Multireference methods are essential for describing diatomic molecules in highly excited states and bond-stretching regions, but they are rarely black-box methods. For a single molecule, electronic states and active spaces are typically chosen manually on a case-by-case basis. For large-scale systematic studies, however, these choices become a central bottleneck. This raises a methodological question: can multireference electronic spectra calculations be automated sufficiently to generate reliable reference data at scale?
This question is motivated by the search for molecular carriers of diffuse interstellar bands (DIBs), one of the longest-standing open problems in astrophysics. While C_{60}^+ is the only DIB carrier firmly identified so far [1], modern high-resolution observations suggest that small molecules, including diatomics, may contribute to some DIB systems [2]. For many plausible candidates, accurate experimental or theoretical gas-phase reference spectra are not yet available. A systematic computational database could therefore help guide targeted laboratory and astronomical studies toward the most promising regions of the chemical search space and, more generally, considerably accelerate the assignment of astrophysical signals.
Here, we present the Electronic Spectroscopy Database for Astrochemical Studies (ESPECDAS), an automated workflow and database ecosystem for calculating electronic spectra of astrochemically relevant diatomic molecules. The electronic structure workflow aims to cover excitation energies up to ~ 7 eV. Molecular states are generated from atomic terms using Wigner–Witmer rules and tabulated atomic energy levels. AVAS-CASCI scans construct and refine active spaces and select relevant state groups. State-averaged CASSCF calculations provide reference functions for final state-specific X2C-MRCI+Q calculations [3], while orbital occupations, transition-dipole continuity, avoided crossings, smoothness, and state degeneracies monitor state character and curve quality. The simulated rovibronically resolved electronic spectra are collected in a database and provided via our web app and Python API, enabling the search by spectral (rovibronic) patterns [4].

References

[1] Campbell et al., *Nature* **523**, 322 (2015).
[2] Ebenbichler, Ončák, Przybilla et al., *Astron. Astrophys.* **695**, A212 (2025).
[3] Werner, Knowles, Knizia et al., *WIREs Comput. Mol. Sci.* **2**, 242 (2012).
[4] Gatt, Ončák et al., ESPECDAS Web App, Proof-of-concept, `https://explore-research.uibk.ac.a`

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Modelling the Impact of Structural and Electronic Disorder Effects on the Charge Transport Properties of Merocyanines

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Merocyanines are push-pull π-conjugated chromophores, consisting of electronic donor and acceptor subunits. Despite their well-known self-assembly and opto-electronic characteristics, which make them optimal molecular platforms for organic electronics and sensing applications, their structure-function relationships remain elusive. We tackle this challenge by modelling, through a multi-scale approach, their structural and charge transport properties by assessing the interplay between intra- and inter-molecular interactions, as well as structural, energetic, and thermal disorder effects.
At first, we modelled the complex thermally-activated solid state phase transitions of merocyanines via extended classical MD simulations, based on careful re-parametrization of the atomistic force field, leading to a perfect match with the XRD experimental data. Insights into the polymorph structural characterization as well as the short-/long-range order at solid state, are obtained by computing a set of order parameters (e.g., nematic order parameter, bond orientational order parameters).[1]
Finally, we model the charge (hole) transport properties of merocyanine single crystals by adopting semi-classical electron transfer theory and kinetic Monte-Carlo simulations.[2] The effect of static disorder is evaluated by computing the site energy distributions considering both electrostatic and polarization effects. Thermal effects are modeled via MD simulations, assessing the impact of molecular motions (phonons) on the charge transfer coupling integrals. While energetic disorder is detrimental for the hole mobility, dynamic effects can open new transport channels, thus impacting the topology of the charge transport. Disorder effects in merocyanines are of the same order of magnitude as those computed for oligoacenes and thioacenes.[3]
Our in-depth computational investigation connects structural and charge transport properties at various scale lengths, providing informative insights into solid state phase transitions, polymorphism as well as a comprehensive understanding of disorder effects on the charge transport properties. Such aspects can be considered for a guided molecular design of next-generation organic opto-electronic semiconductors.

[1] S. Geller, K. Meerholz, D. Fazzi (in preparation).
[2] N. Gildemeister, G. Ricci, L. Böhner, J.M. Neudörfl, D. Hertel, F. Würthner, F. Negri, K. Meerholz, D. Fazzi, J. Mater. Chem. C 2021, 9, 10851-10864.
[3] N. Gildemeister, S. Geller, R. Herzhoff, F. Negri, K. Meerholz, D. Fazzi, Mater. Adv. 2024, 5, 8475-8489.

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Multireference Studies of Strongly Correlated Transition Metal Complexes: When DFT Fails at the Geometry

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Density functional theory remains the workhorse for 3d transition metal chemistry, but for strongly correlated complexes the choice of functional does not merely shift relative energies, it changes the electronic state, and with it the potential energy surface. A structure optimized on a qualitatively wrong state cannot be repaired by a better single-point. Multireference methods are the natural remedy, yet inherit the problem the moment they are applied to a naive DFT geometry, and which structure to trust is rarely asked. The cobalt CO2-reduction catalyst [(Hbbpya)Co]2+ (Hbbpya = NH-bridged bisbipyridine) illustrates the difficulty, GGA and hybrid functionals predict Co(I)-ligand-radical and Co(II)-ligand-diradical states respectively, but calculated EPR parameters agree with experiment only for the former.[1] CASSCF calculations on either DFT-optimized geometry nevertheless converge exclusively to the Co(II) solution, irrespective of active space, orbital treatment, or state selection. The Co(I) state emerges only upon displacement along the metal-ligand coordinate. The practical consequence is diagnostic rather than prescriptive: when CASSCF cannot find a state that experiment demands, the geometry is at least as plausible a suspect as the active space. The iron-oxo complex [(Hbbpya)FeO]+ poses the same question in a considerably less settled form.[2] Several plausible electronic structures — an Fe(IV)-oxo, an antiferromagnetically coupled Fe(III)-oxyl, and a more reduced iron center coupled to both an oxyl and a ligand radical — lie close in energy. Which one a DFT calculation converges to depends not only on the amount of Hartree-Fock exchange admixed but also on the initial orbital guess. How these solutions relate to the species actually observed remains unresolved. We present state-averaged large-active-space CASSCF/EN-NEVPT2 calculations along a relaxed Fe–O coordinate, asking which of these descriptions survive a multireference treatment and whether a two-state situation analogous to that proposed for TauD is operative.[3] All multireference calculations were carried out with the HUMMR program package, employing EN-NEVPT2 and spin-adapted heat-bath CI.[4-6]

[1] Bera et al., ACIE 2025, e202503705.
[2] https://doi.org/10.26434/chemrxiv.10001822/v1
[3] Ye et al. PNAS 2011, 108, 1228.
[4] Ugandi et al. JCTC 2026, 22, 7164.
[5] Ugandi et al., JCC 2023, 44, 2374.
[6] https://hu.berlin/hummr

P50

Piezochromism of tridentate platinum complexes for adaptive materials

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Transition metal complexes are known for their triplet emission and many possible applications including catalysis, optical sensing, and OLEDs. Previous work introduced platinum and palladium complexes whose ligands could be designed to define their crystal structure [1]. This way, the metal-metal interactions and consequently the photophysical properties can be tuned. The complexes show reversible change in their phosphorescence through pressure variation [2]. Because of their responsive behaviour, transition metal complexes are potential components for adaptive materials. Here, we investigate single crystals of new phosphorescent tridentate platinum complexes under pressure from 1 bar to 16 GPa using (time-dependent) density functional theory. In the single crystal, the complexes form dimers of which metal-metal distances decrease by 0.5 Å from 1 bar to 16 GPa. Increasing pressure leads to a red-shift of absorption and phosphorescence spectra. The calculations yield a shift of about -40 meV/GPa for the lowest energy peak of the absorption and indicate delocalized electronic excitations. Furthermore, they provide insight into the influence of different ligands on the piezochromism.

[1] T. Theiss et al., J. Am. Chem. Soc. 145 (2023), 3937.
[2] P. Steeger et al., Nano Lett. 25 (2025), 2628.

P51

Absorption and Response Properties of Molecules Coupled to Optical Quantum Cavities with Coupled Cluster Theory

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In quantum cavities, strong light-matter coupling gives rise to polaritons, hybrid light-matter states with properties distinct from those of isolated molecules. This enables the non-invasive control of quantum phenomena, molecular energy transfer, and chemical reactivity [1]. The accurate description of strong light-matter coupling still presents challenges for electronic structure theory, as it requires a quantum treatment not only of the matter, but also of the electromagnetic field and the light-matter interaction. [2]
In this work, we extend coupled cluster-based approaches to study polaritons in opical cavities. The spin-flip variant of the equation-of-motion coupled cluster theory (QED–EOM–CC) [3] is presented and applied to polaritonic states arising from biradicals. It is demonstrated how the new states alter the topology of excited-state potential energy surfaces, thereby enabling control over photochemical processes. We also exend the linear-response coupled cluster theory to the polaritonic regime (QED–LR–CC) for computing static and dynamic dipole-dipole polarizabilities and explore how resonant and nonresonant effects can alter the pole structure, the magnitude, and the anisotropy of the polarizability tensor. Together, these developments enable highly accurate ab initio descriptions of optical excitations and response properties of molecules in optical quantum cavities, providing reliable benchmarks for future experimental or theoretical developments.

[1] B. Xiang, W. Xiong, Chem. Rev. 2024, 124, 5, 2512-2552
[2] N. Bauman et al., J. Chem. Theory Comput., 2025, 21, 20, 10035-10067
[3] L. Monzel, S. Stopkowicz, J. Phys. Chem. A, 2024, 128, 44, 9572-9586

P52

On the Role of Substituent Arrangements versus Carbon Backbone towards Chirality Observables

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The parameters of optical activity in chiral molecules specific rotation and optical rotatory dispersion (ORD) are pseudoscalar in nature and have been of great interest, as they can be potentially linked to the absolute configuration of the molecule. In this contribution we analyse the role of the arrangement of the substituents versus the influence of the carbon skeleton in different organic chiral molecules towards the sign and magnitude of the observables. Inclusion of weak interactions into the quantum mechanical study leads to a non-negligible parity violating energy difference between the two enantiomers (PVED) [1]. This energy difference can also be viewed as a pseudoscalar observable. We explore if the established arguments for optical activity also suffice for predicting the sign and magnitude of PVED.

[1] R. Berger and J. Stohner, WIREs Comput Mol Sci. (2019), 9, e1396

P53

Understanding Resonance-Enabled Polaritonic Control of Hydrogen Transfer Dynamics and Relaxation

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Molecular vibro-polaritons are currently discussed as a possible tool to modify ground-state reactivity. They arise when vibrational transitions couple strongly to an electromagnetic field, e.g in a cavity. We present numerical open-system quantum dynamics of thioacetylacetone (TAA) undergoing hydrogen transfer, coupled to a cavity mode and a bath. This model system, previously studied in [Fischer and Saalfrank Phys. Chem. Chem. Phys. 25 (2023) 11771], comprises an asymmetric potential energy surface (PES). By accounting for vibrational energy relaxation (VER), we provide reaction rates and corresponding photon-frequency dependent rate-profiles, exhibiting both, suppression and acceleration, depending on microscopic details. To understand the numerical results, analytic expressions for the rate-profiles are provided by utilizing the Jaynes-Cummings model, while treating VER perturbatively. We attribute our findings to the formation of polariton states, representing the light matter hybridization when the light mode is in resonance with a vibrational transition. Our results indicate that a full quantum treatment of both the cavity mode and the bath is required.

P54

Ab initio calculation of energy-dependent Hubbard parameters for DFT+U using real-time TDDFT

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DFT+U [1], the density functional theory (DFT) augmented with parametrized on-site interactions, is known as a cost-efficient approach to calculations of strongly correlated materials. It has recently gained renewed interest in the application to high-throughput calculations. In such applications, ab initio calculation of the screened interaction parameters, or the Hubbard parameters, is an indispensable component. A number of techniques have been developed, including constrained random-phase approximation (cRPA) [2], linear-response approaches [3,4], and a pseudohybrid Hubbard density functional ACBN0 [5]. However, only a few of those techniques, such as cRPA [2], derive these parameters as functions of energy. The effective interactions represented by the Hubbard parameters depend on the quasiparticle energy due to the screening effects. A recent study has shown that energy-independent parametrization gives qualitatively wrong results in some materials [6]. We propose a new linear-response-based calculation scheme for energy-dependent Hubbard parameters [7]. Our scheme extends the minimum-tracking linear-response method [4] to compute Hubbard parameters as a dynamical response. One strength of the linear-response approach is the inclusion of the exchange-correlation (XC) effects, yielding dynamical Hubbard parameters adjusted for a given XC functional. Our new scheme is implemented in the k-point sampling real-time time-dependent DFT (RT-TDDFT) program in CP2K [8]. We discuss the properties of the minimum-tracking linear-response method [4] in comparison to ACBN0 [5]. While neither was found to clearly outperform the other in terms of accuracy in static property calculations, each has a distinct dynamical application depending on its theoretical formulation.

[1] V. I. Anisimov, J. Zaanen, and O. K. Andersen, Phys. Rev. B 1991, 44, 943
[2] F. Aryasetiawan, et al. Phys. Rev. B 2004, 70, 195104
[3] M. Cococcioni and S. de Gironcoli, Phys. Rev. B 2005, 71, 035105
[4] G. Moynihan, G. Teobaldi, and D. D. O'Regan, arXiv 2017, arXiv:1704.08076
[5] L. A. Agapito et al., Phys. Rev. X 2015, 5, 011006
[6] Y. Wu, M. Caserta, T. Chiarotti, and N. Marzari, arXiv 2025, arXiv:2511.23181
[7] K. Hanasaki and S. Luber, arXiv 2026, arXiv:2604.06927
[8] K. Hanasaki and S. Luber, J. Chem. Theor. Comput. 2025, 21, 1879

P55

Exploiting the Softening of Universal MLIPs for Data-Efficient Material-Specific Models

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Machine-learned interatomic potentials (MLIPs) trained on large, general datasets promise near-ab initio accuracy at a cost approaching that of emperical force fields. Whether they deliver this for material-specific observables, in particular rare and reactive events, is far less clear.
We assess five leading universal MLIP frameworks (MACE, SevenNet, GRACE, MatterSim, ORB) on seven chemically diverse systems, covering equivariant and invariant as well as conservative and non-conservative architectures. Strong performance on established benchmarks does not translate into reliable recovery of the target observables: in zero-shot mode the models deviate from the ab initio reference in an architecture-dependent way and fail for transport and high-barrier processes. Fine-tuning on system-specific data removes these deviations and brings all architectures to a common level, improving force predictions by factors of 10–20.
The remaining bottleneck is therefore the reference data itself. We use universal MLIPs as configuration-space generators: long MLIP trajectories are sub-sampled, relabeled with DFT, and used to train material-specific models. This lets us exploit the systematic softening of universal potential energy surfaces - underestimated barriers let the sampling MD reach high-energy, off-equilibrium regions that equilibrium AIMD rarely visits, and after DFT relabeling these structures become exactly the reference data the model was missing.
The conventional route of sub-sampling AIMD provides the yardstick: ten configurations are not enough, 200 succeed only in favorable cases, and 2,000 are a robust default, with moderately dense sub-sampling shortening the required trajectory tenfold. Models built from universal-MLIP-generated structures reach comparable accuracy without the long AIMD run.

P56

Machine-Learning Potentials for redox chemistry in solution

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Over the past two decades, Machine-Learning Potentials (MLPs) have become established tools for atomistic simulations of large condensed-phase systems. Their key advantage is the ability to represent the potential energy surface with near–quantum mechanical accuracy at a fraction of the computational cost.[1] MLPs can be classified into different generations based on the interaction range they capture. The most widely used are Second-Generation MLPs (2G-MLPs), which rely on local structural information to model short-range interatomic interactions. Because this approach neglects long-range electrostatics, Third-Generation MLPs (3G-MLPs) were introduced which incorporate electrostatic contributions via environment-dependent atomic charges. Since both, 2G and 3G MLPs, rely on local information only, their accuracy for systems in which non-local charge transfer is important is limited.[2] A prominent example is distinguishing the oxidation states of transition-metal ions, such as ferrous and ferric ions in aqueous solution, where the near-sightedness of local MLPs fails to account for distant counter-ions.[3] Here, we show that this limitation can be overcome by Fourth-Generation High-Dimensional Neural Network Potentials (4G-HDNNPs),[4] which employ atomic charges obtained from a charge-equilibration scheme as global input features. We find that the model correctly assigns the iron oxidation state in accordance with the total number of chloride ions in the system, irrespective of their positions. These findings show that physical insight into the system is essential for constructing reliable MLPs for simulations of solution-phase redox chemistry involving multiple oxidation states.[3]

References

[1] Behler, J., Csányi, G., "Machine learning potentials for extended systems: a perspective", Eur. Phys. J. B 94, 142 (2021).
[2] Behler, J., "Four generations of high-dimensional neural network potentials," Chem. Rev. 121, 10037 (2021).
[3] E. Kocer, R. El Haouari, C. Dellago, and J. Behler, "Machine learning potentials for redox chemistry in solution," arXiv:2410.03299 (2024).
[4] T. W. Ko, J. A. Finkler, S. Goedecker, and J. Behler, "A fourth-generation high-dimensional neural network potential with accurate electrostatics including non-local charge transfer," Nat. Commun. 12, 398 (2021).

P57

Instanton theory for reaction rates far from equilibrium

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Rate theories are central to understanding rare, reactive events across chemistry and physics. Workhorse approaches, such as transition-state theory, Kramers theory and their semiclassical relatives, rely on the assumption that the system remains close to thermal equilibrium throughout the reactive process. Yet many systems of contemporary interest, including active and biological matter, driven soft materials, and chemical systems under external fields or mechanical load, are continuously driven and thus operate far from equilibrium. For these nonequilibrium processes, equilibrium rate theories can break down.

In this work, we present a method for determining the rate constants and mechanisms of rare events in systems driven arbitrarily far from equilibrium. Far from equilibrium, reaction rates are no longer determined by the familiar free-energy landscape alone but depend on the dynamical details of the reactive trajectories themselves. We therefore formulate our approach as an asymptotic approximation to the stochastic path integral in the weak-noise limit, where the rate is governed by the optimal transition pathway, or instanton, connecting reactant and product states. Unlike in equilibrium, forward and backward paths are no longer related by time-reversal symmetry because detailed balance is broken, leading to qualitatively different reaction mechanisms and rates for forward and backward processes.

We demonstrate that our method consistently extends Kramers–Langer theory beyond equilibrium, with instanton rates showing excellent agreement with numerically exact results across a broad range of model systems for which a naive equilibrium treatment fails. Through applications to phase transitions in complex systems such as active-matter field theories and enzyme-catalyzed reaction–diffusion systems, we highlight how nonequilibrium driving reshapes reactive pathways and alters stability in systems of experimental relevance. Together, these results establish our nonequilibrium instanton rate approach as a practical and physically interpretable tool for rare-event kinetics beyond equilibrium, with natural applications to driven chemical, biological, and soft-matter systems.

References

[1] E. R. Heller, D. T. Limmer, Evaluation of transition rates from nonequilibrium instantons, *Phys. Rev. Res.* **6**, 043110 (2024).

[2] E. R. Heller, D. T. Limmer, Pattern stability in reaction–diffusion systems depends on path entropy, *arXiv:2603.11192* (2026).

P58

Bridging the Gap: A Benchmark Study of Hyperfine Coupling Constants from EPR Spectroscopy to Qubit Decoherence Times

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The accurate prediction of hyperfine coupling constants (HFCCs) bridges a gap from interpreting electron paramagnetic resonance (EPR) spectra [1] to the rational design of stable molecular qubits [2]. However, calculating HFCCs for transition metal complexes represents a significant challenge, largely because these systems are prone to spin contamination and multi-configurational character. To establish a reliable and predictive methodology for both the simulation of EPR spectra and qubit engineering, we extract learnings from a comprehensive benchmark of 40 density functionals, ranging from generalized gradient approximations (GGAs) to spin-component-scaled double hybrids (SCS-DHs), on 34 complexes using the exact two-component (x2c) Hamiltonian. Utilizing the unrestricted Hartree–Fock (UHF) $\langle S^2 \rangle$ expectation value as a qualitative measure of spin contamination [4], which we validate as a reliable indicator of the underlying multi-reference character via explicit CASSCF calculations, we demonstrate that functional performance is strongly correlated with this multi-configurational nature. While performance degrades across all functional classes with increasing UHF $\langle S^2 \rangle$, double hybrid functionals excel for systems with a "spin clean" single-determinant reference. Because an ideal molecular qubit inherently operates in this strictly spin-pure regime, our findings directly establish a highly reliable, first-principles methodology for their theoretical construction. Furthermore, these precisely determined HFCCs provide the essential parameters needed to model spin-phonon coupling and spin dynamics, enabling the calculation of coherence times (T_1/T_2) [3]. For accurate HFCC predictions within this spin-pure regime, we recommend DSD or double hybrid functionals, reserving range-separated hybrids (RS-H) for unavoidably contaminated scenarios if multi-reference methods are unavailable. Finally, to address the breakdown of DFT in highly contaminated regimes, we extend our methodology toward multi-reference HFCC calculations utilizing the RASSCF/RASSI approach in OpenMolcas.

References

[1] M. Munzarová, M. Kaupp, *J. Phys. Chem. A* **103**, 9966 (1999).
[2] J. Cardona et al., *Chem. Sci.* **16**, 11291 (2025).
[3] J. K. Staab, N. F. Chilton, *J. Chem. Theory Comput.* **18**, 6588 (2022).
[4] J. Gräfenstein, D. Cremer, *Mol. Phys.* **99**, 981 (2001).

P59

Towards the construction of a global potential energy surface for H4+ through isotropic direction sampling

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The H3+ ion is the most abundant polyatomic molecule in the universe and is formed through the reaction H2 + H2+ → H3+ + H. Any quantum dynamics calculation involving this highly efficient and exothermic reaction requires a suitable potential energy surface (PES) describing the H4+ system globally under consideration of its permutational symmetry. This work forms the basis for the construction of such a PES by locating all relevant stationary points and determining further reference geometries by analyzing the minimum energy path. The data necessary for constructing the PES is then acquired by multireference configuration interaction calculations through isotropic direction sampling starting from each of the determined reference geometries. This method is aimed at constructing a PES particularly accurate around geometries relevant for the reaction.

P60

Vibrationally Nonadiabatic Dynamics of (Floppy) Molecules in Liquid Environments

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Studying molecules in liquid environments is a powerful but challenging way to probe single-molecule properties and solvent-solute interactions. Sophisticated gas-phase simulations of small molecules are common, yet studies of the effect of liquid environments on dissolved molecules are scarce. Interesting solvent-solute systems include molecules dissolved in superfluid Helium, where the solvent is assumed to interact weakly with the solute, and molecules dissolved in cold liquid Argon to study relaxation processes. In both regimes an accurate quantum mechanical treatment of the solute is required, whereas the solvent can oftentimes be described classically [1]. To date, such simulations remain challenging due to the many degrees of freedom, the floppiness of solute and solvent and the required accuracy of the interaction potentials.
This work is split into two parts: in the first part, a fully quantum-mechanical study of the interaction of Hydronium with Helium is presented. The solute-solvent interactions are described through a vibrationally diabatic representation, obtained via an electronically adiabatic interaction potential based on an accurate potential energy surface [2]. Ro-vibrational energy levels of the H3O+-He system are calculated and analyzed, revealing strong vibration-rotation coupling. In the second part, we investigate the dynamics of HCl and DCl in liquid Argon, simulating the relaxation of vibrationally excited HCl or DCl into the vibrational ground state. The Argon is treated classically and the HCl-subsystem quantum-mechanically, with the interaction again modelled using a nonadiabatic representation based on an accurate electronic potential energy surface [3]. The mixed quantum-classical dynamics are based on the Mapping Approach to Surface Hopping [4], and the resulting relaxation rates are compared to experimental data. In the future, the methodology and interaction potentials will be used to further understand the dynamics of H3O+ in bulk Helium, as well as the properties of dissolved or confined molecules such as D2O in H2O.

References
[1] Oxtoby D. W., Ann. Rev. Phys. Chem., 32, 77-101, (1981)
[2] Schran C., Uhl, F., Behler, J., Marx, D., J. Chem. Phys., 148, 102310, (2018)
[3] Sadeghi, S., Nemati-Kande, E., J. Phys. Chem., 127, 1053 (2023)
[4] Mannouch, J. R., Richardson, J. O., J. Chem. Phys., 158, 104111 (2023)

P61

Ab-initio Auger spectrum of the ultrafast dissociating $2p_{3/2}^{-1}\sigma^*$ resonance in HCl

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We present an ab-initio theoretical approach to calculate the resonant Auger spectrum in the presence of ultrafast dissociation, that follows the evolution of the molecular state all along the potential energy curve. The Auger decay rates are calculated within the one-center approximation and are shown to vary significantly with the inter-nuclear distance. A quantum-mechanical description of dissociation is effectuated by propagating the corresponding Franck–Condon factors using a quasiclassical Green-function propagator. The presented method can describe the resonant Auger spectrum for an arbitrary speed of dissociation, while throughout considering the variation of Auger rates.
The method is demonstrated by deriving the L–VV resonant Auger spectrum of the $2p_{3/2}^{-1}\sigma^*$ resonance in HCl, where the electronic Auger decay and nuclear dissociation occur on the same time-scale. The calculated shapes of Auger spectral lines agree very well with available experimental results. We show that the quantum-mechanical description of nuclear dynamics is essential to achieve this.

P62

All-Electron Machine-Learned Orbital-Free Density Functional Theory for Periodic Systems

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Orbital-free density functional theory (OF-DFT) expresses the total energy directly as a functional of the electron density, without the auxiliary orbitals used in Kohn–Sham DFT, offering more favorable scaling. Its practical accuracy, however, is limited by approximate kinetic-energy functionals. Applications to periodic materials pose a further practical challenge because they commonly rely on nonlocal pseudopotentials that act on orbitals. While a density-functional treatment of these potentials has been proposed [1], an all-electron approach avoids pseudopotentials altogether.

We present, to our knowledge, the first variational all-electron machine-learned OF-DFT framework demonstrated for three-dimensional periodic crystals. The unit-cell density is represented in an atom-centered Gaussian basis capable of describing the nuclear-cusp region. Hartree and electron–nuclear attraction energies are evaluated analytically, leaving an equivariant graph neural network to learn the combined kinetic and exchange-correlation contribution. We apply this approach to CHNO molecular crystals. Training data are generated from all-electron periodic Kohn–Sham calculations for structures drawn from a structure search at 1 GPa, extending the molecular approaches of Refs. [2,3] to crystals.

For held-out structures, the learned functional achieves a mean absolute total-energy error of 0.32 mHa atom^{-1} after variational density optimization and generalizes to unit cells larger than any in the training set. It largely preserves the Kohn–Sham structure rankings and yields a formation-enthalpy convex hull in good agreement with the reference. A single functional can also be trained jointly on QM9 molecules and periodic crystals, placing finite and extended systems on the same footing.

These results demonstrate a unified all-electron OF-DFT treatment of energies and electron densities in finite and periodic systems.

References

[1] Q. Xu et al., *Nat. Commun.* **13**, 1385 (2022).
[2] H. Zhang et al., *Nat. Comput. Sci.* **4**, 210–223 (2024).
[3] R. Remme et al., *J. Am. Chem. Soc.* **147**, 28851–28859 (2025).

P63

Computational protocol for the prediction of energy transfer rates in DNA-liposome hybrids

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In natural photosynthesis, absorbed radiation is concentrated in space through a series of highly-efficient energy transfer steps, which is achieved through the spatial confinement of sub-units by embedding these in the thylakoid membranes and protein complexes, tuning their electrostatic environment and relative alignments. A common strategy in artificial photosynthesis is to mimic these approaches. Two of the many immobilization strategies include attachment to DNA architectures or embedding them into liposome membranes. To assess the efficiency of such bio-inspired light-harvesting systems, it is highly desirable to predict their energy transfer rates computationally.
The design of a computational protocol for the prediction of energy transfer rates in such systems is challenged by the variety of properties influencing the energy transfer, including the interchromophoric distance and alignment, the transition dipole moments and the spectral overlap. These properties can generally not be obtained from a single computational method alone. Here, we present a multiscale computational protocol that combines data obtained from quantum chemical calculations, classical molecular dynamics simulations and, where applicable, experimental measurements to compute the energy transfer rates of anisotropic systems. Further, by modeling the diffusion of sub-units within the liposome membrane, the protocol captures membrane loading and concentration effects. At the case study of several DNA-liposome hybrids of varying DNA sequence lengths, we reveal the effective relationship between distance and energy transfer in these systems and elucidate the impact that not only the adjacent donor-acceptor pairs, but also the non-nearest neighbors have on the energy transfer efficiency. This protocol will aid in the design of novel energy transfer and light-harvesting systems.

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Ab-initio relativistic embedding methods for strongly correlated systems

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During the last ten years, an increasing number of compounds containing 4d-, 5d-, and 4f- elements have been investigated due to their potential technological applications in spintronics, energy harvesting, and quantum computing. These materials exhibit unique competitions between quantum-mechanical effects such as electronic correlation, wave-function topology, spin–orbit coupling, and conventional magnetic states, leading to the emergence of new phases and properties. At present, ab-initio modeling of materials containing 4d-, 5d-, and 4f- elements, while exceedingly helpful for fundamental understanding of their chemistry and physics as well as for optimizing their properties, remains very challenging for the current generation of electronic structure methods due to the need to accurately describe multiple competing effects such as electronic correlation, relativistic effects, and temperature effects. We plan to tackle the challenge of correlated relativistic materials while employing embedding methods such as self-energy embedding theory (SEET). In SEET, the large intractable problem of a strongly correlated molecule or solid is split into a series of manageable calculations. The current version of SEET uses the GW method to treat the weakly correlated orbitals considered as environment, while the system orbitals are treated with an accurate exact diagonalization (ED) solver. However, at present SEET only has a non-relativistic formulation, where both GW and ED are being executed for a non-relativistic Hamiltonian. In order to model ab-initio materials containing 4d-, 5d-, and 4f- elements, a relativist framework for GW and ED as well as the overall embedding construction has to be developed. In this work we mainly present progress on development of ED solver capable of describing relativistic effects for a small number of 4f, 4d, 5d orbitals and discuss possible validations on realistic molecular and solid-state problems.

P65

Multimer Embedding for Predictions of Polymorph Stability

N. Goncharova, A. List, J. Hoja, A.D. Boese (University of Graz/AT)

The structure of molecular crystals is determined by the interplay of thermodynamic stability and kinetic effects. This phenomenon, also called polymorphism, poses a challenge in pharmaceutical development, as the transition to a more stable crystalline form might drastically change the bioavailability of a drug.[1] To prevent an unexpected appearance of a new polymorph, the most favorable crystal arrangement must be identified beforehand, employing for example computational methods to study thermodynamic landscape of crystalline polymorphs. However, achieving the required accuracy to determine the most stable form represents a challenge, as the energy differences between similar structures are often so small, that only rigorous models can reliably rank their relative stability. For fully periodic calculations of molecular crystals utilizing hybrid density functionals and larger basis sets would represent a promising approach, however such calculations are often computationally too expensive and we therefore rely on approximations of such methods like a multimer embedding approach,[2-3] where multimer energies up to trimers, calculated at the PBE0+MBD level, are embedded into a GGA functional (PBE+MBD). In order to broaden our investigation beyond simple electronic energies, we account for the volumetric change of a unit cell with temperature by utilizing the quasi-harmonic approximation (QHA).[4-5] Therefore, the equilibrium unit cell volume and free energy corresponding to certain temperature are obtained with Murnaghan equation of state fits utilizing several unit cell volumes. This allows us to track the impact of thermodynamic conditions on the relative stabilities of polymorphs and to generate phase diagrams. Utilizing this framework we study thermodynamic properties of molecular crystals and compare them to the experimental results obtained in the COST action CA22107 (BEST-CSP).

References

[1] J. Bauer, S. Spanton, R. Henry, J. Quick, W. Dziki, W. Porter, J. Morris, Pharm. Res. 2001, 18, 859-866.
[2] J. Hoja, A. List, A. D. Boese, J. Chem. Theory Comput. 2023, 20, 357—367.
[3] A. List, A.D. Boese, J. Hoja, arXiv:2512.16877, 2025.
[4] J. Hoja, A. M. Reilly, A. Tkatchenko, WIREs Comput. Mol. Sci. 2016, 7, e1294.
[5] G. A. Dolgonos, J. Hoja, A. D. Boese, Phys. Chem. Chem. Phys. 2019, 21, 24333—24344.

P66

HB49rev and HB35revx10: New Highly Accurate Benchmark Sets for Hydrogen-Bonded Systems

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Hydrogen-bonded systems play a crucial role in biological and chemical processes, motivating the development of accurate computational methods for describing the underlying intermolecular interactions. Reliable benchmark data based on high-level methods such as coupled cluster with singles, doubles, and perturbative triples [CCSD(T)], often referred to as the "gold standard" in quantum chemistry, are essential for assessing their accuracy and computational cost.[1, 2]
Herein, we present two new benchmark sets, HB49rev and HB35revx10, based on the established HB49 set of 49 small hydrogen-bonded systems.[3] While HB49 combines CCSD(T) interaction energies with QCISD(T)-optimized geometries, both obtained at the complete-basis-set (CBS) limit through basis-set extrapolation, HB49rev consistently employs CCSD(T) for both geometry optimization and interaction energies, with extrapolation applied to energies and geometries to approach the CBS limit. Extending the benchmark beyond equilibrium structures, HB35revx10 comprises 35 systems at 10 different intermolecular distances. At each distance, the remaining structural degrees of freedom are relaxed using constrained CCSD(T)/CBS optimization, yielding relaxed potential energy curves.[4]
Together, HB49rev and HB35revx10 provide high-level reference data for hydrogen-bonded systems across equilibrium and non-equilibrium geometries close to the theoretical limit, serving as reliable benchmarks for the evaluation and development of computational methods.

References
[1] P. Morgante, R. Peverati, J. Comput. Chem., 40, 839 (2019)
[2] J. Rezac, J. Chem. Theory Comput., 16, 2355 (2020)
[3] A. D. Boese, Mol. Phys., 113, 1618 (2015)
[4] R. Guttmann, P. Hartmann, J. Hoja, E. Masumiam, A. D. Boese, to be submitted

P67

Nonadiabatic Molecular Dynamics Enables Direct Modeling of Plasmon-Driven Processes at the Single Molecule Level

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Research interest in plasmon-driven chemical reactions of functional molecules at nanoscale metals has rapidly increased in recent years in both experimental and theoretical communities. In an ideal scenario, plasmonic catalysis promises selective chemistry under mild reaction conditions ideally involving green light sources with various possible applications in heterogeneous catalysis. However, many of the recently investigated plasmonic systems are highly involved and individual mechanistic steps of the observed chemical transformations are immensely difficult to disentangle experimentally. Moreover, the complexity of the experimentally prepared systems provides a significant challenge for computational modeling, thus impeding joint experimental and theoretical studies targeting reaction mechanisms in plasmon-driven catalysis.
Recently, Wu et al. [1] introduced a numerically efficient theoretical framework to perform nonadiabatic surface hopping dynamics simulations of (relatively) large systems at the semiempirical time-dependent density functional tight-binding level. In principle, the method enables coupled electron-nuclear dynamics simulations of many-electron systems with dense excited-state manifolds, e.g., plasmonic systems, with the capability of directly modeling charge and energy flow at metal–molecule interfaces. In practice, however, the workflow has only been applied to an Au20–CO system [2] thus far, while its applicability to larger functional molecules remains unknown. In our present contribution, we therefore aim to theoretically describe the nonadiabatic dynamics of a covalently linked azobenzene molecule on a tetrahedral Au20 cluster, thereby extending our previous work on azobenzene–gold systems [3]. In accord with recent experimental results [4], we observe that the n$\pi\cdot$ state of AB rapidly decays within a few femtoseconds via coupling to the metal continuum, thus suppressing the AB photoisomerization process. On the other hand, the photoexcited metal primarily relaxes through electron-phonon coupling while retaining its excited-state population. Our findings provide direct atomistic insight into the elementary processes occurring at the AB–gold interface to assist in the future design of functional plasmonic systems.

References
[1] X. Wuet al., J. Chem. Phys. 157, 084114 (2022)
[2] X. Wuet al., Chem. Sci. 14, 4714 (2023)
[3] N. Jahn and E. Titov, ACS Phys. Chem. Au (2026)
[4] Z. Chu et al., J. Am. Chem. Soc. 141, 1949 (2019)

P68

Temperature-Dependent Collapse of Surface-Tethered PNIPAM at a Bioactive-Glass-Relevant Silica–Water Interface: A Dissipative Particle Dynamics Study

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Thermoresponsive polymer-bioactive-glass (BG) hybrids display temperature-controlled aggregation and ion release, but the conformational response of the grafted polymer layer that underlies this behavior is difficult to resolve experimentally. Here, dissipative particle dynamics (DPD) simulations are used to examine the temperature-dependent conformation of a single poly(N-isopropylacrylamide) (PNIPAM) chain (DP = 100) tethered to an amorphous silica surface, a bioactive-glass-relevant interface, in explicit water. A four-bead coarse-grained model (water, silica, PNIPAM, APTES-derived anchor) with temperature-dependent conservative repulsion parameters represents good-, near-onset-, and poor-solvent states at nominal 278, 298, and 308 K. Radius-of-gyration (Rg) probability distributions and surface-normal monomer concentration profiles quantify chain compactness and polymer-layer thickness; free-chain simulations in bulk water serve as controls that isolate intrinsic solvent-quality effects from surface-tethering effects.

The grafted chain evolves from a broad, multimodal swollen coil at 278 K (most probable Rg 31.0 Å) through a heterogeneous pre-transitional ensemble at 298 K (Rg 25.2 Å) to a sharply localized, surface-proximal globule at 308 K (Rg 10.55 Å). The polymer-rich layer thickness decreases about 4.7-fold (107 to 23 Å) while the local monomer concentration near the surface increases about 4.5-fold. Free-chain controls reach comparable dimensions in the swollen and collapsed limits, but the tethered chain is markedly more compact at 298 K, showing that surface grafting reshapes the intermediate conformational ensemble even though the ultimate collapse is governed by the temperature-dependent polymer-water interaction.

These single-chain results provide a molecularly consistent framework for interpreting the aggregation and enhanced Ca2+ release reported for PNIPAM-grafted bioactive-glass nanoparticles [1]. Partial shell retraction below the macroscopic cloud point can weaken steric stabilization, while the accompanying layer thinning and lateral retraction expose more glass surface to water, rationalizing the counter-intuitive enhancement of ion transport upon heating. The work establishes a single-chain conformational reference linking polymer collapse to colloidal stability and ion-release behavior in responsive organic-inorganic biomaterials, providing the baseline for future multi-chain brush, curved-particle, and ion-transport DPD models.

References

[1] A. Nabiyan, Z. Jin, D. S. Brauer, Soft Matter 2024, 20, 5945-5953.

P69

Quantum Electrodynamic Corrections in Molecules

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Precise calculations of systems containing heavy elements rely on a relativistic quantum chemical framework, such as the many-body Dirac equation. Beyond Coulombic interactions, there are additional higher-order relativistic corrections from quantum electrodynamics (QED), which improve the results. In this presentation, the leading order QED corrections, precisely the vacuum polarisation (VP) and electron self-energy (SE), are reported for nuclear magnetic resonance (NMR) shielding and nuclear spin-rotation (NSR) tensors of heavy-element containing diatomic molecules. Usually considered in high-precision few-electron atomic calculations, we investigate the scope of QED corrections in many-electron molecules and apply QED effective potentials [1,2] to molecular properties. We compare the QED corrections obtained from atomic-based and molecular-based frameworks [3,4], and identify the main origin of the different contributions. We constrast four-component and two-component approaches and discuss the impact of the QED effects on molecular properties.

References

[1] E. A. Uehling, Phys. Rev. 48, 55 (1935).
[2] V. V. Flambaum, J. S. M. Ginges, Phys. Rev. A 72, 052115 (2005).
[3] K. Kozioł, I. A. Aucar, K. Gaul, R. Berger, G. A. Aucar, J. Chem. Phys. 161, 064307 (2024).
[4] K. Janke, A. E. Wedenig, K. Gaul, P. Schwerdtfeger, R. Berger, J. Chem. Phys. 162, 104111 (2025).

P70

Higher Order Pair Natural Orbital-Based Coupled-Cluster Methods

A. Jiang (University of Georgia)

Local pair natural orbital coupled-cluster methods have seen much success recently, allowing for the application of "chemically accurate" approaches such as CCSD(T) to significantly larger systems. Notably, the DLPNO-CCSD(T) approach of Professor Frank Neese and coworkers has been utilized to run computations on whole proteins such as crambin (636 atoms) and insulin (789 atoms). At the CCSD(T) level of theory, local approximations have errors which are controllable to within 0.25 kcal/mol through tight parameters. Inspired by the work of Neese et al., we developed analogous models for higher orders of coupled-cluster theory through full quadruples (DLPNO-CCSDTQ). These methods allow for an accurate estimate of quadruples contributions such as (Q) and Q-(Q) on the order of 0.01 kcal/mol, as well as the T-(T) difference within 0.1 kcal/mol.

P71

Spin-Orbit Induced Dynamics in Atom-Ion Collisions

Tomáš Jíra (University of Chemistry and Technology, Prague), Marcin Buchowiecki (University of Szczecin)

This study investigates spin-orbit-induced excitation and quenching dynamics in nitrogen cation and helium collisions utilizing a two-dimensional non-adiabatic wavepacket approach. Because these specific reaction pathways are governed entirely by a minimal set of spin-orbit couplings, they provide an ideal baseline for examining multi-state scattering phenomena. The dynamic simulations are constructed from ab initio potential energy curves and effective coupling elements, and the resulting cross sections are rigorously benchmarked against time-independent quantum scattering calculations and semi-classical Landau-Zener approximations. Our comparative analysis demonstrates that the wavepacket framework yields excellent agreement with traditional quantum scattering across a broad range of collision energies. It successfully tracks transition probabilities from just above the activation threshold up to significantly higher energy regimes where standard time-independent computational routines often face numerical instability. Ultimately, this wavepacket approach proves to be a robust and mathematically straightforward tool for modeling complex spin-orbit transitions in atom-ion collisions.

P72

Ultrafast Intersystem Crossing and Transient 2c–3e S–S Bond Formation in [(bpy)Pt(SPh)$_2$]

Jannis Leif Johann (University of Vienna, Austria), Jesús Lucia-Tamudo (University of Vienna, Austria), Leticia González (University of Vienna, Austria)

Ultrafast Intersystem Crossing and Transient 2c–3e S–S Bond Formation in [(bpy)Pt(SPh)$_2$] Transition metal complexes exhibit remarkable structural and electronic versatility, providing a rich platform for exploring excited-state processes governed by the interplay of metal- and ligand-centered electronic structure. An intriguing example of this interplay is given in [(bpy)Pt(SAr)$_2$] (SAr = sulfur-substituted aryl) complexes, which show potential to the formation of a transient two-center–three-electron (2c–3e) sulfur–sulfur bond in the lowest triplet excited state. Here we characterize this phenomenon computationally for the complex [(bpy)Pt(SPh)$_2$] using (TD)DFT and non-adiabatic excited-state dynamics. To investigate the ultrafast excited-state relaxation, we employ an anharmonic vibronic coupling (AVC) model, an extension of the linear vibronic coupling (LVC) approach within the SHARC[1] framework, to propagate a large ensemble of excited-state trajectories for one structural configuration of the complex. The resulting dynamics reveal a clear relaxation cascade: rapid internal conversion and intersystem crossing funnel the initially excited population into the lowest-lying triplet state (T1), which becomes the dominantly populated state on a sub-picosecond timescale. A conformer search on the T1 potential-energy surface shows that a subset of the rotamers—those in which the sulfur-centered orbitals on the two thiolate ligands are favorably aligned—undergo a structural contraction that reduces the S–S distance. The corresponding frontier-orbital picture exhibits the bonding characteristics expected for a 2c–3e interaction.[2] Together, these results support a mechanism in which rapid intersystem crossing populates the lowest-lying triplet state, enabling sulfur-centered orbital realignment and transient S–S bond formation.

References

[1] S. Mai, P. Marquetand, L. González, WIREs Computational Molecular Science 2018, 8, e1370.

[2] R. Gleiter, G. Haberhauer, Coordination Chemistry Reviews 2017, 344, 263–298.

P73

Quantum chemical study of possible reaction pathways between IO and CH3OO

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The chemistry of iodine plays a major role in the complex chemistry of the marine boundary layer, especially in the unpolluted atmosphere. A particularly important molecule in this context is the IO radical. Experimental studies already proved the possibility of IO being a factor in the depletion of ozone.
Due to the high reactivity of the IO radical, it is involved in many other important reactions. The reaction with peroxy hydrocarbons, especially CH3OO, is of particular interest because of the lack of reliable experimental reaction rates. Even the reaction products are not known for sure, yet. For that reason, the focus of the present work is on the quantum chemical calculation of possible reaction pathways of IO with CH3OO.
The aim of this work is to untangle some of the discrepancies between the different existing studies, experimental and theoretical. The computed reaction energies suggest the initial formation of stable adducts. The reaction energies and enthalpies suggest that several other product channels seem thermodynamically possible. It was possible to calculate a minimum energy path of one possible reactions via a stable adduct and only one barrier with a transition state. It turns out in general that the computed reaction energies are rather sensitive to the treatment of electron correlation, effective core potential, AO basis, and multi-reference effects.

P74

ElemCo.jl: A Julia Package for Electron Correlation Methods

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A Julia package ElemCo.jl for electron-correlation methods is presented. The package is designed to be easy to use and to provide a high level of abstraction to the user. Inputs are Julia script files, which can also be used to define custom methods or access the package's internal functions.

The package provides a wide range of methods, including the closed-shell and open-shell coupled-cluster methods CCSD, CCSD(T), CCSDT, distinguishable-cluster approaches, DCSD and DC-CCSDT, Full CI, a selected CI method CIPHI, EOM-CCSD and EOM-DCSD, etc. Furthermore, the package includes, e.g., the two-determinant coupled-cluster and frozen-reference coupled-cluster methods, which can be used to calculate excited states, and tensor-decomposed distinguishable-cluster methods, which can be used to calculate ground-state energies of large systems.

The one- and two-electron integrals can either be computed (using the Libcint library) or read from a file in the FCIDUMP format, including the possibility to read similarity-transformed (transcorrelated) or embedded Hamiltonians.

The package is open-source and available via the Julia package manager or on GitHub at https://github.com/fkfest/ElemCo.jl. Documentation can be found at https://elem.co.il. The inputs can be generated using the jlmol visualizer https://jlmol.com, which is available as a web-application https://app.jlmol.com and a stand-alone application https://github.com/fkfest/jlmol.

P75

Probing Isotope Effects on Metal–H_2 Chemical Bonding in Cu-, Zn-, and Bimetallic Cu/Zn-HKUST-1 Metal–Organic Frameworks

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Metal–organic frameworks (MOFs) are promising materials for hydrogen storage and isotope separation due to their tunable pore structures and strong adsorption sites at open metal centers (OMCs). Chemical affinity quantum sieving (CAQS) enables isotope selectivity through differences in the zero-point vibrational energies and adsorption enthalpies of hydrogen isotopologues.[1,2]
Among MOFs, Cu-based HKUST-1 is a well-studied material for hydrogen isotope separation. Zn substitution is particularly interesting because it has been reported to increase hydrogen isotope selectivity in other MOFs.[3] However, although Zn substitution can strengthen hydrogen adsorption, it can also decrease the pore size. Therefore, mixed Cu/Zn HKUST-1 systems are investigated to explore the effects of partial Zn substitution. Despite extensive studies, how Zn substitution changes the chemical bonding at the OMC remains insufficiently understood.[4]
In this work, DFT calculations at the B3LYP-D4/def2-TZVPP level were applied to investigate the influence of the OMC on $H_2/D_2/T_2$ adsorption. Substitution of Cu by Zn increases H_2 adsorption strength but also induces structural changes and distorts the framework symmetry. When only one Cu atom is substituted by Zn, H_2 adsorbs more strongly at the Zn site than at the Cu site. H_2 adsorption was further studied by progressively increasing the number of adsorbed H_2 molecules up to five. Anharmonic zero-point vibrational energy corrections were calculated using VPT2.
To capture geometric isotope effects upon H_2 adsorption, NEO-DFT (Nuclear–Electronic Orbital DFT) geometry optimizations were performed.[5] Smaller paddle-wheel fragments were used to overcome convergence limitations for the larger systems. Finally, energy decomposition analysis was done to investigate changes in metal–H_2 bonding upon Zn substitution. The interaction is dominated by σ-donation from H_2 to the OMC together with π back-donation. Comparing the individual bonding-energy components will clarify how Zn substitution modifies the metal–H_2 interaction and contributes to changes in hydrogen adsorption and isotope effects.

References
[1] I. Savchenko,Chem. Phys. Lett. 2017, 670, 64–70.
[2] H. Oh, Eur. J. Inorg. Chem. 2016, 4278–4289.
[3] W. Zhuang, J. Chem. Phys. 2025, 163, 214709.
[4] C. Jiang, Nanoscale Adv. 2022, 4, 2077–2089.
[5] S. Hammes-Schiffer, J. Chem. Phys. 2021, 155, 030901.

P76

Investigation of HCl-H2O electrolyte using AIMD simulations

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Renewable and eco-friendly energy storage systems are the key elements to address the current global challenges such as the reduction of fossil fuels and climate change. Improving the efficiency of the electrochemical batteries and capacitors is the most promising solution in that direction. To realize this goal in practice, novel electrode materials and electrolytes need to be developed. To achieve the optimal performance of the current and novel electrochemical systems the fundamental mechanistic understanding of the processes takin place within the bulk of electrolytes and at the electrode-electrolyte interfaces has to be gained. Electrochemical processes in liquid electrolytes and at the interfaces exhibit complex nature due to the interplay between various physico-chemical effects such as electrostatic interactions, quantum effects, thermodynamics and hydrodynamic motion. Such effects operate on different length and time scales thus, to tackle this problem, an effective combination of various computational and analytical methods has to be developed. In this work a prototypical HCl-H2O liquid electrolyte is investigated using ab initio molecular dynamics simulation using Quickstep module of CP2K software.[1] HCl concentration range between 0.44 molal and 3.21 molal is considered. Radial distribution functions and coordination numbers of various species within the electrolytes are investigated in detail. It is shown that O-O RDF and coordination number vary with concentration whereas for other species the respective properties are less sensitive to concentration. A particular emphasis of this study is made on HCl dissociation reaction dynamics and energetics. Temporal evolution of proton movement in form of Grotthuss diffusion is monitored and relevant cations, i.e, Eigen and Zundel are investigated. For all concentrations HCl and surrounding 4 H2O molecules are isolated at the instant of HCl dissociation. Identifed clusters are used as input for quantum chemical density functional theory calculation including Cosmo correction, as implemented in Turbomole 7.9 program [2], to estimate energetics of the reactions.

References
[1] J. Chem. Phys. 152, 194103 (2020)
[2] J. Chem. Theory Comput. (2023) 19 (20): 6859-6890

P77

Analytical gradients and non-adiabatic couplings within the algebraic diagrammatic construction scheme

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Accurate description of excited states is crucial for understanding a wide variety of photochemical and photophysical processes. Because these processes involve complex electronic transitions, reliable methods for modeling excited-state properties are essential. The algebraic diagrammatic construction (ADC) family of methods provides a powerful and efficient framework for calculating such properties, offering a good balance between accuracy and computational cost. In this work, we extend ADC methods by implementing analytical energy gradients within the Q-Chem electronic structure package [1]. This extension enables the exploration of excited-state potential energy surfaces (PES), geometry optimizations, and the characterization of minimum-energy crossing points (MECPs). Furthermore, we implement non-adiabatic couplings (NACs) for ADC(2), ADC(2)-x, and ADC(3) using the summed-state approach [2,3]. The performance and applicability of the implementations are demonstrated on selected molecular test systems.

References

[1] Rehn, D. R.; Dreuw, A. J. Chem. Phys. 150, 174110, 2019.
[2] Tajti, A.; Szalay, P. G. J. Chem. Phys. 131, 124104, 2009.
[3] Kim, M.; Rehn, D. R.; Dreuw, A.; Faraji, S. Phys. Chem. Chem. Phys. (in preparation).

P78

Emergence in Computational High-Pressure Chemistry: Equations of State of Molecular Clusters

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As a first approximation, many properties of compounds can be understood by considering the smallest possible model system; in the case of certain molecular properties, a single molecule is sufficient. For example, many simulated vibrational modes agree well with experimental spectra using this single-molecule approximation. However, this approach is limited to properties that are mostly governed by intramolecular effects [1,2]. Intermolecular effects like hydrogen bonds or packing effects cannot be captured by single-molecule approaches. In particular, packing effects are important in the context of high-pressure chemistry, as molecules move closer together with increasing pressure. Cluster approaches are one way to include intermolecular effects, but the question remains what cluster size and structure are needed to reproduce experimental data.
In this work, we present an automatic workflow for generating molecular clusters and compressing them in a dynamic, computationally inexpensive approach, linked with the Conformer Rotamer Ensemble Sampling Tool (CREST) [3] using the eXtended Hydrostatic Compression Force Field (X-HCFF) [4]. We show that for simple compounds like hydrogen, methane, and ethane, the resulting equations of state (V/V_0 curves) are in quantitative agreement with experimental data when large clusters of over 100 molecules are used. As the pressure-dependent molecular volumes converge with increasing cluster size, they can be considered emergent properties. This work paves the way for analyzing other pressure-dependent properties, or more complex compounds, using molecular clusters.

References

[1] C.-M. Hsieh, B. Grabbet, F. Zeller, S. Benter, T. Scheele, N. Sieroka, T. Neudecker, *ChemPhysChem* **2022**, *23*, e202200414.
[2] R. Weiß, F. Zeller, T. Neudecker, *J. Chem. Phys.* **2024**, *160*, 084101.
[3] P. Pracht, S. Grimme, C. Bannwarth, F. Bohle, S. Ehlert, G. Feldmann, J. Gorges, M. Müller, T. Neudecker, C. Plett, S. Spicher, P. Steinbach, P. A. Wesołowski, F. Zeller, *J. Chem. Phys.* **2024**, *160*, 114110.
[4] T. Stauch, *J. Chem. Phys.* **2020**, *153*, 134503.

P79

Chemical Reaction Machine Learning: Data-Centric Shortcuts for Reaction Energetics and Transition State Geometries

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State-of-the-art quantum chemical methods for reaction property prediction require proficient users and are too expensive for everyday use in synthetic laboratories. Yet, the toolkit has expanded tremendously during the past decades: Advances in thermochemistry, conformer sampling, and algorithms designed to locate critical points on the potential energy surface (PES) have enabled skilled computational chemists to calculate almost any desired property of almost any given reaction. However, their computational cost prevents routine application in laboratories around the world. Machine learning (ML) offers a paradigm shift, delivering comparable predictions within seconds, yet chemical reaction ML remains unfamiliar to many theoretical chemists, often conflated with machine-learned interatomic potentials (MLIPs) or molecular structure-to-property prediction, which are inherently different in many regards. This talk will introduce direct chemical reaction ML: models that predict e.g., reaction energetics and transition state geometries starting with as little as connectivity information only, similar to how experimental colleagues envision their molecules. Unlike MLIPs, which approximate the PES, chemical reaction ML learns patterns across the chemical transformations themselves, mimicking chemical intuition and enabling rapid screening of synthetic pathways and mechanistic hypotheses. The data used to train such models is also very different from other ML applications in chemistry, has to be high in quality, both regarding the level of theory and the methodology to obtain the required properties. Currently, this data is the rate-limiting step in the field. As for undergraduate students laying the foundation for their chemical intuition, the reactions visited during learning are highly important. Selecting the most informative training data is a non-trivial task, requiring not only chemical domain knowledge but also insights from mathematics, data science, and machine learning: curriculum learning, active learning, and uncertainty quantification. Dataset generation is costly but built on state-of-the-art quantum chemical software and advances. Close collaborations between disciplines — theoreticians and experimentalists, method developers and data curators — are crucial to move chemistry forward. The talk will display ongoing work as well as results and conclusions from generative and predictive frameworks already developed (ChemTorch, GoFlow, ChiFlow).

P80

Photophysical Properties of a MR-TADF Emitter in Complex Environments

Jennifer Kremper (Heinrich-Heine University Düsseldorf / DE), Oliver Weingart (Heinrich-Heine University Düsseldorf / DE), Jan Meisner (Heinrich-Heine University Düsseldorf / DE)

Thermally activated delayed fluorescence (TADF) emitters are crucial for highly efficient organic light-emitting diodes OLEDs, because they boost light emission by converting triplet excitons into fluorescent singlet excitons without relying on heavy metals. The efficiency of TADF emitters is largely determined by the energy gap between singlet (S1) and triplet (T1) states, as well as key factors like oscillator strength and spin-orbit coupling.[1] Furthermore, the accurate treatment of the chemical surrounding is essential to predict their photophysical behavior in optoelectronic applications. In this study, we employ a hybrid computational approach[2] combining quantum mechanics/molecular mechanics (QM/MM) with density functional theory (DFT), time-dependent (TDDFT), and multireference configuration interaction (DFT/MRCI[3]) to investigate the excited-state behavior of a well-known TADF emitter (DiKTA[4]) in various solvents (acetonitrile, cyclohexane, water), MOF-5 and polymethylmethacrylate (PMMA) as polymer matrix. To account for environmental effects on molecular geometries, we perform QM/MM optimizations using DFT and TDDFT to obtain realistic excited-state conformations.[2] The photophysical properties are subsequently calculated with DFT/MRCI to accurately compute the electronic excitation energies.[3] By applying this method to different environments, we quantify the impact of polarity and rigidity on the above mentioned key TADF parameters. This computational framework can assist in tailoring TADF emitters for specific applications, guiding experimental efforts in material selection for organic light-emitting diodes OLEDs and other optoelectronic devices. Our findings highlight the necessity of incorporating environmental effects in theoretical models to achieve accurate predictions of photophysical behavior in real-world applications.

References

[1] A. Vollrath, M. N. A. Fetzer, J. Kremper, et al. (submitted).
[2] M. Bracker , et al. Phys. Chem. Chem. Phys. 2019, 21(19), 9912-9923
[3] C.M. Marian et al., WIREs Comput. Mol. Sci., 2019, 9(2), e1394.
[4] C. Prentice et al., Chem. Eur. J. 2023, 29, e202202998.

P81

Computational investigation on red-light driven molecular motors enabled by triplet-triplet annihilation upconversion

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Light-driven molecular rotary motors convert photon energy into controlled directional motion and offer opportunities in areas ranging from photopharmacology and optogenetics to photoresponsive materials. However, many molecular motors require ultraviolet or high-energy visible light for direct excitation, limiting their application in biological and other light-sensitive environments. Shifting motor operation toward longer wavelengths is therefore an important challenge. Here, we investigate triplet–triplet annihilation photon upconversion (TTA-UC) as an indirect excitation strategy for driving a second-generation Feringa-type molecular rotary motor with lower-energy light. In TTA-UC, two low-energy photons are harvested through a sensitizer–annihilator system and converted into a higher-energy singlet excitation that can subsequently be transferred to the molecular motor. Particular attention is given to organic radical sensitizers with doublet character as alternatives to conventional metal-based photosensitizers. Using quantum-chemical calculations, we evaluated the excited-state energetics and energy-transfer requirements of sensitizer–annihilator–motor combinations involving the radical sensitizers TTM–TPA and BNS and the annihilators perylene and rubrene. The poster presents the energy-transfer pathway from low-energy excitation to molecular rotation, compares the investigated sensitizer–annihilator combinations, and highlights the key energetic design criteria emerging from the calculations. These results provide a basis for the rational design of molecular rotary motors that can be indirectly activated using lower-energy visible light.

P82

Internal conversions with non-adiabatic instanton theory

S.L. Krug (ETH Zurich, Switzerland), M.R. Fiechter (ETH Zurich, Switzerland), J.O. Richardson (ETH Zurich, Switzerland)

Radiationless decay competes with light emission across many applied fields, such as organic electronics and photovoltaics. Whenever an excited molecule returns to its ground state, internal conversion drains population that would otherwise be emitted as light. Controlling this loss channel is central to organic light-emitting diodes and light-harvesting materials, while the same process underlies the photostability of UV-absorbing molecules. I will approach such internal conversions through nonadiabatic instanton theory, which locates an optimal, local tunneling path between a reactant and a product potential energy surface (PES). This instanton is a stationary-action path – minimal with respect to the hopping point between the surfaces and a saddle point with respect to the imaginary time of the transition. Beyond a substantial reduction in computational cost, it grants a classical-mechanical interpretation of the dynamics and, with it, insight into the reaction mechanism. Locating this path is intricate even in simple systems, and especially so in the deep inverted regime or when the two surfaces do not intersect at all; these regimes are governed by so-called branch-point instantons. These have so far been treated rigorously only for harmonic oscillators; since real systems are rarely purely harmonic, I extend the analytical treatment to Morse oscillators by means of a semiclassical Morse propagator. Internal conversion (S1 -> S0) in naphthalene is a prime example of such an anharmonicity-dominated reaction. As the parent of the acene family on which many photoactive materials are built, and a prototypical aromatic for radiationless transitions, it is a natural model system; moreover, its S1 -> S0 conversion can be modeled by two Morse oscillators, so that the essential physics beyond the harmonic picture already emerges from a one-dimensional treatment along the reaction coordinate.

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The ultranonlocal LAK meta-GGA: An efficient density functional for molecules, materials, and interfaces

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Over the past decade, meta-GGAs have significantly advanced the accuracy–efficiency trade-off in density functional theory, approaching hybrid-functional accuracy while retaining nearly GGA computational efficiency. Meta-GGAs like B97M-V and SCAN provide accurate descriptions of a wide range of energy-related properties. The recently developed Lebeda–Aschebrock–Kümmel (LAK) meta-GGA [1,2] additionally captures the ultranonlocality of the derivative discontinuity to achieve accurate band gaps for the right reason [3].

In our contribution, we briefly explain the non-empirical design strategy of the LAK meta-GGA and highlight its performance for molecular systems [2], covering thermochemistry, kinetics, and non-covalent interactions, and for extended materials, including band gaps [1] and defect formation energies [4].

The combination of high accuracy for both molecules and materials makes LAK particularly promising for a unified description of complex surface and interface problems. Indeed, on a recently established benchmark database for adsorption energies covering layered, porous, ionic, and metallic surfaces, LAK achieves near-chemical accuracy and provides consistent performance across the considered surface classes [5]. Remarkably, LAK achieves the best overall performance among all tested dispersion-corrected GGAs, meta-GGAs, and hybrid functionals, even without adding a dispersion correction. This is because LAK incorporates near-equilibrium dispersion interactions directly into the exchange-correlation functional by design [2].

Finally, we discuss remaining challenges and future directions in meta-GGA development, with particular emphasis on the description of spin-polarized systems and magnetic materials [6].

References

[1] T. Lebeda, T. Aschebrock, and S. Kümmel, *Phys. Rev. Lett.* **133**, 136402 (2024).
[2] T. Lebeda and S. Kümmel, *Phys. Rev. B* **111**, 155133 (2025).
[3] T. Lebeda, T. Aschebrock, J. Sun, L. Leppert, and S. Kümmel, *Phys. Rev. Mater.* **7**, 093803 (2023).
[4] J. V. Bazantes, T. Lebeda, A. Ramasamy, K. Pokharel, R. Zhang, J. P. Perdew, and J. Sun, *arXiv:2604.05385* (2026).
[5] G. Kler-Young, F. della Pia, T. Lebeda, F. Berger, G. Csányi, and A. Michaelides (in preparation).
[6] R. Maniar, C. Shahi, J. Sun, J. P. Perdew, and T. Lebeda (in preparation).

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Light-induced electron dynamics in open-shell systems via the restricted open-shell method

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The description of correlated, ultrafast electron dynamics in polyatomic many-electron molecules is generally a challenging task - and even more so for open-shell systems. The time-dependent configuration interaction (TDCI) method has been shown to correctly describe the light-induced excitation processes on the natural time scale of the electrons. By employing spin orbitals constructed from molecular orbitals, monitoring the electronic wavepacket evolution in the TDCI framework becomes possible, and the Jellyfish software package provides specialised time-dependent tools to analyse the light-driven dynamics. [1, 2] Investigating the dynamics of an open-shell molecule is a greater challenge, as it requires the construction of the spin-adapted wavefunction to avoid spin contamination and guarantee a physically solid foundation. In this poster, we present and discuss the light-driven electron dynamics simulation in open-shell molecules using a restricted open-shell approach.

References

[1] F. Langkabel, P. A. Albrecht, A. Bande, P. Krause, Chem. Phys., 557, 111502 (2022)
[2] F. Langkabel, P. Krause, A. Bande, WIREs Comput Mol Sci., 14, e1696 (2024)

P85

Freezing the Unfreezable: para-Hydrogen Microsolvation Tunes CH5+ Fluxionality

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Understanding how molecular solvation shapes structure and dynamics at the quantum level is a central goal of modern Solvation Science. Protonated methane (CH5+), a highly fluxional molecule known for its large-amplitude motion (LAM) presents a uniquely sensitive system for exploring the effects of microsolvation in para-hydrogen clusters (pH2)n at low temperatures (15 K). We investigate CH5+-(pH2)n using ring polymer molecular dynamics (RPMD) simulations based on highly accurate high-dimensional neural network potentials. These RPMD simulations serve as basis for our IR spectra computation using a neural network dipole moment surface. All networks are parametrized using CCSD(T) theory. We find that microsolvation significantly modulates the LAM of CH5+ as a function of pH2 cluster size n, and for small clusters transiently freezes the CH5+ molecule. This dynamical change leaves clear spectroscopic fingerprints: The emergence of a partial scrambling band and systematic mode shifts connected to the structural differences between frozen and fully flexible CH5+. As larger cluster sizes provide a broader interaction site with pH2, we observe the reentrance of full scrambling of CH5+. Together, the simulation and spectral results are in excellent internal agreement and provide a coherent picture of how para-hydrogen controls the fluxionality of CH5+ at 15 K.

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Simulating mechanochemical disassembly pathways of PdnL2n supramolecular architectures

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Mechanochemical activation provides a powerful route to trigger the disassembly of supramolecular PdnL2n architectures, yet the underlying bond rupture and fragmentation pathways remain difficult to resolve at the atomistic level. Here, we apply ramped steered molecular dynamics to probe force-induced Pd-N bond rupture in a Pd2L4 cage and a giant Pd12L24 nanosphere, complementing experimental ultrasound-induced disassembly studies of these systems.[1] Because of the size of these solvated assemblies, conventional ab initio dynamics are impractical for this problem. Therefore, the machine-learned interatomic potential (MLIP) Universal Model for Atoms S (UMA-S)[2] was fine-tuned for Pd-N bond dissociation in coordination chemistry using a dataset of over 1.6 million structural configurations spanning six different relevant molecular motifs. Fine-tuning was necessary to correct structural artifacts as the pre-trained model produced unphysical, asymmetric distortions upon geometry optimization, whereas the fine-tuned potential enables stable, reactive simulations of solvated supramolecular assemblies beyond the reach of conventional quantum chemical approaches. The simulations of Pd2L4 reveal that the onset of cage disassembly is strongly influenced by how force is transmitted through the ligands. Pulling two adjacent ligands apart, referred to as cis pulling, promotes Pd-N bond rupture more efficiently than trans pulling, where force is applied to two opposite ligands. Across all pulling scenarios, ligand dissociation is observed, leaving behind Pd-coordinated fragments whose composition depends on the pulling geometry. In the Pd12L24 nanosphere, multipoint pulling reveals a stepwise disassembly mechanism in which successive Pd-N bond rupture events progressively fragment the nanosphere into smaller Pd-coordinated species and free ligand. These results demonstrate how fine-tuned MLIPs can extend computational mechanochemistry from small mechanophores to large, reactive supramolecular coordination assemblies, opening a route to molecular-level mechanistic insights in systems previously beyond computational reach.

References
[1]David, T.; Lennarz, R., et al., Mechanochemical disassembly pathways of self-assembled polymer-decorated PdnL2n supramolecular architectures. Nat. Commun. 2026, 17 (1), 5459.
[2] Wood, B.; Dzamba, M., et al., UMA: A family of universal models for atoms. Advances in Neural Information Processing Systems 2026, 38, 129391-129427.

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First-Principle Investigations on BN-Modulated Graphene Nanoribbons

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Using spin-polarized PBE first-principles calculations, we investigate saturated and unsaturated BN-modulated zigzag graphene nanoribbons[1, 2]. Structural relaxation reveals that the changes in the local geometry leads to electron density redistribution, modifying the magnetic behavior of the nanoribbons. The localized spin-polarized edge-states of the pristine zigzag nanoribbon are preserved in BN-modulated systems. However, in the BN modulated systems, the geometrical asymmetry leads to unequal contributions on the edges, giving rise to a net magnetic moment in system. This can be further tuned based on the BN content, resulting in a tunable net magnetic moment perpendicular to the ribbon. Bader charge analysis is employed to examine the charge distribution and its correlation with the magnetic response. In addition, the electronic structure shows a spin-dependent band-gap variation upon BN modulation, indicating a strong dependence of the electronic properties on the edge chemistry. These trends are also relevant to 2D BN/graphene lateral heterostructures, where interface engineering may provide another route to control edge-state magnetism[3]. Overall, these preliminary results suggest that BN modulation is an effective strategy for tuning the magnetic and electronic properties of graphene nanoribbons.

References

[1] T. Das, S. Chakrabarty, Y. Kawazoe, G. Das, Tuning the electronic and magnetic properties of graphene/ h -BN hetero nanoribbon: A first-principles investigation. AIP Advances, 2018. 8: p. 065111.

[2] H. Lee, Y.-W. Son, N. Park, S. Han, J. Yu, Magnetic ordering at the edges of graphitic fragments: Magnetic tail interactions between the edge-localized states. Physical Review B, 2005. 72(17): p. 174431.

[3] M. P. Levendorf, C.-J. Kim, L. Brown, P. Y. Huang, R. W. Havener, D. A. Muller, et al., Graphene and boron nitride lateral heterostructures for atomically thin circuitry. Nature, 2012. 488(7413): p. 627-632.

P88

Mixed Metal Oxides in the Gas Phase: The Study of the FeAlO3+ and Al2O3+ Clusters

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Elucidating the atomistic structure of active sites in heterogeneous catalysis remains a fundamental challenge, due to their intrinsic complexity and dynamic nature under operating conditions. The study of those systems involves techniques such as matrix isolation and gas-phase clusters, which enable detailed structural and electronic characterisation at the molecular level. In the realm of gas-phase clusters experiments, the investigation of mixed metal oxide catalysts uses action spectroscopy approaches, where mass-selected ions (emulating the catalyst's active site) are characterised by techniques such as infrared photodissociation spectroscopy (IRPD) and X-ray absorption spectroscopy (XAS) [1]. Aluminium oxide (alumina) is a prototypical oxide material widely employed in heterogeneous catalysis, both as an active phase and as a support, due to its chemical and physical properties. The incorporation of transition metals into the alumina framework has emerged as an effective strategy to tailor catalytic performance, for instance, in isobutane synthesis [2]. In this regard, the study of the active site of iron-doped alumina is pivotal to understanding its catalytic activity. Herein, FeAlO3+ and Al2O3+ clusters were investigated as model systems for iron-doped alumina active sites by combining IRPD and XAS with electronic structure calculations. Structural assignment is achieved through comparison of experimental spectra with simulated harmonic infrared. The results indicate that the measured cluster adopts a "key-like" structural motif. Further insight into the electronic structure is obtained from X-ray absorption spectroscopy at both the oxygen K-edge and iron L-edge. The combined analysis reveals that FeAlO3+ is best described as an oxyl species with a general quintet spin state, in which the iron centre is in the +3 oxidation state. These findings afford a detailed atomistic picture of a prototypical iron-doped alumina motif, highlighting the power of combining action spectroscopy with simulations to resolve structure–property relationships in complex oxide systems. This work provides fundamental insights into the nature of the active sites in mixed metal oxides and establishes a framework for the rational design of improved catalytic materials.

References

[1] Schwarz, H., Asmis, K.R., et al. Chem Eur. J., 2019, 25, 2112.
[2] Venugopalan, A. T., Kandasamy, P., et al. Catal. Commun., 2021, 150, 106263.

P89

CO Reactivity on Pristine and Co- and Cu-Substituted NiF2(F2) (001) Surface Models: A First-Principles Study

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Electrochemical fluorination on a nickel anode in anhydrous hydrogen fluoride (Simons process) is an important industrial route towards fluorinated compounds. Despite its widespread application, its mechanism is not well understood. It is believed to involve higher-valent nickel fluorides formed in situ under electrochemical conditions. The existence of higher-valent nickel centres on the anode was confirmed recently. [1] To study the reactivity of higher-valent nickel surfaces, we constructed a twice-oxidised NiF2(F2) (001) surface model. This surface model contains both a higher-valent nickel centre and a surface [F2]- unit, which is readily available as a fluorine source. Previously, we have investigated the reactivity of methane [2], ethene [3] and carbon monoxide [4] on this surface. In the case of CO, we have also considered explicit co-adsorption of up to two HF molecules. Within this model we were able to reproduce most of the experimentally observed products. As nickel is the only metal experimentally known to facilitate Simons-type fluorination, a computational comparison with other related transition metals may provide further insights into the underlying mechanism. Here, we present the results of the CO adsorption study on the pristine surface. Furthermore, we extend it with models in which either a surface or subsurface nickel atom is substituted by cobalt or copper. The interaction of CO with these modified surfaces is then investigated and compared with that on the pristine NiF2(F2) surface.

References
[1] G. Senges, A. G. Buzanich, T. Lindič et al., Chem. Sci. 15 (2024) 4504.
[2] T. Lindič and B. Paulus, Materials 17 (2024) 2062.
[3] T. Lindič, J. Ahmad and B. Paulus, ChemPhysChem 27 (2026) e202500117.
[4] T. Lindič and B. Paulus, Mol. Phys. 124 (2026) e2593939.

P90

Excitonic effects in layered Poly(heptazine imide) from many-body perturbation theory

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Poly(heptazine imide) (PHI) has emerged as a promising, metal-free photocatalyst and charge storage material in solar batteries, owing to its suitable band gap and favorable band edge positions.[1][2] Despite such broad application potential, a comprehensive understanding of its photo-response, especially excitonic effects and their dependence on spatial confinement and interlayer couplings remains elusive. Herein, we employ many-body perturbation theory, i.e., simulations on the GW/BSE level, to account for self-energy corrections and electron-hole interactions, both neglected in single-particle approaches such as TDDFT. We investigate the quasiparticle band structure and excitonic effects in various PHI models, including monolayer, bilayer, and bulk structures with different stacking configurations. The simulations reveal localized, Frenkel-type excitons with high binding energies that are modulated by the structural dimensionality and the degree of interlayer stacking. Specifically, the eclipsed AA-stacking configuration exhibits a larger interlayer distance and a reduced quasiparticle band gap. Such configurations exhibit dark bound excitons that are long-lived, due to the out-of-plane band dispersion and concomitant forbidden optical selection rules, compared to their bright excitonic counterparts. We hypothesize that the existence of dark bound excitons is instrumental to charge accumulation and delayed photocatalysis observed in PHI materials. Conversely, the staggered AB-stacking configuration with reduced interlayer distance and broken structural symmetry gives rise to bright bound excitons together with an increased quasiparticle band gap. Our findings provide insights into the optoelectronic properties of layered PHI materials and demonstrate the possibility of property engineering via controlled stacking configurations. The results offer guidance for the rational design of PHI-based functional devices.

References

[1] H. Schlomberg, J. Kröger, G. Savasci, M. W. Terban, S. Bette, I. Moudrakovski, V. Duppel, F. Podjaski, R. Siegel, J. Senker, R. E. Dinnebier, C. Ochsenfeld and B. V. Lotsch, Chem. Mater., 2019, 31, 7478–7486.

[2] Wang and S. Wang, Coordination Chemistry Reviews, 2022, 453, 214338.

P91

Investigation of H3O+-nHe geometries and He-migration

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The H3O+-nHe cluster is chosen as a benchmark-system to investigate molecular systems in superfluid environments. Interesting questions include, e.g., the onset of rotational decoupling with increasing number n of He-atoms. As a first step towards quantum dynamic calculations for the H3O+-nHe clusters, the potential energy surfaces are analyzed. H3O+-nHe clusters ranging from three to thirteen helium atoms are investigated. Specifically, geometries of potential minima, transition state geometries for He-migration and dissociation energies are examined and characterized. The onset of new solvation shells with increasing number of He-atoms are studied. The minimum geometry of the H3O+-4 He cluster turns out to be of special interest, differing from the geometry obtained by imaginary-time pathintegral calculations.(by Schran et al.) In this context, the influence and importance of zero point energies of the minimum geometries is discussed. Future steps might include, e.g., quantum dynamics calculations studying He-migration, vibrational predissociation and rotational decoupling using methods like MCTDH.

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Development and combination of novel basis set convergence accelerators for CCSD(T) and demonstrations on large molecular complexes

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While the local natural orbital (LNO) method of our group [1] enables relatively routine access to CCSD(T) for systems with hundreds of atoms, approaching their complete basis set (CBS) limit still remains highly challenging. Namely, large atomic orbital (AO) basis sets often augmented with diffuse functions are needed, leading to higher cost.
To accelerate the convergence to the CBS limit, we introduce and combine here three efficient tools developed recently in our MRCC program suite. The present authors introduced a novel, general Floating Orbital Grid (FOG) approach that automatically generates a non-atom-centered basis (with center position, exponents, etc.) between or around interacting monomers. The advancement over the common single midbond functions is that FOG is applicable irrespectively of the size, shape, and number of the interacting units.
Then, we combine the FOG method with complementary basis set convergence accelerators, namely an LNO-accelerated density-based basis set correction (DBBSC) [3]; and a highly accurate explicitly correlated CCSD(F12*)(T+) approach [4]. Benchmarks on medium-sized complexes demonstrate that the FOG method added to X-ζ AO bases by itself performs as well as augmented X-ζ or $(X+1)$-ζ interaction energies. Remarkably, combining FOG with DBBSC or F12 methods using double-ζ basis delivers the accuracy of conventional quadruple-ζ quality energies (MAE $\sim$ 0.4 kcal/mol) at a cost significantly lower than triple-ζ. This outstanding performance extends to complexes of real-life drug molecules with full-sized protein binding pockets (300+ atoms), supramolecular binding, and catalyst–substrate interactions, where LNO-CCSD(T)/double-ζ + FOG + DBBSC computations reproduce CBS references within 0.5 kcal/mol.
Together, these advances systematically accelerate the AO basis set convergence to the CBS limit of CCSD(T) or even LNO-CCSDT(Q) [5]. This enables high-throughput computation of CCSD(T)-level benchmark results to assist the development and assessment of lower-cost (DFT, SQE, ML) models [6].

References

[1] P. R. Nagy, *Chem. Sci.* **15**, 14556 (2024).
[2] B. D. Lőrincz, P. R. Nagy, *J. Phys. Chem. A* **128**, 10282 (2024) & (2026) in preparation.
[3] D. Mester, P. R. Nagy, M. Kállay, *J. Chem. Theory Comput.* **20**, 7453 (2024).
[4] M. Kállay et al., *J. Chem. Theory Comput.* **19**, 174 (2023).
[5] B. D. Lőrincz*, V. Fishman* et al., *J. Phys. Chem. A* **130**, 3233 (2026).
[6] Zhao, Lőrincz et al., *Sci. Adv.* eadz8521 (2026); Puleva et al., *Nature Commun.* **16** (2025).

P93

Effects of Mechanical Strain and Local Curvature on Hydrogen Isotope Selectivity in Graphene Membranes

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Atomically thin graphene is a promising platform for hydrogen isotope separation due to its impermeability to most atoms and selective proton permeation [1]. The experimental work of the Lozada-Hidalgo group demonstrated that mechanically exfoliated monolayers of graphene can effectively separate hydrogen ion isotopes, achieving a separation factor (H+/D+) of 10 at room temperature [2]. Under environmental conditions, graphene membranes are not perfectly flat [3]. Thermal fluctuations and hydration forces induce intrinsic in-plane and out-of-plane corrugations. Experimental studies have shown that high-proton-conductivity regions are observed near ripples, wrinkles, and strained regions, indicating that local lattice deformation is a key factor in hydrogen isotope transfer [4]. In this work, we investigate how controlled biaxial tensile strain (1–5%) and local curvature influence isotope selectivity (H+, D+, T+) in graphene membranes. Moderate tensile strain increases interatomic spacing and reduces the activation barrier for proton transfer, while local curvature modulates the barrier according to its out-of-plane deformation. The results provide insight into how biaxial strain and ripples can modulate proton conductivity and isotope separation performance in graphene membranes.

References

[1] Friedrich, M., & Stefanelli, U., Communications in Mathematical Physics, (2019), 379(3). [2] Lozada-Hidalgo, M., Hu, S., Marshall, O., Mishchenko, A., Grigorenko, A. N., Dryfe, R. A. W., Radha, B., Grigorieva, I. V., & Geim, A. K., Science, (2016), 351(6268). [3] Mogg, L., Zhang, S., Hao, G. P., Gopinadhan, K., Barry, D., Liu, B. L., Cheng, H. M., Geim, A. K., & Lozada-Hidalgo, M. Nature Communications, (2019), 10(1).
[4] Wahab, O. J., Daviddi, E., Xin, B., Sun, P. Z., Griffin, E., Colburn, A. W., Barry, D., Yagmurcukardes, M., Peeters, F. M., Geim, A. K., Lozada-Hidalgo, M., & Unwin, P. R. Nature, (2023), 620(7975).

P94

Deriving and Benchmarking Triplet–Triplet Exciton Couplings for Density Functional Tight-Binding

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Triplet–triplet exciton interactions are central to multi-exciton processes in organic semiconductors, yet their quantitative description remains challenging for large molecular systems. In this work, we focus on establishing and validating a density functional tight-binding (DFTB)-based framework for computing triplet–triplet exciton couplings. First, we benchmark DFTB triplet excitation energies and relevant electronic properties against higher-level electronic-structure methods for representative organic semiconductor molecules and dimers. This provides a systematic assessment of the accuracy and transferability of the underlying DFTB parametrization for triplet states. On this basis, we derive a working expression for the effective triplet–triplet coupling that separates Coulomb and exchange-like contributions, formulated in terms of DFTB quantities such as transition densities or state-specific density matrices. The resulting scheme is designed to be computationally efficient while retaining a clear physical interpretation of the individual coupling components. These benchmarking and derivation steps lay the groundwork for future applications of DFTB to multi-exciton phenomena in extended organic semiconductor systems, such as triplet–triplet annihilation and singlet fission.

P95

QM/MM investigation of structure and reactivity of PSTK dependent on the protonation state of the Thiaminediphosphate cofactor

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Thiamine diphosphate (ThDP), the active functional form of vitamin B1, is a common cofactor for a multitude of enzymatic systems. This ThDP cofactor is known to exhibit uncharacteristically low pKa values at key sites.[1] The low pKa values have been attributed to the protonation of the cofactor and nearby residues[1] both of which are central to the catalysis as well as regulation.[2] We are, therefore, investigating the ThDP cofactor using QM and combined QM/MM methods. Of interest is the behavior of both the isolated ThDP molecule as well as the assembly of the cofactor in the pocket of its host protein. Due to the dimeric structure of the protein, cooperativity between active sites needs to be taken into account in the investigation of the catalytic action.[1] Conformational analysis and the corresponding energies, pKa values, the electric fields acting upon the cofactor as well as the hydrophobicity of the surrounding cavity will be used in combination to provide quantitative insights into the forces acting on the cofactor and the protein environment. Our findings will shed light on the functionality of the protein and open avenues for biomimetic molecular design and catalysis.

References

[1] J. Uranga, F. Rabe von Pappenheim, K. Tittmann, and R. A. Mata, The Journal of Physical Chemistry B 2023 127 (44)

[2] S. Dai, L.M. Funk, F.R. von Pappenheim, V. Sautner, M. Paulikat, B. Schröder, J. Uranga, R.A. Mata, K. Tittmann, Nature 573, 609–613 (2019)

P96

Accelerating plane wave Hartree-Fock and hybrid functional calculations

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Hartree-Fock and hybrid functional calculations with plane wave basis sets are often demanding to converge. Despite their popularity in modern computational solid-state physics, corresponding convergence acceleration schemes still lag behind their counterparts in DFT calculations as well as Hartree-Fock calculations performed with localized basis sets. This work aims at enabling the well-established commutator direct inversion in the iterative subspace acceleration scheme, CDIIS in short, for plane wave calculations with an acceptable overhead per iteration. It builds on previous work that introduced the projected CDIIS method and presented results for large systems (hundreds of electrons). Our results show that this approach is also applicable to smaller systems (less than a hundred electrons) efficiently and therefore is a promising candidate for general use in Hartree-Fock and hybrid functional calculations. Results were produced with the Vienna ab Initio Simulation package (VASP) and the density-functional toolkit (DFTK), proving applicability across different code bases.

P97

Fourth-Generation High-Dimensional Neural Network Potentials for Atomistic Simulations in Solution

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Machine learning potentials have become essential tools in chemistry and materials science, providing accurate and efficient representations of high-dimensional potential energy surfaces for atomistic simulations. Among these, high-dimensional neural network potentials (HDNNPs) offer an approach for modeling complex molecular systems. In this work, we compare two generations of HDNNPs, namely 2G-HDNNPs and 4G-HDNNPs, in their ability to describe organic molecules in aqueous solution. 2G-HDNNPs are highly effective for systems in which the atomic properties are dominated by local chemical environments. By relying on descriptors such as atom-centered symmetry functions (ACSFs) constructed from the neighborhood of each atom within a finite cutoff radius, they accurately capture short-range bonding and local interactions. However, this locality also represents their main limitation: interactions from atoms beyond the cutoff, such as non-local charge transfer, cannot be described explicitly. To overcome this limitation, 4G-HDNNPs incorporate a physically motivated treatment of non-local charge transfer through charges obtained from a global charge equilibration (QEq) scheme that respond to structural changes throughout the entire system. As a result, they can capture non-local charge-transfer effects and long-range electrostatic interactions, making them particularly well suited for systems in which the electronic structure depends on distant atomic environments. In this work, we demonstrate and compare the performance of both approaches for organic molecules solvated in water, demonstrating the advantages of 4G-HDNNPs for systems with non-local charge-transfer effects.

P98

Assessing the frozen natural virtual orbitals and frozen core approximations for fully self-consistent GW

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The GW approximation of many-body perturbation theory has historically been performed in an approximate one-shot perturbative fashion known as G_0W_0. However, some recent methodological advances now allow to systematically perform "exact" GW calculations, *i.e.*, fully self-consistent GW, for realistic systems. While this latter has the same formal scaling as G_0W_0, the self-consistency inevitably introduces a prefactor proportional to the number of iterations. In this work, we investigate two approximations that reduce the computational cost of each single iteration and therefore of the overall self-consistency procedure. These two approximations are defined by discarding some set of orbitals, namely core and high-lying (natural) virtual, in the computation of the dynamic GW self-energy. The errors, as well as the computational benefits, introduced by these two approximations are systematically assessed for molecules first - on a set of 34 small molecules as well as 24 medium-sized organic acceptors - and then for solids where some simple semi-conductors and insulators are considered.

P99

Electronic states of diarylethene switch-gold nanocluster hybrids

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Diarylethenes (DAEs) are promising photochromic molecules because of their thermal stability, reversible switching, and high fatigue resistance under repeated light irradiation[1]. Coupling DAEs with gold nanoclusters may further enhance their optoelectronic performance due to the tunable plasmonic properties and signal-enhancement capability of gold nanostructures[2]. In this work, we investigate how interaction with Au10, Au19, and Au20 clusters modifies the electronic and optical properties of open- and closed-form (stiff) DAE switches[3]. Ground-state properties are studied using density functional theory (DFT) at the B3LYP+D3(BJ) and ωB97X-D levels of theory. Excited-state properties are calculated using linear response time-dependent DFT (TD-DFT) with the same functionals. The excited states are further analyzed using the Fraction of Transition Density Matrix (FTDM) analysis[4], which distinguishes local (molecular or gold) excitations from charge-transfer contributions. The calculated UV-Vis spectra show clear spectral changes upon DAE adsorption on gold clusters, indicating that metal-molecule interactions significantly affect the electronic and optical properties. These results provide insight into the design of gold-DAE hybrid systems for nanoscale optoelectronic and sensing applications.

References

[1] H. Tian and S. Yang, "Recent progresses on diarylethene based photochromic switches," Chem. Soc. Rev., 2004, 33, 85–97.

[2] B. M. Briechle, Y. Kim, P. Ehrenreich, A. Erbe, D. Sysoiev, T. Huhn, U. Groth and E. Scheer, "Current–voltage characteristics of single-molecule diarylethene junctions measured with adjustable gold electrodes in solution," Beilstein J. Nanotechnol., 2012, 3, 798–808.

[3] M. Kleinwächter, "Highly constrained dithienylethenes," PhD dissertation, Humboldt-Universität zu Berlin, 2019.

[4] E. Titov, "On the low-lying electronically excited states of azobenzene dimers: Transition density matrix analysis," Molecules, 2021, 26, 4245.

P100

How to model Spin-Crossover cages in solution

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Spin-crossover (SCO) systems are transition-metal, often Fe(II), complexes capable of switching between low-spin (LS) and high-spin (HS) configurations, in response to external stimuli, such as temperature. This phenomenon is governed by a balance between enthalpic and entropic contributions. [1] Spin-crossover cages, consisting of multiple Fe(II) centers, are of particular interest because they can serve as molecular sensors in solvent environments. Yet their rational design remains largely empirical, underscoring the need for a computational workflow capable of guiding experimental efforts. The aim of this project is to develop a computational methodology with predictive power. Starting from a series of mononuclear Fe(II) complexes in various solvents, for which experimental data for the SCO temperature T1/2 are available, we showcase the challenges and limitations of current computational methodologies. As a first step, a DFT screening of different functionals and dispersion correction schemes was performed to obtain HS and LS energies, while adding thermal and entropic corrections with the standard rigid-rotor-harmonic oscillator model (RRHO).[2] The switching temperatures T1/2 show an extreme dependence on the chosen density functional, which could be attributed to a lack of accurate ΔEHS-LS prediction due to the single reference character. Comparison with experimental data revealed, however, that the local hybrid functional LH20t yielded very good results. To obtain benchmark reference data, CASSCF/NEVPT2 calculations were performed but the multireference treatment over-stabilizes the HS configuration, which is not sufficiently corrected by NEVPT2, leading to a qualitatively incorrect negative ΔEHS-LS. Strategies to improve the description of dynamic correlation are currently underway. In addition, the RRHO model was not able to correctly capture T1/2 modulations arising from different solvents. These initial results show the shortcoming of currently available methods and the need for more realistic chemical models accounting for explicit environments.[3]

References

[1] Halcrow, M. The Effect of Ligand Design on Metal Ion Spin State. Crystals 2016, 6 (5), 58. https://doi.org/10.3390/cryst6050058.

[2] Gorges, J. Towards Understanding Solvation Effects on the Conformational Entropy of Non-Rigid Molecules. Phys. Chem. Chem. Phys. 2022, 24 (20), 12249–12259. https://doi.org/10.1039/D1CP05805C.

[3] Podewitz, M. Generating Reliable and Reproducible Data for Reusable Quantum Chemical Reaction Free-Energy Profiles.

P101

Static DFT Is Enough for Reactive Chemistry, Not for Dispersion: MACE Potentials for Methanol on Si(001)

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Reactive machine-learned interatomic potentials (MLIPs) are currently filling the cost-accuracy gap between DFT and force fields for dynamic modeling of chemical reactions. MLIP training based on fine-tuning of foundational models is currently the most data-efficient way of obtaining performance and transferrable models. MLIPs trained on AIMD frames and active learning approaches are most commonly used. In this project, we aim at probing the lower bound of required ab-initio data for MLIP training by fine-tuning MACE based foundational models on static DFT data. We therefore use available DFT geometry optimizations, NEB calculations and optimized transition state structures from our previously investigated reaction network of methanol on Si(001), previously investigated by Göbel et al. [1] We compare the fine-tuning of MACE-MP0 with its multiheaded successor MACE-MH1. By splitting of the train/test set and exclusion of a full reaction path from the train and testset, we investigate accuracy and transferability of the obtained ML models. We find that the static-only dataset is enough to enable chemically sensible reactive MD simulations of methanol adsorption on Si(001) at 500 K that recover the reaction network from the dynamics, particularly when using multihead models. Furthermore, we find that training with and without D3 dispersion corrections yields energetically indistinguishable models for the tested configurations. We trace this back to a gap in the sampled configuration space of the training set: reaction-path frames never populate the longer-range structures representing the molecule approaching the surface, so the label choice is inert regardless of whether dispersion is included in the DFT reference. More generally, this identifies the sampled configuration space, rather than the choice of reference labels, as the binding constraint when MLIPs are trained from reaction-network data.

References

[1] J. Phys. Chem. C 2025, 129, 10911

P102

Fragment Molecular Orbital Methods for Excited States and Nonadiabatic Dynamics in Large Molecular Assemblies

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We present a fragment molecular orbital (FMO)-based electronic structure method for simulating optical properties and nonadiabatic excited-state dynamics in large molecular assemblies containing hundreds of electronically coupled molecules.
In our approach, the excited states are represented in a quasi-diabatic basis of local-excitation and charge-transfer configurations. Matrix elements of the full electronic Hamiltonian are obtained from embedded monomer and dimer calculations. For computational efficiency, we employ a tight-binding approximation, yielding FMO-LC-TDDFTB, although the formulation can be combined with higher-level electronic-structure methods as well. Analytical energy gradients enable nonadiabatic dynamics using Ehrenfest propagation and its collapse-to-a-block decoherence correction.
To demonstrate the accuracy, efficiency and scope of the method, we studied a range of organic materials. Relative to unfragmented LC-TDDFTB, fragmentation introduces only small deviations in excited-state energies while greatly reducing computational cost. The ground-state self-consistent calculation scales nearly linearly, whereas the effective scaling of the excited-state calculations is approximately quadratic to cubic, depending on fragment size [1]. We simulated exciton transport in an aggregate of 30 anthracene molecules comprising 720 atoms, following the migration and delocalisation of an initially localised excitation over picosecond time scales [2]. We further investigated the bacterial light-harvesting complex LH2, treating all 27 bacteriochlorophyll chromophores within a unified fragment-based framework. This enables on-the-fly nonadiabatic dynamics of the complete pigment aggregate beyond a conventional Frenkel-exciton description and resolves energy flow between the B800 and B850 pigment rings.
The method has also been combined with a generalised Tavis–Cummings Hamiltonian to describe strong light–matter coupling between large molecular aggregates and quantised photon modes [3]. Adding photonic states to the quasi-diabatic basis yields a polaritonic Hamiltonian that enables the calculation of polarisation-dependent spectra and dispersions in aggregates containing hundreds of molecules. All methods are implemented in the open-source DIALECT software package [4, 5].

References

[1] R. Einsele, J. Hoche, R. Mitrić, J. Chem. Phys. 2023, 158, 044121.
[2] R. Einsele, R. Mitrić, J. Chem. Theory Comput. 2024, 20, 6587–6603.
[3] R. Einsele, L. N. Philipp, R. Mitrić, J. Chem. Phys. 2024, 161, 154106.
[4] R. Einsele, X. Miao, L. N. Philipp, R. Mitrić, J. Phys. Chem. A 2025, 129, 6994–7007.
[5] DIALECT, https://github.com/dialect-rs/DIALECT-rs.

P103

Diffusion Mechanisms of Oxygen in $CaMnO_3$ Perovskites and Related $CaMnO_{2.5}$ Ordered Oxygen Vacancy Structures

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Perovskite oxides with the ABO_3 composition are promising redox materials for various applications of two-step thermochemical cycles, such as thermochemical energy storage (TCS) and splitting of H_2O or CO_2. Their tunable composition and defect chemistry allow them to be tailored to various applications. Under reducing conditions, these materials undergo significant non-stoichiometric reduction ($ABO_{3-\delta}$, $0 < \delta < 0.5$), leading to the formation of oxygen vacancies (V_O) and a subsequent topotactic phase transition: The initial perovskite structure with disordered V_O at a low reduction extent ($\delta \approx 0$) transforms into ordered oxygen vacancy (OOV) phases at a high reduction extent ($\delta \approx 0.5$). These two limiting cases provide the background for this study: The perovskite $CaMnO_{3-\delta}$, being a promising candidate for TCS, and its corresponding maximally reduced OOV structure $CaMnO_{2.5+\delta}$ are investigated for their redox thermodynamics and oxygen diffusion kinetics using first-principles density functional theory (DFT). At low reduction ($\delta \approx 0$), the formation energy of isolated V_O is calculated as 1.94 eV, with (nearly) isotropic diffusion barriers of 0.7 eV in all crystallographic directions. At $\delta = 0.5$, two energetically favorable OOV structures are identified for $CaMnO_{2.5}$: First, the experimentally observed structure with all Mn atoms showing a pyramidal 5-coordination, and the V_O being aligned in lines parallel to the crystallographic *c*-direction. Second, a Brownmillerite-type phase with alternating layers of octahedral and tetrahedral Mn atoms and the V_O being aligned in lines parallel to the *a*-direction. Both OOV structures have a formation energy of 1.80 eV per V_O; however, they reveal significant differences in diffusion pathways and activation energies compared to the disordered perovskite phase. The results demonstrate that both the V_O formation energy and the oxygen mobility are strongly dependent on vacancy ordering, with implications for the rational design of perovskites with tailored thermodynamics and redox kinetics. This work provides fundamental insights into how the defect structure changes with reduction and governs oxygen diffusion kinetics, which is key knowledge for optimizing perovskites in energy applications.

P104

Impact of higher-order terms on parity-violating rotational transitions in chiral molecules

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The potential energy surface of a molecule is invariant under inversion of the nuclear coordinates when assuming only electromagnetic interactions, leading to a parity-conserving nuclear-motion Hamiltonian. Consequently, the eigenfunctions describing nuclei moving in such a potential can be chosen to have a well-defined parity [1,2]. However, within the framework of electroweak quantum chemistry, parity is violated. The presence of the weak interaction, mediated by the Z^0 boson, transforms the potential energy surface into a parity-nonconserving one for systems containing more than three atoms [3]. Within this study, perturbation theory is applied for predicting parity-nonconserving energy contributions to rotational transitions within a vibrational state. Predictions are accomplished for the prototypical chiral molecule CHAtFI, for which large parity-violating effects have been found in previous studies [4]. In zeroth-order, the rovibrational wavefunction is a pure product of rigid rotor and harmonic oscillator wavefunctions, whereas refined models construct it from rotational-vibrational perturbation theory [5]. This approach incorporates the coupling between rotations and vibrations, comprising Coriolis forces, centrifugal distortion and changes in molecular shape during vibration. The purpose of this work is to compare the results obtained with and without including third-order derivatives of the parity-nonconserving part of the Born-Oppenheimer hypersurface. Implications for previous estimates of parity-violating contributions based solely on the gradient, which is analytically available in our group [6], are discussed.

References

[1] F. Hund, Z. Phys. 43, 805 (1927).
[2] M. Bartelmann et al., Theoretische Physik 3 | Quantenmechanik, Springer, 2018.
[3] R. Berger and J. Stohner, WIREs Comput. Mol. Sci. 9, e1396 (2019).
[4] R. Berger and J. Stuber, Mol. Phys. 105, 41 (2007).
[5] G. Riley et al., Mol. Phys. 38, 877 (1979).
[6] S. A. Brück et al., J. Chem. Phys. 158, 194109 (2023).

P105

Unraveling GaPt with Machine-Learned Force Fields: Intermetallic Compounds and Oxide Interaction

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Heterogeneous catalysis constitutes a vivid research area; a recently developed new concept is the usage of supported catalytically active liquid metal solutions (SCALMS). SCALMS are based on a matrix metal with a low melting point like gallium, containing catalytically active elements like platinum dissolved in it at low concentrations. The dynamic nature of the liquid metal alloy allows for the formation of isolated catalytic sites, with the gaseous reactants being unable to enter the matrix metal. This enables for the combination of advantages of heterogeneous and homogeneous catalysis, like easy product separation and defined properties of isolated reaction sites.
The dynamic nature of this systems, without stable individual bonds between the atoms, makes the simulation of SCALMS a difficult task. The need for sufficiently large simulation cells and simulation times makes machine-learned force fields (ML-FFs) the method of choice for molecular dynamics (MD) simulations. Since they do not rely on any preselected geometries or bonding situations, they enable the description of highly dynamical SCALMS. Their linear scaling with system size further enables the simulation of large time and length scales.
This poster presents how ML-FF simulations can be used to study the real-time formation of different intermetallic phases in liquid GaPt SCALMS model systems, and the resulting structures are compared to literature and experiments. Furthermore, a brief insight into the interaction of liquid GaPt alloys with oxide layers is given.

References
[1] M. Moritz, S. Maisel, N. Raman, H. Wittkämper, C. Wichmann, M. Grabau, D. Kahraman, J. Steffen, N. Taccardi, A. Görling, M. Haumann, P. Wasserscheid, H.-P. Steinrück, C. Papp, ACS Catal. 2024, 14, 6440-6450
[2] Egger, F.; Mölkner, A.; Steffen, J.; Taccardi, N.; Raman, N.; Zobel, M.; Görling, A.; Wasserscheid, P.; Haumann, M. Advanced Science 2025, 12, e11498

P106

Modelling the quantum dynamics of protonated methane as a benchmark for fluxional molecules

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Protonated methane, CH5+, is the prototypical example of a fluxonial molecular system and its infrared spectroscopy is considered "one of the holy grails of rotational-vibrational spectroscopy". The ro-vibrational states of CH5+ display a unique level scheme, which results from a complex entanglement of rotational and tunneling motions. In this work a new model for the quantum dynamics of CH5+ based on the work of Wodraszka and Manthe is presented. The model uses the symmetry of the multi-well system for the representation of the vibrational wavefunctions as well as the rotational wavefunctions, leading to a minimal model. This minimal model agrees closely with numerically exact calculations, while the new rotational functions lead to a symmetry-compliant representation of the wave function and provide an alternative picture of the entanglement of internal motion and rotation.

P107

Orbital-Resolved Graph Neural Networks for Interpretable Prediction of K-edge XAS in Transition Metal Complexes

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The structural and electronic diversity of transition metal complexes (TMCs) poses a persistent challenge for accurate spectral prediction. To address this, a dataset of transition metal complexes is presented, providing high-quality pre-edge X-ray absorption spectra for a broad range of nickel complexes drawn from the tmQM dataset [1]. For each complex, the Ni K-edge pre-edge features were computed, providing detailed information about 1s to 3d transitions, oscillator strengths, and their sensitivity to the local coordination environment (geometry and ligand identity) and oxidation state. To enable Explainable AI (XAI), the dataset is further enriched with orbital-resolved features, including molecular orbital compositions, atomic contributions to transitions, and decomposed oscillator strengths. These features go beyond spectral prediction; they allow machine learning models to reveal which orbitals, atoms, and structural motifs govern pre-edge signatures, transforming black-box predictions into chemically interpretable insights. By bridging the gap between small-molecule datasets such as QM9 [2] and the chemically rich transition-metal space, this work aims to accelerate the interpretation of XAS spectra.

References

[1] Balcells, D.; Skjelstad, B. B. tmQM Dataset Quantum Geometries and Properties of 86k Transition Metal Complexes. Journal of Chemical Information and Modeling 2020, 60 (12), 6135–6146.

[2] Kotobi, A.; Singh, K.; Höche, D.; Bari, S.; Meißner, R. H.; Bande, A. Integrating Explainability into Graph Neural Network Models for the Prediction of X-ray Absorption Spectra. Journal of the American Chemical Society 2023, 145 (41), 22584–22598.

P108

Mechanistic Insights in Natural and Modified Hammerhead Ribozyme from MD-QM/MM simulations

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Hammerhead ribozymes are catalytically active RNA motifs that have been studied extensively, both experimentally and computationally. However, the molecular mechanism of RNA self cleavage and the role of ions remain debated. Although Mg2+ ions were identified in X-Ray structures, the role of cations in the nucleophile activation and product stabilization are still an open question.[1] Moreover, recent experimental results show that epigenetic modifications on key bases may affect and even increase the catalytic activity of the Hammerhead ribozyme. Molecular Dynamics (MD) simulations are a powerful tool to investigate active site dynamics and obtain mechanistic insights into the altered reactivity induced by sequence modifications.

We devised a computational MD-QM/MM workflow to study the molecular mechanism of RNA self-cleavage in natural Hammerhead ribozyme. MD trajectories are simulated with the standard 12-6-4 Lennard-Jones-type non-bonded parameters for Mg2+by Li and Merz [2] and with the improved nucleic acid-ion force field developed in our group.[3] Additionally, we parametrized the proposed modified nucleobases following the modXNA approach and RESP scheme and incorporated the pseudo-nucleobases into the Hammerhead motif with good structural stability.

The MD simulations verify the positions of two magnesium cations within the active site of the Hammerhead ribozyme. While the simulation with 12-6-4 Lennard-Jones model shows advantages in capturing the positions of magnesium cations in the active region, the new force field demonstrates a better description of the inline-alignment of the nucleophilic attack. MD simulations with the deprotonated general base show the reorientation of the proton of 2'-OH, which supports the predicted nucleophile activation mechanism by the proton abstraction via general base catalysis. These results are foundational for the understanding of ribozyme activity and provides fundamental knowledge into the enhanced activity of modified RNA sequences.

References

[1] Mir, A.; Chen, J.; Robinson, K.; Lendy, E.; Goodman, J.; Neau, D.; Golden, B. L. Biochemistry 2015, 54, 6369–6381.

[2] Li, P.; Song, L. F.; Merz, K. M. Jr. J. Phys. Chem. B 2015, 119, 883–895.

[3] Christanell, L.; König, K.-J.; Holzinger, J.; Schütz, A. K.; Fingerhut, B. P. J. Phys. Chem. B 2026, 130, 6106–6119.

P109

Using Constrained DFT for modelling excited states in periodic systems

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Constrained Density Functional Theory (CDFT) offers a set of tools that, when carefully used, can access the low-lying energy states that cannot be achieved using conventional DFT [1]. In its broader definition it encompasses not only constrains on electron density, but also on spin, which allows for modelling of excited states. This study uses such spin constrain, complemented by the creative usage of Hubbard potential, in solid state plane-wave theory calculations to break the symmetry of a supramolecular unit cell and allow for localization of both electron and electron hole on singular molecules in selected excited states. Combined with GGA functionals and ΔSCF approach it gives a cheap method of describing a change in phosphorescence spectrum due to platinum complex's interaction with its molecular environment. After an initial success in modelling a supramolecular crystal with a flat platinum (II) complex, where a similar crystal had an added hydrogen bond to the platinum atom shifting the phosphorescence spectrum by 13 nm [2], two more systems were selected to be studied. Set of similar crystals sharing a platinum complex, but different in their chemical environment were found in literature with differing spectra [3], and the same approach was used to try and recreate the experimentally measured differences in phosphorescence. Additionally, a pair of crystals where our earlier attempts failed to show a difference in absorbance spectra in higher-level solid state calculations were also studied [4]. CDFT offered not only computationally efficient but also quantitatively correct results. Studies of all mentioned systems allowed for refining the method as well as finding its limitations and strengths.

References

[1] X. Gonze et. al., J. Chem. Theory Comput., 2022, 18, 10, 6099–6110
[2] D. Glosz et. al., Chem. Eur. J., 2024, 30, 40, e202400797
[3] V. Sivchik et. al., Chem. Eur. J., 2021, 27, 5, 1787-1794
[4] E. Kuzniak et. al., Inorg. Chem. Front., 2020, 7, 1851–1863

P110

Non-van der Waals Heterostructures from an Integrated Computational Approach

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Heterostructure interfaces formed by stacking two-dimensional (2D) materials provide a promising route toward achieving advanced electronic and magnetic functionalities at the nanoscale. The automated construction and computational investigation of these systems rely on precise lattice matching between the constituent 2D materials. The resulting supercells can contain hundreds or even thousands of atoms, considering that the individual 2D layers are often subjected to structural strain. In addition, the relative rotational alignment (twist angle) between layers introduces an extra degree of freedom, generating moiré patterns that can be exploited to tailor and enhance interfacial electronic and magnetic properties.

In this work, we perform a high-throughput screening of non-van der Waals (non-vdW) 2D heterostructures. Non-vdW 2D materials originate from non-layered bulk crystals [1] and can be synthesized through both top-down exfoliation techniques and bottom-up growth methods. Our methodology utilizes AFLOW-Hetbuilder, a recently developed computational tool based on the coincidence lattice algorithm for constructing stacked 2D material systems [2–5]. We investigate interfacial binding phenomena across a broad range of heterobilayers, focusing on their structural, electronic, and magnetic properties [6].

The formation of strong covalent and ionic interactions at the interfaces results in the development of hybrid interface bands and pronounced magnetic coupling effects. Additionally, we compare the binding energetics of non-vdW heterostructures with those of conventional van der Waals (vdW) heterostructures. The unique interfacial characteristics of non-vdW 2D heterostructures establish them as promising candidates for advanced technologies, with potential applications in electronics, spintronics, and energy-storage devices [6].

References

[1] R. Friedrich, M. Ghorbani-Asl, S. Curtarolo, et al., Nano Lett. 2022, 22, 989.
[2] D. S. Koda, F. Bechstedt, M. Marques, and L. K. Teles, J. Phys. Chem. C 2016, 120, 10895.
[3] https://zenodo.org/record/4721346 .
[4] C. Oses, M. Esters, D. Hicks, et al., Comput. Mater. Sci. 2023, 216, 111808.
[5] C. Oses, M. Esters, D. Hicks, et al., Comput. Mater. Sci. 2023, 217, 111889.
[6] A. Nihei, T. Barnowsky, and R. Friedrich, Acta Mater. 2026, 315, 122370.

P111

Modelling the Collective Dynamics of Photoinduced Dimerization in a Metal–Organic Framework

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Photoinduced dynamics and reactivity are well understood at the single-molecule level. How these dynamics unfold collectively among many molecules embedded in a periodic matrix, and how they translate into a macroscopic response, remains largely unexplored. One reaction suited to explore this is the [2+2] photodimerization, which we study within a metal–organic framework (MOF) whose stilbene-derived linkers serve as the reacting units, organised by the periodic matrix [1]. We develop a program that automates construction of the periodic framework model from its linker and metal node and sets up the resulting structure for molecular dynamics simulations. The workflow can be generalised for investigations of molecular frameworks with variable network topologies. The molecular environment and framework dynamics of the MOF are described by our previously developed force field MOF-FF [2], which permits molecular dynamics at length and time scales beyond the reach of electronic-structure methods. The force-field description is validated against DFT calculations and the experimentally observed macroscopic structural response. To achieve an efficient and reliable description of the light-induced dynamics, we mimic the excited-state dynamics of the organic linkers by switching the interatomic potential from reactant to product bonding. Combined with the atomistic representation of the molecular matrix, this approach captures the collective dynamics and follows how the photodimerization of individual and multiple centres impacts the periodic structure. Since the photoinduced reaction is assumed not to proceed to completion throughout the crystal under experimental conditions, the spatial pattern of dimerized linkers matters. Our approach therefore enables us to systematically correlate different patterns of reacted linkers with the resulting macroscopic response of the crystal. We present an automated workflow, its validation, and the resulting macroscopic response, and outline how this approach can be extended to other frameworks to elucidate how the reaction cascades into macroscopic effects.

References

[1] Y.-X. Shi et al., Angew. Chem. Int. Ed. 2019, 58, 9453.
[2] S. Bureekaew et al., Phys. Status Solidi B 2013, 250, 1128–1141.

P112

A benchmark study of density functional approximations for predicting the electrical properties of transition metal oxides

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The choice of pseudopotentials (PPs) and density functional approximations (DFAs) is critical for developing a reliable computational cookbook for predicting electrical properties using first-principles methods. In our recent work, we showed that the choice of PPs has only a negligible influence on calculated electrical properties of several transition metal oxides (TMOs). However, experimentally determined atomic structures provide better agreement with experimental electrical conductivity than computationally relaxed structures [1].
In this work, we benchmark a broad range of DFAs, including LDA [2], GGA [3], meta-GGA [4], hybrid functionals [5], and Hubbard corrections [6] with tabulated values from AFLOW [7] and with self-consistently calculated values across a wide range of materials, covering metallic systems to insulators, and including magnetic cases. We compute band gap and intrinsic electrical conductivity using Wannier functions as implemented in the BoltzWann code [8]. We use an automated workflow manager, AiiDA [9], to calculate our benchmarking results.
We report the preliminary benchmarking results by direct comparison with experimental data. We identify strengths and limitations of each functional class and provide a cookbook for best practice DFAs for specific material sets. The take-home message of our work is that improving the band gap prediction does not necessarily improve the prediction of the activation energy of electrical conductivity.

References

[1] A. Öztürk et al., *RSC Adv.* **16**, 33 (2026).
[2] J. P. Perdew, A. Zunger, *Phys. Rev. B* **23**, 5048 (1981).
[3] D. R. Hamann, *Phys. Rev. B* **88**, 085117 (2013).
[4] J. Sun et al., *Phys. Rev. Lett.* **115**, 036402 (2015).
[5] J. Heyd et al., *J. Chem. Phys.* **118**, 8207 (2003).
[6] V. I. Anisimov et al., *J. Phys.: Condens. Matter* **9**, 767 (1997).
[7] C. E. Calderon et al., *Comput. Mater. Sci.* **108**, 233 (2015).
[8] G. Pizzi et al., *Comput. Phys. Commun.* **185**, 422 (2014).
[9] G. Pizzi et al., *Comput. Mater. Sci.* **111**, 218 (2016).

P113

Hydrogen Bond Strength and vSFG spectroscopy of the Water-air Interface

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In present work, we use the strength of hydrogen bonds (HBs) as calculated from energy decomposition analysis based on absolutely localized molecular orbitals (EDA ALMO) for the water molecules on the water-air interface to decipher the vSFG spectra. The contributions of hydrogen bonded water molecules to the different frequency range is examined as a function of HB strength, inter-molecular charge and intra-molecular asymmetry. Similarly, we also establish a linear relationship between the HB strength/charge transfer and instantaneous vibrational frequency of the OH modes of water molecules.

P114

Capturing emergent behaviour in complex reaction networks through quantum chemistry-based kinetic modelling

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Understanding the behaviour of complex chemical systems requires going beyond the description of individual elementary reactions and establishing how microscopic reaction mechanisms give rise to experimentally observable outcomes. However, constructing predictive kinetic models for such systems remains challenging due to the presence of multiple competing pathways, coupled equilibria and reaction steps governed by different kinetic regimes.

We present a quantum chemistry-informed kinetic modelling approach applied to a complex reaction network inspired by the prebiotic nucleotide activation chemistry reported by Liu et al. [1], in which methyl isonitrile serves as a chemical activating agent. By combining quantum chemical calculations of reaction energetics with mechanistic kinetic modelling, we construct a reaction network describing the formation of activated nucleotide species under different reaction conditions.

The developed model incorporates approximately 30 elementary processes, including barrier-mediated transformations, protonation equilibria, conformational interconversion and diffusion-controlled steps. By translating molecular-level reaction energetics into time-dependent concentration profiles, the approach enables direct prediction of product yields and comparison with experimental observations.

Despite the complexity of the reaction network and the inherent uncertainty associated with quantum chemical energy estimates, the model reproduces key experimental trends, including the relative efficiency of different activation conditions and the effects of pH and reagent concentrations on product formation. These results demonstrate how quantum chemistry-informed kinetic modelling can provide mechanistic insight into complex reaction networks and establish a connection between molecular-scale energetics and emergent chemical behaviour.

References

[1] Liu et al. Harnessing chemical energy for the activation and joining of prebiotic building blocks. Nature Chemistry 2020, 12, 1023-1028.

P115

Resonance Raman Spectroscopy of a BODIPY Derivative: Computational Insights from Monomer to Dimer

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BODIPY dyes are an important class of chromophores with strong absorption, high photostability, and tunable electronic properties, making them attractive for applications in molecular photonics, bioimaging, sensing, and light-harvesting systems. Understanding how intermolecular interactions and aggregation influence their excited-state properties and vibrational signatures is therefore of considerable interest. Resonance Raman spectroscopy is particularly well suited to this purpose, as it selectively probes vibrational modes coupled to electronic excitations and provides direct insight into vibronic interactions.

Here, we investigate one-photon absorption and resonance Raman spectra of a BODIPY derivative in its monomeric and dimeric forms using a time-dependent formalism. In this approach, the relevant polarizability tensor elements are expressed in terms of Fourier transforms of time-dependent correlation functions involving nuclear wavepackets propagated on excited-state potential energy surfaces. This formulation describes the spectroscopic response through nuclear wavepacket dynamics, thereby avoiding the explicit sum over intermediate vibronic states required in the corresponding time-independent formulation.

Density functional theory (DFT) and time-dependent density functional theory (TDDFT) are used to systematically characterize the electronic structure and construct model potential energy surfaces. Vertical-gradient, adiabatic-shift, and adiabatic-Hessian approximations are assessed, together with the influence of exchange-correlation functional, basis set, molecular conformation, and explicit and implicit solvation. The monomer serves as a reference system for establishing and validating the computational workflow through comparison with experiment. Among the harmonic models considered, the vertical-gradient model provides a favorable balance between computational cost and agreement with experimental spectra.

For the dimer, the treatment is extended beyond the Born–Oppenheimer approximation by constructing a vibronic-coupling Hamiltonian that explicitly accounts for the coupling between electronic and nuclear degrees of freedom. The resulting quantum dynamics are treated using the multiconfiguration time-dependent Hartree (MCTDH) method, enabling the interplay between vibronic coupling, exciton dynamics, and the resulting spectroscopic signatures to be investigated.

Overall, the developed framework combines systematic electronic-structure calculations and time-dependent resonance Raman simulations with a beyond-Born–Oppenheimer treatment of the dimer, providing a route to investigate the role of vibronic coupling and exciton dynamics in BODIPY aggregates.

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Electron attachment as a route to exotic reactivity: the case of sulfur heterocycles

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Electron attachment can open the way to unusual reactivity, including the opening of the aromatic ring of benzene. [1] Here, we explore whether ring opening induced by low-energy electrons also occurs in a series of sulfur heterocycles: thiophene, benzothiophene, and dibenzothiophene. We combine negative-ion mass spectrometry with electronic-structure calculations to investigate electron attachment, fragmentation, and possible ring-opening reactions in these molecules. We observe that parent anion clusters dominate the negative-ion mass spectra, while the minimum cluster size required for detectable attachment decreases with increasing molecular size. This is consistent with the negative electron affinities of the isolated molecules. Clustering lowers the vertical attachment energy and provides a way to dissipate the excess energy of the incoming electron, allowing the initially formed anion to survive long enough for detection. The decreasing HOMO–LUMO gaps and vertical attachment energies across the series support this picture. Electron attachment also produces strikingly different fragmentation patterns in the isolated molecules. In thiophene, the parent anion dominates over the anion formed by hydrogen loss; in benzothiophene, the two channels have comparable intensities, while in dibenzothiophene the parent anion is no longer detected. Fragmentation into smaller ions is also increasingly suppressed with molecular size. To understand these differences, we identify energetically accessible anionic structures that can be reached through bond cleavage and hydrogen migration. In contrast to the cyclic precursors, these structures are thermodynamically favorable for electron stabilization: the corresponding anionic geometries lie below their neutral counterparts. We also determine threshold energies for hydrogen loss, which produces closed-shell cyclic anions. The similar energetics of these competing processes do not, however, explain their markedly different experimental efficiencies. This points to an important role of reaction dynamics, which we plan to investigate in future work. Overall, the results show how increasing molecular complexity can strongly affect electron-driven reactivity in sulfur heterocycles.

References

[1] A. Pysanenko, I. S. Vinklárek, J. Fedor, M. Fárník, S. Bergmeister, V. Koštál, T. Nemirovich, and P. Jungwirth, "Gas phase $C_6H_6^-$ anion: Electronic stabilization by opening of the benzene ring," J. Chem. Phys. 157, 224306 (2022). https://doi.org/10.1063/5.0130998

P117

Heteroatom Doping for Enhanced Domain-Based Charge Transfer in Organic Dyes

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This work presents a computational investigation of domain-based charge transfer in π-conjugated organic dyes using localized orbitals of pair-coupled cluster doubles (pCCD). The equation-of-motion pCCD with singles (EOM-pCCD+S) approach enables quantitative and directional analysis of charge-transfer processes among the donor (D), bridge (B), and acceptor (A) domains. A series of 33 carbazole-based prototypical organic dyes was systematically designed by introducing nitrogen (N), oxygen (O), and sulfur (S) heteroatoms at three positions within the π-conjugated bridge, generating mono-, di-, and tri-doped structures. The results reveal that both the type and position of heteroatom doping play a crucial role in controlling the direction and magnitude of charge transfer. For mono-doped dyes, the forward donor-to-bridge-to-acceptor (D $\rightarrow$ B $\rightarrow$ A) charge transfer increases when the heteroatom is shifted toward the acceptor, with nitrogen-doped systems exhibiting the strongest charge-transfer character. In di-doped dyes, enhanced charge transfer is observed when the heteroatoms occupy the terminal bridge positions, particularly when nitrogen is positioned closer to the acceptor. Nitrogen-doped systems consistently exhibit greater forward charge transfer than their oxygen- and sulfur-doped counterparts. Among all investigated systems, the tri-nitrogen-doped dye (NNN) exhibits the highest directional D $\rightarrow$ B $\rightarrow$ A charge transfer of 42.6%, demonstrating the effectiveness of increasing nitrogen content within the π-bridge for promoting charge-transfer efficiency. These findings demonstrate that strategic heteroatom doping provides an effective approach for tuning directional charge-transfer pathways in π-conjugated organic dyes and offers valuable molecular-design principles for developing efficient organic sensitizers for dye-sensitized solar cells (DSSCs).

P118

REMP up your ADC

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The algebraic diagrammatic construction (ADC) family of methods is a robust and widely used approach for computing excited states of molecular systems of realistic size.[1] In particular, its formulation in an intermediate-state representation provides convenient access not only to excitation energies but also to excited-state and transition properties. As a perturbation-theoretical method, ADC fundamentally depends on the choice of the Hamiltonian partitioning. Traditionally, the Møller-Plesset (MP) partitioning has been employed. In this work, we introduce a novel ADC scheme consistent through third order, derived from the hybrid, parametrized REMP partitioning,[2] which was recently proposed and successfully applied to the ground state by R. Fink and coworkers. REMP is obtained by setting the unperturbed Hamiltonian to a mixture of the respective ones of the "Retaining the Excitation degree" (RE)[3] and MP perturbation-theoretical approaches. As such, REMP-ADC enables excited-state calculations to benefit from the same tunable mixing of MP and RE, and effectively bridges the already established standard ADC and RE-ADC methods.[4] REMP-ADC(2) and REMP-ADC(3) were implemented in adcc and benchmarked against high-level theoretical reference data. We find that REMP-ADC(3) provides highly accurate excitation energies, with a mean absolute error of 0.12 eV, improving upon the performance of both standard ADC(3) and RE-ADC(3), while maintaining the formal arithmetic scaling of N^6. Remarkably, this optimal performance is achieved with a 15% MP contribution in the mixing, which is consistent with the recommended mixing ratio for the REMP ground state.

References

[1] Dreuw, A.; Papapostolou, A.; Dempwolff, A. L., J. Phys. Chem. A, 2023, 127, 6635-6646.
[2] Behnle, S; Fink, R. F., J. Chem. Phys., 2019, 150, 124107.
[3] Fink, R. F., Chem. Phys. Lett., 2006, 428, 461-466.
[4] Leitner, J.; Dittmer, L. B.; Schneider, F.; Behnle, S.; Fink, R. F.; Dreuw, A., J. Chem. Phys., 2026, 164, 164101.

P119

Optimized grids for the real-space resolution of the identity in low-scaling GW

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The GW approximation is a Green's-function-based method for calculating charged excitations in molecules and materials [1]. Low-scaling formulations using Gaussian basis sets have enabled GW calculations for systems containing thousands of atoms [2]. These formulations employ the resolution of the identity (RI). However, the resulting three-index RI tensors still require substantial memory, and matrix operations involving these tensors remain computationally expensive. The real-space resolution of the identity (RI-RS) represents three-center integrals on a numerical real-space integration mesh [3,4]. In this representation, a three-index tensor is factorized into products of two-index matrices using numerical integration. Tensor operations involving three-index quantities can therefore be replaced by matrix operations involving only two-index quantities, leading to cubic computational scaling with system size for GW calculations with a small computational prefactor [5]. The computational cost and accuracy of RI-RS, however, depend critically on the selection of the real-space grid points for the numerical integration [4,5,6]. Until now, grids have been available primarily for the def2-TZVP basis set [4,5]. Here, we optimize compact atom-centered RI-RS grids for additional Gaussian basis sets, including the MOLOPT basis-set families used in CP2K [7,8]. We benchmark the resulting grids against conventional Coulomb-metric RI and determine the number of real-space points required to converge GW calculations. We further examine their transferability across different molecular environments and the numerical robustness of the grid optimization.

References

[1] D. Golze, M. Dvorak, and P. Rinke, Front. Chem. 7, 377 (2019).
[2] R. Pasquier, M. Graml, and J. Wilhelm, J. Chem. Theory Comput. 22, 540–557 (2026).
[3] R. M. Parrish, E. G. Hohenstein, N. F. Schunck, C. D. Sherrill, and T. J. Martínez, Phys. Rev. Lett. 111, 132505 (2013).
[4] I. Duchemin and X. Blase, J. Chem. Phys. 150, 174120 (2019).
[5] I. Duchemin and X. Blase, J. Chem. Theory Comput. 17, 2383–2393 (2021).
[6] F. A. Delesma et al., J. Chem. Phys. 160, 024118 (2024).
[7] T. D. Kühne et al., J. Chem. Phys. 152, 194103 (2020).
[8] M. Iannuzzi et al., J. Phys. Chem. B 130, 1237–1310 (2026)

P120

Simulating Chloroalkane-Methanol Binary Liquid Mixtures and Correlating with Femtosecond Thermal Lens Spectroscopy

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Understanding thermal transport in associated liquid mixtures requires molecular-level insight into the interactions governing energy dissipation1. In this work, molecular dynamics simulations are employed to investigate the thermal transport pathways in MeOH-chloroalkane binary mixtures (Dichloromethane, chloroform, and carbon tetrachloride), with femtosecond thermal lens spectroscopy serving as experimental validation2. Although the chloroalkanes exhibit negligible absorption at the excitation wavelength, their incorporation into methanol produces pronounced composition-dependent changes in the photothermal response. Molecular dynamics simulations performed using GROMACS-2023 reveal that the observed behavior originates from changes in the local liquid structure. Analysis of hydrogen-bond dynamics, O···Cl interactions, and intermolecular organization demonstrates that specific solvent-solvent interactions govern thermal energy transport. In particular, the MeOH-Carbon tetrachloride system exhibits solvophobic clustering arising from polar-nonpolar interactions, leading to distinct thermal transport characteristics compared with the other binary mixtures. Hydrogen-bond lifetime analysis indicates a highly dynamic local environment across all compositions, while structural correlations provide a molecular-level explanation for the experimentally observed photothermal response. The combined simulation and experimental approach establishes a direct relationship between the liquid's microscopic structure and macroscopic thermal transport.

P121

Understanding the Electronic Structure of a Radical-Functionalized Azobenzene MOST Candidate

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Azobenzenes substituted with Gomberg radicals present a challenging computational problem due to their pronounced multireference character and the strong dependence of the electronic structure on the molecular conformation and the choice of initial electronic state. We investigated the electronic and photochemical properties of these molecules to assess their photoswitchability and establish a computational strategy for treating systems beyond the reach of conventional multireference methods. A combination of different DFT-based methods was employed supported by NEVPT2 calculations to characterize the electronic ground state as well as low-lying excited states and explain experimental observations. Our results demonstrate that carefully selected single-reference approaches provide a qualitatively reliable description of complex open-shell systems that are otherwise computationally prohibitive.

P122

Development and benchmarking of vibe-qc, a molecular and periodic electronic-structure code targeting correlated methods for solids

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vibe-qc is a Python/C++17 electronic-structure code for molecules and solids. Its long-term goal is an ab initio Cyclic Cluster Model (AICCM) that brings correlated wavefunction methods to periodic systems. Because the CCM is a real-space Gamma-point method, in which a finite cluster subject to periodic boundary conditions represents the infinite crystal, the molecular method stack is a direct prerequisite: a correlated method available for molecules transfers to the periodic cluster without reformulation in reciprocal space. This contribution reports the current state of the code and benchmark results on molecular and periodic chemical systems. The molecular stack comprises restricted and unrestricted Hartree-Fock and Kohn-Sham DFT with analytic nuclear gradients and the full libxc functional library, Moller-Plesset theory (MP2, RI-MP2, SCS/SOS, and the B2PLYP and DSD-PBEP86 double hybrids), DLPNO-CCSD, CASSCF and internally contracted CASPT2, TDDFT, D3(BJ)/D4 dispersion, effective core potentials, and CPCM solvation. Semi-empirical methods are also implemented. Periodic boundary conditions are currently treated at the Hartree-Fock and Kohn-Sham DFT level through three routes: a BIPOLE/Ewald path with multi-k analytic gradients, GDF, and a GPW route, together with band structures and densities of states. Correctness is established by continuous benchmarking against established quantum-chemical codes. Molecular and periodic total energies are validated against reference implementations across the available methods, covering both self-consistent-field results and density-fitted variants. Representative tests span closed- and open-shell main-group molecules, geometry optimizations and harmonic frequencies, heavy-element complexes via ECPs, and ionic solids. Reproducible reference outputs and automatic per-job citation tracking are part of the standard workflow. The code is developed largely with AI coding assistance, which shortens the path from method specification to a testable implementation and shifts the limiting factor for reliability from implementation effort to verification. We therefore treat systematic benchmarking against reference data as the decisive activity and present the supporting results accordingly.vibe-qc is a Python/C++17 electronic-structure code for molecules and solids. Its long-term goal is an ab initio Cyclic Cluster Model (AICCM) that brings correlated wavefunction methods to periodic systems.

P123

High-Order Response Functions with Duschinsky Coupling and Finite Temperature: Application to Two-Dimensional Resonance Raman Spectroscopy

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Coherent multidimensional spectroscopy offers a wealth of information about ultrafast dynamical processes in molecular systems.[1] In this work, we derive closed-form expressions for multi-time correlation functions underlying higher-order nonlinear spectroscopic response functions within the harmonic approximation.[2] Our method includes displacements, frequency changes, and Duschinsky rotation between the vibrational modes of different electronic states while retaining polynomial scaling with the number of degrees of freedom. Finite temperature is accounted for without summation over vibrational eigenstates. To show the capabilities of our method, we apply it to calculate fifth-order two-dimensional resonance Raman (2DRR) [3,4] spectra of two-mode model systems and naphthalene. Comparison with the displaced harmonic oscillator model shows that Duschinsky rotation modifies the positions and relative intensities of diagonal and cross peaks. Overall, our results demonstrate that 2DRR spectroscopy is particularly sensitive to changes in normal coordinates between electronic states and can provide distinct signatures of Duschinsky coupling in molecular systems.

References

[1] Cho, M. Coherent Two-Dimensional Optical Spectroscopy. Chem. Rev. 2008, 108, 1331–1418.

[2] Philipp, L.N.; Begusic, T. High-Order Response Functions with Duschinsky Coupling and Finite Temperature: Application to Two-Dimensional Resonance Raman Spectroscopy, arXiv, https://arxiv.org/abs/2608.03879.

[3] Ren, H.; Biggs, J. D.; Mukamel, S. Two-dimensional stimulated ultraviolet resonance Raman spectra of tyrosine and tryptophan: A simulation study. J. Raman Spectrosc. 2013, 44, 544–559.

[4] Guo, Z.; Molesky, B. P.; Cheshire, T. P.; Moran, A. M. Two-Dimensional Resonance Raman Signatures of Vibronic Coherence Transfer in Chemical Reactions. Top. Curr. Chem. 2017, 375, 87.

P124

Combining DFT, MLP, and Monte-Carlo Approaches to Model Area-Selective Deposition Processes

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Area-selective atomic layer deposition (AS-ALD) has emerged as a key strategy for nanoscale patterning in advanced material design complementing conventional lithography processes. However, achieving high selectivity and process reliability requires precise control of the surface chemistry. Here, computational approaches are used to reveal the fundamental mechanisms governing reactivity and selectivity. These insights enable predictive tuning of inhibitor and precursor chemistry as well as substrate functionalization, guiding the rational design of selective deposition processes.
For inhibitor based AS-ALD several studies investigating the interactions of the inhibitor with the non-growth surface are available. However, comprehensive investigations of interactions between precursor molecules and the blocking layer are still sparsely available as a realistic model for the blocking layer must be derived at first. With the present work we want to highlight the currently available approaches to derive realistic blocking layer models. These approaches span a wide range of size scales and balance accuracy against computational cost. Density functional theory (DFT) is frequently employed to identify the most stable blocking layer configurations with high accuracy. However, its use is typically restricted to a small number of structural variations and relatively small surface cells, which limits direct comparison to experimental coverages and defect structures. Random sequential adsorption (RSA) simulations provide an extremely fast way to generate inhibitor layers models on large surface models containing hundreds to thousands of adsorbates. As RSA simulations naturally sample many distinct packing motifs on extended surfaces, they are often closest to experiment. Still, RSA simulations rely on the quality and physical realism of the initial input parameters. More recently, machine-learning potentials (MLP) have enabled molecular dynamics and metadynamics simulations that are orders of magnitude faster than DFT while approaching DFT accuracy when trained on suitable reference data. These approaches enable us to explore the dynamical evolution and restructuring of blocking layers on large systems and timescales, albeit at higher cost than static RSA simulations. We firmly believe that the interplay of all three methods will enable efficient as well as accurate modelling of interactions between precursors and blocking layers in future investigations.

P125

From Hydrostatic to Anisotropic Compression: The Development of two new Mechanochemical Simulation Methods

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Two new mechanochemical simulation methods that subject molecular systems to anisotropic strain are introduced. The implementations were developed for the Python-based chemical framework PySCF[1,2] for electronic structure simulations. Both methods are based on existing approaches for hydrostatic compression, namely the recently developed eXtended Hydrostatic Compression Force Field[3] (X-HCFF) method and the Gaussians on Surface Tesserae Simulate Hydrostatic Pressure[4–6] (GOSTSHYP) model. Anisotropy in the new approaches is introduced by placing a family of planes of a certain width in or along the van-der-Waals cavity of the system. The width of the family of planes determines the extension of the middle cylindrical part and the two outer half-shells. In the half-shells, the pressure is increased with distance to the origin of the planes' normal vector. This models these high-pressure sites as potential impact zones of a collision as for example in a ball milling setup. Therefore, the pressure is decreasing with distance to the origin of the plane in the middle part. On average, the same effective a priori pressure as in the original X-HCFF and GOSTSHYP approaches is applied, merely the distribution differs. With these new approaches named eXtended Anisotropic Compression Force Field (X-ACFF) and Gaussians On Surface Tesserae Simulate Anisotropic Strain using Half-shells (GOSTSASH), first insights into the underlying mechanisms occurring under anisotropic compression on the molecular level can be gained. Initial results for molecular model systems for both methods of hydrostatic and anisotropic compression are compared and presented.

References

[1] Q. Sun et al., Wiley Interdiscip. Rev. Comput. Mol. Sci. 2018, 8, e1340.
[2] Q. Sun et al., J. Chem. Phys. 2020, 153, 024109.
[3] T. Stauch, J. Chem. Phys. 2020, 153, 134503.
[4] M. Scheurer et al., J. Chem. Theory Comput. 2021, 17, 583–597.
[5] F. Zeller et al., J. Chem. Phys. 2022, 157, 184802.
[6] A. Pausch, F. Zeller, T. Neudecker, J. Chem. Theory Comput. 2025, 21, 747–761.

P126

SHARC meets Q-CHEM: interface for TDDFT dynamics with non-adiabatic and spin-orbit couplings for arbitrary states

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Recent developments in quantum chemistry have enabled reconstruction of spin-orbit couplings between all multiplet components based only on the information provided by a single individual transition, making evaluation of arbitrary spin–orbit couplings possible with single-reference methods. This approach implemented in Q-Chem is based on applying the Wigner–Eckart theorem for the triplet excitation operators, construction of the reduced excitation matrix elements with spin–orbit integrals, and applying the Wigner–Eckart theorem again to the spin–orbit reduced matrix elements. Single-reference methods have also progressed in treatment of the strong electron correlation. For example, the spin-flip approach implemented within a variety of methods enables an accurate treatment of spin near degeneracy. In this contribution, we interface these approaches to treat excited-state dynamics within the SHARC package. We demonstrate applicability of the approach to the reverse LIESST process of $[Fe(tpy)2]^{2+}$, requiring spin–orbit couplings between quintet and triplet state manifolds, which we treat with spin-flip TDDFT.

P127

Can a solvated electron reach the reactant?

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Electron-induced chemistry has been extensively explored in gases, solids, and even plasma, whereas corresponding processes in liquids remain comparatively understudied. A major experimental challenge is that electrons emitted from an electron gun can be strongly attenuated by the vapor phase above a liquid surface before reaching the solution. Here, we address this limitation by reducing the vapor-phase thickness using a continuous-flow liquid microjet and investigate whether incident electrons can penetrate the vapor layer and induce chemical reaction in solution. We chose electron-induced nitrate reduction as a model system. The resulting nitrite was detected through a well-established analytical reaction with 2,3-diaminonaphthalene, yielding a fluorescent triazole derivative. However, the experimental fluorescence spectra were crowded and difficult to assign reliably by inspection alone. To identify which molecular species contributed to the observed emission, we conducted excited-state calculations, vibronic effects modelling, excimer modelling, and pKa estimates by means of quantum chemistry. By combining the quantum chemical computations with a cleverly designed experiment, we address the central question: Can an incident electron reach the solvated reactant?

P128

Tensile Testing on an Atomistic Scale: Gold–Carbene Interfaces under Mechanical Strain

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Self-assembled monolayers (SAMs) of N-heterocyclic carbenes (NHCs) on gold offer superior chemical and thermal stability compared to their thiol-based counterparts, making them attractive for functionalized surfaces exposed to mechanical stress.[1] Despite advances in NHC–functionalized surfaces – both experimentally and theoretically, mechanistic insight into the effects of mechanical strain is still lacking. Here, we present periodic density functional theory (DFT) simulations of the force-induced detachment of two representative NHC classes, an imidazole derivative (IMe(Me) 2) and the stronger Σ donating and π accepting cyclic alkyl amino carbene (CAAC(Me) 2), adsorbed on Au(111).[2,3] By mapping the complete force-extension pathway, we find that both NHCs extract a two- atom gold cluster before an Au–Au bond in the gold lattice ruptures. Counterintuitively, despite CAAC(Me)2 exhibiting a stronger adsorption energy than IMe(Me)2 (by 0.28 eV), its rupture force is significantly lower by a factor of two. The force-extension pathways reveal distinct structural evolution of the carbene–Au interface, accompanied by differences in Au coordination and vertical fragmentation energies during detachment. Bader charge analysis shows the extracted Au–carbene fragment retains a nearly constant charge for IMe(Me)2 over much of the pulling range, whereas CAAC(Me)2 exhibits continuous charge depletion, reflecting distinct charge-transfer pathways at the two interfaces. These results provide the first atomistic insight into the mechano–response of NHC- functionalized gold interfaces and establish a framework for rationally designing more robust carbene-based surface coatings.

References

[1] G. Kaur et al., Chem. Commun. 2022, 58, 13188–13197.
[2] K. Denk, P. Sirsch, W. A. Herrmann, J. Organomet. Chem. 2002, 649, 219.
[3] V. Lavallo, Y. Canac, A. DeHope, B. Donnadieu, G. Bertrand, Angew. Chem. Int. Ed. 2005, 44, 7236.

P129

Spin–Orbit Coupling in TDDFT: From the Tamm–Dancoff to the Random Phase Approximation

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Many light-induced molecular processes involve transitions between states of different spin multiplicity, which are formally forbidden in non-relativistic quantum theory. Intersystem crossing, a nonradiative transition between such states, is important in photochemistry and photophysics, with applications spanning molecular photonics, energy conversion, biomedicine, and materials science. Spin–orbit coupling (SOC) provides a key mechanism for facilitating these otherwise spin-forbidden transitions, making the accurate evaluation of SOC matrix elements essential for describing such processes and their associated spectroscopic properties. Several approaches have been developed for the theoretical evaluation of SOC matrix elements. In this work, the Breit–Pauli spin–orbit Hamiltonian is treated within the spin–orbit mean-field (SOMF) approximation. For electronically excited states, time-dependent density functional theory (TDDFT) provides a computationally efficient framework applicable to relatively large molecular systems. SOC matrix elements within TDDFT are often evaluated using the Tamm–Dancoff approximation (TDA), in which only excitation amplitudes are retained. While the TDA simplifies the underlying eigenvalue problem, it neglects the de-excitation contributions included in the random phase approximation (RPA). Here, an existing TDA-based implementation is extended to the RPA formalism by solving the non-Hermitian Casida equation and incorporating both excitation and de-excitation amplitudes (X and Y) into the evaluation of SOC matrix elements. The implementation is validated and benchmarked using a set of 14 molecules. SOC matrix elements obtained with RPA are compared with their TDA counterparts to assess the influence of de-excitation contributions and identify cases in which the two approaches differ significantly. The results quantify the effect of going beyond the TDA and assess the relevance of de-excitation contributions to SOC matrix elements within the TDDFT formalism.

P130

Electron Dynamics and Spin Selectivity in Chiral Molecules and Interfaces

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Chiral-induced spin selectivity (CISS), i.e., the phenomenon by which electron transport through chiral molecules becomes spin-selective, has been well-observed in experiments [1,2]. However, a quantitative theoretical explanation remains challenging, especially because experimentally measured spin polarizations are orders of magnitude larger than expected from the intrinsic spin-orbit coupling of organic molecules alone [3,4].
In this work, we systematically investigate charge and spin dynamics in several experimental settings using relativistic real-time time-dependent density functional (RT-TDDFT) simulations. We observe chirality-dependent spin dynamics when a chiral molecule adsorbs on a ferromagnetic substrate. This can in turn modify the magnetic state of the ferromagnet, in agreement with CISS adsorption experiments [5]. We further demonstrate that RT-TDDFT yields spin-resolved photoelectron spectroscopy signals up to tens of percent, in unprecedented quantitative agreement with CISS experiments [6]. In addition, real-time pump-probe simulations qualitatively agree with time-resolved photoelectron circular dichroism (TR-PECD) experiments [7] and predict a spin polarization of up to ten percent in prospective spin-resolved TR-PECD experiments.
Overall, this work emphasizes that a dynamical, nonequilibrium description of the molecule-metal interface provides important insight into the interplay between molecular chirality and electron spin.

References

[1] Ray, K., Ananthavel, S.P., Waldeck, D.H. & Naaman, R. Science 283, 814–816 (1999).
[2] Naaman, R., Paltiel, Y. & Waldeck, D.H. J. Phys. Chem. Lett. 11, 3660–3666 (2020).
[3] Bloom, B.P., Paltiel, Y., Naaman, R. & Waldeck, D.H. Chem. Rev. 124, 1950–1991 (2024).
[4] Naskar, S., Mujica, V. & Herrmann, C. J. Phys. Chem. Lett. 14, 694–701 (2023).
[5] Ben Dor, O. et al. Nat Commun 8, (2017).
[6] Möllers, P.V., Göhler, B. & Zacharias, H. Israel Journal of Chemistry 62, (2022).
[7] Wanie, V. et al. Nature 630, 109–115 (2024).

P131

Molecular Insights into Alkene Hydrogenation with Manganese(I) NacNac Catalysts

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Manganese(I) complexes have emerged as earth-abundant alternatives to precious-metal catalysts for hydrogenation reactions,[1-2] yet efficient Mn(I) catalysts for alkene hydrogenation remain scarce and are hitherto predominantly based on phosphine, pincer, or N-heterocyclic carbene ligands.[3-5] Here, we investigate a novel Mn(I) NacNac catalyst system for alkene hydrogenation, combining experimental studies with DFT calculations to elucidate its mechanism and underlying reactivity.[6] The mechanism of alkene hydrogenation catalyzed by manganese(I) NacNac complexes was investigated using density functional theory (DFT) in conjunction with experimental mechanistic studies. The calculated catalytic cycle reveals an outer-sphere pathway involving a manganese hydride as the catalytically active species, with hydride transfer to the alkene identified as the rate-determining step. Further analysis provides insights into the intricate role of methanol in the catalytic cycle. Computed activation barriers across a range of substituted styrene substrates reproduce the experimentally observed reactivity trends, with electron-deficient double bonds showing enhanced reactivity. Furthermore, the differing catalytic activities of substituted NacNac manganese complexes can be rationalized based on their electronic and structural properties, providing a molecular-level understanding of catalyst and substrate effects. These results demonstrate the predictive potential of DFT for relating catalyst structure and substrate electronics to catalytic reactivity, providing a computational basis for the rational design and optimization of this promising type of hydrogenation catalysts.

1. P. Schlichter, C. Werlé, Synthesis 2022, 54, 517–534.
2. S. Elangovan, M. Garbe, H. Jiao et al., Angew. Chem. 2016, 128, 15590–15594.
3. S. Weber, B. Stöger, L. F. Veiros et al., ACS Catal. 2019, 9, 9715–9720.
4. N. F. Both, A. Spannenberg, H. Jiao et al., Angew. Chem. Int. Ed. 2023, 62, e202307987.
5. M. Hruzd, S. L. Kleynemeyer, C. Michon et al., Chem. Commun. 2025, 61, 2969–2972.
6. C. Hofer, P. E. Hartmann, A. D. Boese, N. C. Mösch-Zanetti, to be submitted

P132

Deciphering the Solvent Dependence of Hydrogen Generation from Seawater and Silicon Waste

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Hydrogen is a promising sustainable energy carrier and an essential reducing agent in chemical synthesis. This work explores a green, waste-valorizing route for hydrogen generation under mild conditions, developed in cooperation with our experimental collaborators.[1] The process utilizes untreated seawater, strongly basic catalysts (OH^-/$OtBu^-$), and industrial polymethylhydrosiloxane (PMHS) waste. Interestingly, experimental data reveal that dimethyl sulfoxide (DMSO) exerts a unique and massive accelerating effect, vastly outperforming other solvents. To elucidate the underlying reaction mechanism without conformational complexity, density functional theory (DFT) calculations were performed using a simplified $HSiMe_3$ model. The reaction proceeds via the addition of the base (OR^-) to the electrophilic silicon, forming a pentavalent intermediate ($[HSiMe_3(OR)]^-$). This intermediate prefers an axial configuration and subsequently generates H_2 via the formation of a dihydrogen bond ($H^{\delta-} \cdots H^{\delta+}$ interaction). This pathway aligns well with literature reports on related systems.[2] Overall, the computational data predict OH^- to be a significantly more effective catalyst than $OtBu^-$. However, modeling the unique accelerating effect of DMSO proved computationally challenging. As standard continuum models (COSMO, CPCM) and more sophisticated implicit approaches (SMD, DCOSMO-RS) were insufficient to reproduce the observed solvent effects, the methodology was extended to include explicit microsolvation. Preliminary evaluations indicate that neither purely implicit nor these combined implicit/explicit models currently capture the full extent of the observed solvent dynamics, underscoring the severe complexity of the specific solute-solvent interactions. Ultimately, this ongoing investigation aims to comprehensively rationalize the reaction mechanism, providing predictive insights to guide experimental development while serving as a benchmark study to evaluate the performance of various solvation strategies in highly specific environments.

[1] M. Hamdaoui, et al., Angew. Chem. Int. Ed. 2026, e9919359
[2] E. D. Voronova, et al., Inorg. Chem. 2020, 59, 17, 12240

P133

Wavelength-Controlled Fragmentation of Mixed-Functionalized Norrish Type-I Acylgermanes

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Novel Norrish-type I acylgermane photoinitiators with mixed functionalization were synthesized to control the formation of different radicals via the excitation wavelength generating various polymer microstructures [1]. Two different aryl-carbonyl chromophores were introduced in tetra-substituted acyl germanes to increase solubility and design absorption characteristics in different wavelength ranges.
TD-DFT (CAM-B3LYP/def2-TZVP) analysis assigned complementary UV/Vis bands to the mesitoyl and *para*-alkoxybenzoyl units enabling wavelength-selective excitation. Photo-CIDNP investigated the competition between two alternative α-cleavages of primary Ge/benzoyl radical pairs in the presence of a monomer quencher (butyl acrylate). By shifting the irradiation window, the balance between mesitoyl- and *para*-alkoxybenzoyl-derived radicals can be systematically tuned, which was confirmed by steady-state LED irradiation NMR experiments.
This effect was investigated in detail by TD-DFT computing the excited state dissociation curves of the different substituents into radicals. Conical intersections and crossings of the excited states before and after the triplet state transition state are responsible for the formation of different radicals depending on the absorbed wavelength. These PEG-acylgermanes combine polar media compatibility with light-tuned radical generation, converting small adjustments in wavelength into mechanistic control during photopolymerization.

Acknowledgements
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References
[1] *Angew. Chem. Int. Ed.* **2026**, *65*, e5559644, https://doi.org/10.1002/anie.5559644.

P134

Probing Oxyl Character and Reactivity in a Biomimetic Non-Heme Oxoiron(IV) Complex

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High-valent iron–oxygen species are key intermediates in enzymatic oxidation processes. While commonly described as Fe(IV)=O units, the electronic structure of these species can involve substantial ligand-centered radical character, leading toward an Fe(III)–oxyl description. Distinguishing between these electronic structures is essential for understanding their bonding and reactivity.

Recently, a non-heme oxoiron(IV) complex bearing the TMC-HOR ligand was synthesized as a biomimetic model of human catalase (CAT) and was shown to form 1-syn and 1-anti upon oxidation using different oxidants.[1] The two species exhibit remarkable different reactivity, with 1-syn decomposing upon addition of triethylamine (TEA), while 1-anti remains stable even in the presence of the stronger base triazabicyclodecene (TBD). Furthermore, 1-anti cannot be generated when the precursor is deprotonated prior to oxidation, suggesting an important role of the protonated alcohol functionality and its molecular environment.

In this study, the electronic structure of these complexes was investigated using density functional theory (DFT) and multireference methods. Particular emphasis was placed on determining whether the experimentally observed 1-anti species is adequately described as an Fe(IV)–oxo complex or exhibits significant Fe(III)–oxyl character. Computational electronic-structure analysis was complemented by calculated M¨ossbauer and resonance Raman parameters to enable comparison with experimental spectroscopic data. Redox and acid–base reactions were additionally investigated to explore the influence of ligand protonation and secondary-sphere interactions.

The combined electronic-structure and spectroscopic analysis reveals a clear distinction between the two isomers. While 1-syn is consistent with an Fe(IV)–oxo description, 1-anti exhibits pronounced Fe(III)–oxyl character, supported by substantial oxygen-centered spin density, Mossbauer isomer shifts, and its characteristic Fe–O vibrational properties. These findings establish 1-anti as a notable example of oxyl character in a biomimetic non-heme iron complex and demonstrate how subtle changes in coordination geometry and ligand environment can substantially alter the electronic structure of high-valent iron–oxygen species.

References

[1] D. Jyoti Barman, T. Lohmiller, S. Katz, M. Haumann, P. Hildebrandt, W. Nam, K. Ray, Chemistry – A European Journal 2025, 31, e202404468.

P135

Systematic Active-Space Construction for the Multiconfigurational Description of a Mn(I) Photosensitizer Candidate

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Developing photosensitizers based on earth-abundant first-row transition metals is an attractive strategy for more sustainable and cost-effective light-driven applications.[1] In this context, the Mn(I) complex $[Mn(HMTI)(CN)_2]^-$is investigated as a potential photosensitizer. This system is inspired by its isoelectronic Fe(II) analogue, $[Fe(HMTI)(CN)_2]$, for which a nanosecond metal-to-ligand charge-transfer (MLCT) excited-state lifetime has been reported.[2] Since experimental data are currently unavailable for the study Mn(I) complex, a reliable theoretical description of its excited-state electronic structure is particularly important. The excited-state manifold was first investigated using density functional theory (DFT) and time-dependent DFT (TD-DFT) calculations.TD-DFT reveals a dense manifold of low-lying singlet, triplet, and quintet states, with several triplet and selected quintet states showing significant spin contamination. This unbalanced description of the low-lying excited states highlights the limitations of a single-reference treatment and demonstrates the need for a multiconfigurational approach. Charge-transfer analysis further indicates that the most intense transition in the visible region has mixed MLCT, ligand-centered (LC), and metal-centered (MC) character, emphasizing the involvement of both metal- and ligand-centered orbitals. To obtain a balanced multiconfigurational description of these states, a sequence of CAS(10,12), CAS(14,14), RAS(14,14), and RAS(18,18) active spaces was investigated by systematically incorporating metal-centered σ and d orbitals together with ligand π and $\pi*$ orbitals. Despite the progressive enlargement of the active space, the low-lying singlet-state pattern remains broadly preserved, with the most intense transition consistently predicted between 1.97 and 2.05 eV. These results identify the orbital components required for the active space and provide a systematic framework for establishing a balanced multiconfigurational description of the low-lying excited states of $[Mn(HMTI)(CN)_2]^-$.

References

[1] C. Förster, K. Heinze, Chem. Soc. Rev. 49, 1057 (2020).
[2] J. T. Malme et al., J. Am. Chem. Soc. 145, 6029 (2023).

P136

Machine Learning-Guided Molecular Insights into Metal-Catalyzed Asymmetric Reactions: A Case Study with a Pharmaceutically Relevant Version of Suzuki-Miyaura Coupling

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Transition metal-catalyzed asymmetric transformation is a well-known tool for synthesis of a structurally complex molecular motif which can be used in drug discovery. Development of a room-temperature reaction with a high stereo-induction from easily accessible starting materials often requires exhaustive experimentation. The use of data science and statistical modelling dares to circumvent this problem by generating a large number of numerical data for reaction discovery in a time-bound fashion. However, it is still a challenge to identify the molecular features from a machine learning-guided study to control the reaction outcomes, as desired. Herein, we present a gradient boosting-guided protocol on a pharmaceutically relevant version of Rh-catalyzed Suzuki-Miyaura coupling reaction between racemic allyl halide substrates, non-racemic bis-phosphine-based biaryl catalysts, and boronic acid coupling partners. The predictive power of our final model is found to be reasonable with an RMSE of 4.28 – 6.51. The bite angle in the metal-catalyst geometry, the size and volume of the chiral pocket inside the bis-phosphine ligand, and molecular fitting of the coupling partner/allyl halide substrate inside the pocket are found to be of utmost importance for controlling the reaction output, i.e., enantiomeric excess. We have discovered a library of 1577 highly asymmetric ($\geq$ 98% ee) coupling reactions that have experimentally untested combinations of substrate, catalyst, coupling partner, time, and temperature. We have further extracted a subset of 191 reactions doable at 25°C without excessive heating. Our machine learning-guided approach assists a synthetic chemist in accessing easily-tunable molecular parameters to control the outcome of an asymmetric transformation, without the need for experimental trial-and-error methods.

P137

Spatially Resolved Vibrational Dynamics of Water in Aqueous $Ba(SCN)_2$ Using Machine-Learned Potentials

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Understanding how ions influence the dynamics of their surrounding water is important for describing solvation and electrolyte behavior at the molecular level. In this work, we study an aqueous $Ba(SCN)_2$ solution using spatially resolved spectroscopic analysis based on first-principles molecular dynamics (FPMD) simulations [1].
A direct first-principles treatment of the system over experimentally relevant timescales is computationally demanding. We therefore developed a workflow to train a set of machine-learned interatomic potentials (MLIPs) on reference structures, energies, forces, and maximally localized Wannier centers (MLWCs) obtained from short FPMD simulations. The energy and force labels are used to train a MACE [2] MLIP, which is then employed to generate a longer molecular-dynamics trajectory of the electrolyte. Separately, a machine-learning model based on the SCFNN framework [3] is trained on the MLWC positions obtained from the FPMD simulations and used to predict the Wannier centers of water molecules along the MACE trajectory.
We apply time- and space-resolved correlation analysis of the molecular dipoles to obtain radially resolved infrared absorption spectra of water molecules. In addition, radially resolved vibrational correlations are calculated from the atomic velocities. These complementary analyses allow us to characterize correlated and anticorrelated water motions as functions of both frequency and intermolecular distance.
Our results show clear differences between water molecules in the vicinity of Ba^{+2}, those near SCN^-, and those located farther from the ions. Even at a concentration of approximately 0.5 M, the ions noticeably influence the collective vibrational dynamics of the surrounding water and the hydrogen-bond network. Overall, the combination of FPMD and machine-learning models provides an efficient approach for extending first-principles-quality spectroscopic analysis to longer simulation times and for investigating ion-specific effects in aqueous electrolytes.

References

[1] Heyden M, Sun J, Funkner S, u.a. Dissecting the THz spectrum of liquid water from first principles via correlations in time and space. Proceedings Of The National Academy Of Sciences. 2010;107(27):12068-12073. doi:10.1073/pnas.0914885107
[2] Erhard LC, Schörghuber J, Comas-Vives A, Madsen GKH. How Realistic Are Idealized Copper Surfaces? A Machine Learning Study of Rough Copper–Water Interfaces. ACS Materials Au. 2026;6(2):379-389. doi:10.1021/acsmaterialsau.5c00174
[3] Gao A, Remsing RC. Self-consistent determination of long-range electrostatics

P138

Controlling Excited States of Anthracenes through Heteroatom Substitution

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Introducing heteroatoms into acenes is a powerful route to controlling their spectral properties, but a predictive picture that holds between chemically distinct heteroacene families has been hard to pin down. In this work, we apply the algebraic diagrammatic construction (ADC) scheme for the polarization propagator to a diverse set of aza-, phospha-, and halogen-substituted anthracenes, examining how both the nature and the ring position of a substituent govern the two lowest $\pi\pi^*$ states, $^1\mathrm{L}_s$ ($^1\mathrm{L}_b$) and $^1\mathrm{B}_b$. By tracing the diagonal elements of the ADC matrix down the perturbative hierarchy ADC(2) $\rightarrow$ ADC(1) $\rightarrow$ ADC(0), we isolate the physical origin of the intensity redistribution between these states. The orbital-energy gap between the participating particle–hole configurations emerges as the dominant factor controlling their mixing and, in turn, the oscillator strength. Substitution at the apical sites enlarges this gap and yields bright α-bands, while substitution at lateral sites keeps the configuration mixing balanced and transitions weak. Along the halogen series, keeping positions fixed, increasing electronegativity progressively intensifies the $^1\mathrm{L}_s$ band. Together these results provide a unified, physically transparent framework for rationally tuning excited-state brightness in substituted acenes.

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Combinatorial Structure Search and First-Principles Stability Analysis of Borophene Phases on Ag(111)

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Borophene, the two-dimensional allotrope of boron, shows an inherent structural polymorphism governed by the density and arrangement of hexagonal holes within its pristine triangular lattice. The substrate-dependent nature of this polymorphism under experimental conditions, observed on Ag(111)1,2, Ir(111)3, Au(111)4, Al(111)5, and Cu(111)6 motivates a systematic theoretical investigation of the thermodynamically accessible phase space. We introduce a combinatorial structure-search algorithm that performs substrate-constrained commensurate cell matching followed by hexagonal hole pattern enumeration and graph-based connectivity filtering and subsequent multi-registry stacking, without prior assumption of phase identity. All screened candidate structures are then relaxed using spin-polarised density functional theory at the PBE+D3(BJ) level of theory. Within a thermodynamic stability window of 0.2 eV above the ground-state $\beta 12$ phase, corresponding to experimental synthesis temperatures up to 800 K, the algorithm predicts several novel borophene polymorphs on Ag(111) besides the known experimental phases. A systematic energetic decomposition and Pearson correlation analysis highlight the role of intrinsic boron–boron bonding governing phase stability. Simulated constant-current STM fingerprinting further identifies three distinct morphological classes, each with characteristic bias-dependent properties. The methodology is broadly applicable to other combinations of two-dimensional materials on substrates where polymorphism is determined by vacancy patterning.

References

[1] Mannix, A. J. et al. Science 2015, 350, 1513–1516.
[2] Feng, B. et al. Nature Chemistry 2016, 8, 563–568.
[3] Vinogradov, N. A. et al. ACS Nano 2019, 13, 14511–14518.
[4] Kiraly, B. et al. ACS Nano 2019, 13, 3816–3822.
[5] Li, W. et al. Science Bulletin 2018, 63, 282–286.
[6] Wu, R. et al. Nature Nanotechnology 2019, 14, 44–49.

P140

Vibrational Spectroscopy of Molecules in High Pressure Enviroments with GOSTSHYP

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Vibrational spectroscopy in high pressure environments provides a sensitive probe of pressure-induced changes in molecular structure and bonding. Experimental advances employing the diamond anvil cell have made infrared and Raman spectroscopy under pressures of several Gigapascals increasingly accessible. Computational methods that can efficiently predict vibrational properties of molecules under such conditions are therefore highly desirable. The Gaussians On Surface Tesserae Simulate HYdrostatic Pressure (GOSTSHYP) model describes hydrostatic pressure by means of repulsive Gaussian potentials distributed over the surface of a molecular cavity, thereby providing an efficient molecular approach to quantum chemistry for high pressure environments. Recent applications have demonstrated its potential for vibrational spectroscopy, including the calculation of pressure-dependent Raman spectra of zwitterionic glycine in good agreement with experiment. However, vibrational frequency calculations with GOSTSHYP have so far relied on numerical second derivatives of the energy, which become computationally demanding even for moderately sized systems. The benefits of analytical second derivatives for high-pressure vibrational calculations have previously been demonstrated for the related X-HCFF approach. Here, we present the derivation, implementation, and application of an analytical nuclear Hessian for the GOSTSHYP model. Starting from the GOSTSHYP energy and its analytical gradient, we derive all Hessian contributions, including derivatives of the molecular cavity, the surface Gaussian potentials, and the corresponding three-center integrals. The resulting expressions have been implemented into the TURBOMOLE program suite and validated against numerical derivatives. First applications demonstrate the use of analytical GOSTSHYP Hessians for the efficient calculation of pressure-dependent harmonic vibrational frequencies and infrared spectra. This development enables routine vibrational analyses of molecular systems under hydrostatic pressure and substantially extends the spectroscopic capabilities of GOSTSHYP.

P141

The Aperiodic Defect Model: Treating Aperiodic Defects in Fully Periodic Systems with Wavefunction Methods

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Defects are aperiodic perturbations in otherwise periodic systems, yet their electronic structure is almost exclusively treated using periodically repeated supercells. This introduces artificial defect-defect interactions, and, for charged defects, the need for compensating background charges, resulting in slow convergence toward the thermodynamic limit. At the same time, the large supercells required by conventional approaches severely limit the applicability of systematically improvable wavefunction methods.
In this contribution, I will present the Aperiodic Defect Model (ADM) [1], a quantum embedding framework that resolves this conceptual inconsistency by embedding a single defect into the mean field of the pristine periodic system. This eliminates spurious periodicity without sacrificing the rigorous description of the crystalline environment and seamlessly incorporates the local electronic structure of the defect into the extended host. The resulting molecular-like Hamiltonian provides direct access to the powerful toolbox of molecular quantum chemistry, including coupled cluster, multireference, and excited-state approaches, whose application to periodic systems can be very difficult if feasible at all. Altogether, the ADM establishes a general framework for quantitative studies of defects and other localized phenomena in complex periodic materials.
The capabilities of ADM will be demonstrated for a negatively charged monovacancy in phosphorene [2], a demanding benchmark involving structural reconstruction, charge localization, and long-range polarization.

References

[1] R. Lavroff, D. Kats, L. Maschio, N. Bogdanov, A. Alavi, A. Alexandrova, and D. Usvyat, “Aperiodic defects in periodic solids“, J. Chem. Phys. 163, 084105 (2025).
[2] C. Rickert, L. Barta, E. Flach, D. Kats, and D. Usvyat, “Application of the aperiodic defect model to a negatively charged monovacancy in phosphorene“, J. Chem. Phys. 164, 234118 (2026).

P142

How good are excited-state geometries from subsystem TDDFT?

Anton Rikus (University of Münster), Johannes Neugebauer (University of Münster)

Subsystem time-dependent density-functional theory in the uncoupled approximation (uncoupled frozen-density embedding, FDEu) lowers the cost of excited-state calculations on a chromophore in an explicit environment and resolves the response into fragment contributions. Analytical gradients for FDEu have been available since 2016 and are implemented both in Serenity and in Koala (using the TDA), but the quality of the excited-state geometries they produce has not been assessed systematically. This poster reports such a benchmark.

For a set of chromophore–solvent clusters we optimize both the ground state and the lowest excited state at three levels of treatment: the isolated chromophore, the FDEu-embedded chromophore, and the full supersystem, which serves as the reference (TDA/PBE0/def2-TZVP, revAPBE non-additive kinetic and PBE non-additive exchange–correlation functionals). Every minimum is confirmed by frequency analysis.

FDEu reproduces supersystem vertical absorption energies closely (mean error +5 meV, MAE 23 meV), but the agreement deteriorates markedly at the emissive minimum (+48 meV, MAE 52 meV), with a systematic overestimation. To separate the two sources of this error, the emission error is telescoped through vertical single points in which each method is evaluated at the other's geometry. The two possible decompositions agree to better than 10 meV, so the split is well defined: an electronic contribution of +34 meV (MAE 35 meV) and a smaller, sign-indefinite geometric contribution of +14 meV (MAE 32 meV). The latter carries no definite sign because the optimizer minimizes the total excited-state energy rather than the excitation energy itself.

The outliers are as informative as the averages. Charge-transfer states and S1/S0 seam crossings have to be excluded, since the FDEu chromophore spectrum contains no inter-subsystem excitations and TDDFT cannot describe the near-degeneracy with its own reference. In one cluster the two methods converge to different intermolecular arrangements, hydrogen-bonded versus π-stacked, precisely because the supersystem stabilizes the stacked form through charge-transfer admixture that FDEu structurally cannot represent. Finally, control optimizations started from the supersystem geometry show that excited-state minima are more sensitive to the starting structure than ground-state ones, which sets a practical floor on the accuracy any such benchmark can claim.

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Re-purposing Optimization Trajectories: Do Hessians from Quasi-Newton Schemes Yield Reasonable Free Energies?

L. S. Rindt (Heidelberg University), P. Pracht (Heidelberg University)

Second derivatives of the potential energy with respect to nuclear coordinates, known as the nuclear Hessian, are essential for the calculation of molecular vibrational frequencies, thermodynamic properties, and for numerical geometry optimization procedures[1]. However, their computation is often prohibitively expensive, even at DFT level, for moderately sized organic and drug-like molecules. In this work, we explore whether the quasi-Newton framework can be leveraged to reconstruct approximate Hessians from gradients accumulated along the optimization trajectory, requiring only an initial guess Hessian.
Standard single-secant update schemes rarely improve absolute free energies over the pure guess Hessian on a selected dataset calculated with the semiempirical GFNn-xTB methods[2]. Nevertheless, certain variants outperform the guess Hessian for relative free energies on S30L, demonstrating that quasi-Newton update schemes can yield meaningful curvature improvements. We find that single-secant methods are fundamentally limited by their reliance on only the most recent secant condition, causing previously accumulated curvature information to be overwritten.
Multi-secant methods can alleviate this loss, but tend to struggle with redundancy inherent to optimization trajectories. To address these challenges, we propose a rigorous secant selection alongside a novel update scheme that restricts the Hessian corrections to the subspace in which the guess Hessian violates the secant conditions.

References
[1] H. B. Schlegel, WIRES Comput. Mol. Sci., 2011, 1, 790-809.
[2] C. Bannwarth, S. Grimme, et. al, WIRES Comput. Mol. Sci., 2021, 11, e1493.

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A Perturbative Triples Approach for EOM-CC-(D)EA/(D)IP Treatments of Excited States in Strong Magnetic Fields

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Magnetic White Dwarfs (WDs) provide atmospheric environments where the magnetic field strength can reach magnitudes around 200kT. Electronic structures of atoms and molecules in such environments can differ greatly from the weaker/field-free cases. Spectra from stellar objects enable studying of their atmospheric composition and provide glimpses into the behaviour of species in these environments. Theoretical calculations are needed for their assignment due to experimentally inaccessible conditions.[1] Methods such as finite-field(ff) coupled-cluster(CC) as well as equation-of-motion-CC(EOM-CC) theory[2-4] provide excitation energies and transition probabilities of appropriate quality. For predictions capable of comparison and assignment of observable spectra, triple excitations must be considered. Due to compounding factors leading to varying electronic characters, consistent targeting of states despite different magnetic field strengths and orientations is vital. We exploit the flexibility offered by different EOM-CC variants to target these states consistently. The suite of EOM-CC variants implemented as ff-methods is extended via the DEA/DIP approaches that add/remove two electrons to/from their CC reference states.This gives access to specific multiconfigurational states not attainable via existing EOM-CC variants. The implementation of one-electron transition properties like the transition dipole moments for analysis of available spectra are presented as well. The work of Matthews and Stanton[6] is extended to enable perturbative triples approaches for DEA and DIP, completing approximations at this level of theory for the suite of ff-EOM-CC variants available. This presentation discusses the results of an investigation on several absorption lines of the C+ cation, the presence of which has been hypothesised on WD-J2023, previously established as a hot DQ type(carbon-rich atmosphere) and estimated to exhibit temperatures >25.000 K. We combine different EOM-techniques to probe transitions between excited states, to capture the prominent multiconfigurational character of the target C+-states. DIP, which describes C2 in field-free studies well, is applied to C2 in a magnetic field to probe low-lying transitions.

References

[1] M.A.Hollands, S.Stopkowicz, M.-P.Kitsaras, F.Hampe, S.Blaschke, J.J.Hermes, Mon.Not.R.Astron.Soc., 520, 3560(2023)
[2] S.Stopkowicz, J.Gauss, K.K.Lange, E.I.Tellgren, T.Helgaker;J.Chem.Phys.146, 154105(2017)
[3] F.Hampe,S.Stopkowicz;J.Chem.Phys.146,154105(2017)
[4] M.-P.Kitsaras,L.Grazioli,S.Stopkowicz;J.Chem.Phys.160,094112(2024)
[5] D.A.Matthews,J.F.Stanton;J.Chem.Phys.282016;145(12):124102
[6] QCUMBRE, Quantum Chemical Utility enabling Magnetic-field dependent investigations Benefitting from Rigorous Electron-correlation treatment. F.Hampe,S.Stopkowicz,M.-P.Kitsaras,L.Grazioli,S.Blaschke,L.Monzel, with contributions from N.Groß,Ü.P.Yergün,C.-M.Röper,C.Harrer,M.Bonsignore,E.Paulus,S.Góger,www.qcumbre.org

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Quantum-classical non-adiabatic dynamics in extended solid-state materials: a SHARC-VASP approach

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Non-adiabatic dynamics in materials encompasses a broad range of phenomena, from charge-carrier transfer and recombination to light-induced structural and electronic changes. Accurately modeling these processes is challenging even for isolated molecules, and becomes more arduous in extended periodic systems. For molecules, Tully's surface hopping [1] is one of the simplest yet successful quantum-classical methods used to model excited-state dynamics. However, its application to extended periodic materials remains far less explored, mainly due to the intrinsic difficulty and high computational cost of accurately describing excited states in solids. For this reason, most state-of-the-art applications still rely on coarse approximations [2,3], such as a simple Kohn–Sham orbital picture to represent the material's excitations and the neglect of the electron-nuclear back-reaction, i.e., the effect of electronic excitation on nuclear dynamics.

By leveraging some of these concepts, we recently developed a new SHARC-VASP [4] interface that extends SHARC [5]—originally designed for molecular systems—to solid-state materials. We investigate the excited-state non-radiative relaxation of bulk silicon using different initial-condition sampling schemes based on either ground-state ab initio MD or Wigner quantum sampling, and discuss the resulting differences. We further explore machine-learning potentials trained on ab initio MD data to accelerate both MD and quantum sampling. Finally, we present an outlook on improving the current excited-state description by incorporating our recently developed restricted open-shell Kohn–Sham scheme in VASP [6], enabling a more accurate and efficient treatment of excited states in solids.

References

[1] J. C. Tully, *J. Chem. Phys.* **93**, 1061–1071 (1990).
[2] B. Smith, A. V. Akimov, *J. Phys.: Condens. Matter* **32**, 073001 (2019).
[3] O. V. Prezhdo, *Acc. Chem. Res.* **54**, 4239–4249 (2021).
[4] M. Romanelli, M. Sahre, G. Kresse, S. Mai, L. González, *ChemRxiv* 10.26434/chemrxiv.15004801 (2026).
[5] S. Mai et al., *SHARC 4.1: Surface Hopping Including Arbitrary Couplings*, DOI: 10.5281/zenodo.20927309 (2026).
[6] M. Sahre, M. Romanelli, M. Marsman, L. González, G. Kresse, *arXiv:2605.28637* (2026).

P146

Perturbative Spin-Orbit Coupling in CP2K for Molecules, Bulk Materials, and Surfaces

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Spin-orbit coupling (SOC) lifts electronic-state degeneracies, modifies ionization energies and band gaps, and gives rise to topological surface states. For large and structurally complex systems containing heavy elements, efficient formulations of SOC are needed. Here, we present a perturbative SOC implementation in CP2K[1] based on relativistic Hartwigsen-Goedecker-Hutter pseudopotentials.[2] The SOC matrix elements are evaluated analytically in a Gaussian atomic-orbital basis, and their sparsity enables linear-scaling construction with the number of atoms. We benchmark the method for the SOC81 molecular dataset[3] and for 88 crystals comprising alkali halides, elemental solids, and compound semiconductors.[4] For the molecules, the SOC-corrected highest occupied molecular-orbital energies agree with all-electron PBE reference values within 42 meV. For the crystals, spin-orbit splittings obtained with the TZV2P-MOLOPT basis agree with all-electron SOC reference calculations with mean absolute deviations below 20 meV. For a Bi2Te3 slab, the method also reproduces the SOC-induced topological Dirac surface state, with bands near the Dirac cone agreeing within 20 meV with plane-wave calculations.[5] These results establish the perturbative HGH-SOC implementation in CP2K as an accurate and computationally efficient approach for molecular, bulk, and surface electronic-structure calculations, with a formulation well suited to large atomistic models.

References

[1] M. Iannuzzi et al., "The CP2K Program Package Made Simple," J. Phys. Chem. B 130, 1237–1310 (2026).
[2] C. Hartwigsen, S. Goedecker, and J. Hutter, "Relativistic Separable Dual-Space Gaussian Pseudopotentials from H to Rn," Phys. Rev. B 58, 3641–3662 (1998).
[3] P. Scherpelz, M. Govoni, I. Hamada, and G. Galli, "Implementation and Validation of Fully Relativistic GW Calculations: Spin–Orbit Coupling in Molecules, Nanocrystals, and Solids," J. Chem. Theory Comput. 12, 3523–3544 (2016).
[4] W. P. Huhn and V. Blum, "One-Hundred-Three Compound Band-Structure Benchmark of Post-Self-Consistent Spin-Orbit Coupling Treatments in Density Functional Theory," Phys. Rev. Materials 1, 033803 (2017).
[5] B. Samal and J. Wilhelm, "Perturbative Spin–Orbit Coupling for Bulk and Surface States with Gaussian Pseudopotentials," 2026, in preparation.

P147

Girsanov path reweighting for core set Markov state models

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Girsanov path reweighting enables the recovery of unbiased dynamics from path generated under a modified potential energy function. The method is based on reweighting biased path ensemble averages by introducing the relative path probability of observing a path at the target potential compared to the biased potential. A fundamental limitation of Girsanov reweighting arises from the divergence of path probabilities between the two systems with increasing path length. This necessitates the use of kinetic models with high time resolution, such as core set Markov state models, to accurately evaluate reweighted dynamics. In this work, we present a reweighted estimator for core set Markov state models. The approach is implemented within the Deeptime package and validated on both a low-dimensional test system and a molecular system.

References

[1] S. Kieninger and B. G. Keller, J. Chem. Phys., 2021, 154, 12.
[2] S. Kieninger, S. Et.al. arXiv preprint arXiv:2303.14696.
[3] https://github.com/anyschaefer/deeptime , Accessed: 2026-08-14.
[4] J.-H. Prinz, et al. J. Chem. Phys., 2011, 134, 17.
[5] J.-L. Schäfer, B. G. Keller, J. Phys. Chem. B, 128, 25, 6014–6027, 2024.
[6] J.-O. Kapp-Joswig, B. G. Keller, J. Chem. Inf. Model., 63 (4), 1093–1098, 2023.
[7] Vanden-Eijnden, Eric, et al. J. Chem. Phys. 129.17 (2008). [8] O.Lemke, B. G. Keller. J. Chem. Phys. 145.16 (2016).

P148

FrayedEnds: Basis-Set-Free Quantum Chemistry

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Basis-set-free quantum chemistry represents the spatial part of the orbital basis without relying on predefined sets of basis functions. The most promi- nent choice, Alpert multiwavelets, formally spans an L^2 basis that is individu- ally truncated for each orbital. The key advantage of such a representation lies in its conceptual simplicity combined with guaranteed numerical precision: the user can essentially ignore the details of the underlying basis. In practice, however, this flexibility has long come at a cost: existing methods required time-consuming re-implementation in first-quantized form, and correlated methods in particular remained computationally expensive. The latter limita- tion was addressed by the direct determination of optimal pair-natural orbitals [1], which delivered a computationally affordable correlated method directly at the basis-set limit. Building on this, [2] decoupled the determination of the orbital basis from the many-body treatment, allowing general, off-the-shelf many-body methods to be used on a compact set of optimal orbitals. Finally, [3] formulated a self-consistent scheme in which the orbital basis and the many-body state are improved cyclically, refining one against the other. This essentially realizes a CASSCF approach with a given number of active orbitals obtained through a different algorithm in a formally complete L^2 basis. Within this fully variational scheme, nuclear and alchemical gradients can be computed directly using the Hellmann-Feynman theorem. By using a basis- set-free approach we open the possibility of defining a fully-differentiable quantum chemistry framework. The formalism can be used for both ground and excited states and shows promise for usage in tandem with quantum computing methods [4], where optimal active spaces are crucial to increasing accuracy while keeping the number of required qubits low.

References

[1] J. S. Kottmann, F. A. Bischoff, and E. F. Valeev (2020). J. Chem. Phys., doi.org/10.1063/1.5141880

[2] J. S. Kottmann, P. Schleich, T. Tamayo-Mendoza, and A. Aspuru-Guzik (2021). J. Phys. Chem. Lett., doi.org/10.1021/acs.jpclett.0c03410

[3] E. F. Valeev, R. J. Harrison, A. A. Holmes, C. C. Peterson, and D. A. Penchoff (2023). J. Chem. Theory Comput., doi.org/10.1021/acs.jctc.3c00732

[4] F. Langkabel, S. Knecht, and J. S. Kottmann (2026). APL Comput. Phys., doi.org/10.1063/5.0316008

P149

A Novel Variational Quantum Trajectory Approach to Nuclear Quantum Dynamics

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Trajectory-based approaches to molecular Quantum Dynamics have been revisited several times throughout the past decades due to their potential to overcome the curse of dimensionality and allow on-the-fly potential energy surface exploration. They have, however, remained less successful than widely used methods such as Multiconfigurational Time-Dependent Hartree (MCTDH) as consequence of numerical challenges or methodological deficiencies. In this work, we investigate the behaviour of a novel factorisation ansatz for the wavefunction that combines time-dependent Gaussian functions with variational expansion coefficients. We derive working equations to solve the time-dependent Schrödinger equation, and propose a dynamical gauge that accounts for the redundancy of the chosen ansatz with respect to the temporal degree of freedom. Taking advantage of the appealing properties of Gaussians, analytical expressions are obtained for all matrices required for the new propagation scheme. Furthermore, we present a proof-of-concept implementation of the method in Python, which explores its numerical efficiency and stability. The new approach is tested on various one-dimensional models, and ideas for the extension to multidimensional dynamics are presented.

P150

Calculation of Energy Potential Surfaces and Absorption Spectra of Rubidium-Noble Gas Mixtures

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Nuclear spin states of noble gas atoms, due to their protection from decoherence via a closed shell, were identified as suitable components for quantum technologies. This protection however begs the question how one can manipulate these nuclei states reliably. One suitable solution are optically controlled interactions via spin exchange collisions with alkali atoms.

The presented work investigates the potential energy curves and absorption spectra of the rubidium-noble-gas exciplexes RbNe, RbAr, RbKr and RbXe. The interaction was calculated using ab initio electronic-structure calculations, mainly Multi-Reference Configuration Interaction (MRCI) along with Spin-Orbit-Coupling calculations as well as hyperfine-splitting evaluations with DIRAC. The results were used to find possible quasi-bound states between the two atoms as well as their excitation energies. In the asymptote, energies show low deviation to the corresponding experimental values.

The ground- and excited-state energies in addition to the corresponding dipole moments were subsequently used to reconstruct the absorption spectra of such metallic vapor-noble gas mixtures. In order to obtain realistic results, the interatomic distances were weighted by an already known and widely used probability distribution, where the Boltzmann-factor is expanded by the area enclosing one of the atoms. This weighing is essential, since the energies of the exciplex depend strongly on the distance between the interacting atoms, which in general leads to a line-broadening.

Special attention was paid to the visibility of the so-called red- and blue-Satellites, i.e., sidebands which occur on the energetically low and high side of the principal absorption lines. Their appearance is related to extrema in the excitation energy curves and can therefore be used to gain further insight in the underlying exciplex interaction. The achieved results agree with available experimental data and provide further insight into the binding behaviour and spectroscopic properties of rubidium-noble-gas complexes.

P151

The Cavity-Born-Oppenheimer Framework: Electronic Reshaping and Multireference Effects under Vibrational Strong Coupling

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When molecules are placed in a non-classical photonic environment, such as an optical or nanoplasmonic cavity, strong light-matter coupling can form hybrid states called polaritons. Recent experiments have shown that, in the vibrational strong coupling (VSC) regime, when quantized cavity modes couple to molecular vibrational degrees of freedom, the resulting hybrid states can alter chemical and physical properties. The Cavity-Born-Oppenheimer (CBO) framework is a robust and extensible quantum chemistry approach that accurately captures these complex interactions [1,2]. In this contribution, I will provide a general introduction to the CBO framework and its recent methodological extensions, which are designed to capture VSC phenomena. Specifically, I will discuss the development of the CBO Configuration Interaction (CBO-CI) method, which provides a rigorous, ab initio foundation for describing ground and excited states under strong coupling. Since the cavity frequency in VSC is much lower than electronic transitions, the cavity does not hybridize electronic states into polaritons. Instead, the cavity vacuum induces a static, amplified reshaping of the electronic structure. This acts as a vacuum-field Stark-like effect, shifting dipoles and lifting degeneracies without electronic resonance. Furthermore, I will discuss the collective nature of VSC in molecular ensembles. We will explore the multireference character of these systems by analyzing Self-Consistent Field (SCF) stability within the CBO-Hartree-Fock (CBO-HF) approach. Under strong-coupling conditions, cavity-mediated interactions can induce critical SCF instabilities, resulting in cavity-induced, broken-symmetry CBO-HF solutions. These results emphasize the importance of multireference techniques for accurately characterizing the ground-state electronic structure, collective dynamics, and potential modifications to molecular property landscapes under VSC. To facilitate these investigations, all of the discussed methods have been implemented in the Psi4-CBO program package. Built upon the foundational architecture of the Psi4 quantum chemistry software and developed in Python, Psi4-CBO offers a comprehensive and accessible toolkit for understanding polaritonic chemistry and the nuanced molecular changes induced by quantum light.

References

[1] T. Schnappinger et al., Cavity Born-Oppenheimer Hartree-Fock Ansatz: Light-Matter Properties of Strongly Coupled Molecular Ensembles, J. Phys. Chem. Lett., 19, 8024 (2023).

[2] T. Schnappinger, M. Kowalewski, Molecular Polarizability under Vibrational Strong Coupling, J. Chem. Theory Comput., (2025).

P152

Can Triple Excitations Restore Spin Symmetry of Excited States within the ADC Framework?

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Algebraic diagrammatic construction (ADC) methods are accurate and systematically improvable quantum chemical methods for the description of excited states, built upon Møller–Plesset perturbation theory. The recently introduced fourth-order scheme, ADC(4), achieves a mean absolute error (MAE) of only 0.09 eV for singly excited states of closed-shell molecules, a substantial improvement over the MAEs of ADC(2) and ADC(3) of 0.20 eV and 0.23 eV, respectively.[1] Its performance for open-shell systems, however, has not been explored yet. Spin contamination (SC) remains a key challenge in open-shell calculations and a central criterion for assessing the accuracy of single-reference methods. Lower-order ADC methods are susceptible to SC,[2] and while higher-order correlation effects (triple and quadruple excitations) are known to reduce SC of excited states in coupled-cluster theory,[2] this aspect has not been studied in the context of ADC.
Here, we present the first implementation of the consistent second-order $\langle S^2 \rangle$ evaluation for ADC(4) and investigate whether spin symmetry can be restored through the inclusion of triple excitations. We find that SC is reduced systematically for unrestricted Hartree–Fock reference states: the MAE of $\langle S^2 \rangle$ decreases from 0.0126 au at the ADC(2) level to 0.0076 au at the ADC(4) level for first-order properties, and from 0.0078 au to only 0.0017 au for second-order properties, respectively. This shows that ADC methods have the potential to systematically remove SC.

References

[1] Leitner, J. et al., J. Chem. Phys. 157, 184101 (2022).
[2] Stahl, T. L. et al., J. Chem. Phys. 160, 204104 (2024), Stahl, T. L. et al., J. Chem. Phys. 164, 209901 (2026).

P153

Periodic g-xTB — Approaching DFT Accuracy for Periodic Systems at Semiempirical Cost

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It is well known that certain condensed systems and properties benefit greatly from the inclusion of non-local exchange. Examples include the description of electron-phonon couplings, polaron formation, and charge localization in general—phenomena central to the study of energy materials and heterogeneous catalysis. However, periodic non-local exchange interactions require dense k-point meshes and regularization techniques to reach convergence to the periodic limit. Due to the often prohibitive computational demand of non-local exchange, large-scale studies must frequently rely on lower-level semi-local density functionals such as PBE or r^{2}SCAN, or on machine-learning interatomic potentials trained on their data.

Here, we present the extension of the g-xTB method [1] to periodic systems both via a γ-point periodic algorithm implemented in the open-source `tblite` library, and a complete k-point periodic algorithm via an interface to CP2K. g-xTB improves upon the established GFNn-xTB family of semiempirical methods through several key advances, including a charge-adaptive minimal basis approaching double-ζ quality, the new and improved D5 dispersion correction, and a complete tight-binding expansion up to fourth order for a more accurate description of charged systems. However, the most important improvement is the inclusion of approximate non-local exchange in a range-separated fashion, imitating its reference method ωB97M-V/def2-TZVPPD. Consequently, g-xTB exhibits characteristic features of RSH functionals, including large frontier-orbital gaps, accurate reaction barrier heights, and balanced thermochemistry. Overall, it reaches DFT-level accuracy for molecular systems, as demonstrated by a WTMAD-2 of 7.6 kcal mol^{-1} on the GMTKN55 benchmark suite.

Preliminary results of the periodic implementation show improvements over GFN2-xTB, e.g., in X23b lattice energies (MAE 2.05 kcal/mol) and relative DMC-ICE13 polymorph energies (MAE 0.23 kcal/mol), cutting errors at least in half compared to GFN2-xTB. Overall, our periodic implementation demonstrates that g-xTB transfers well to extended systems, preserving the favorable properties of its RSH reference method. At the same time, the new terms introduce only a limited computational overhead, retaining comparable efficiency to other semiempirical methods. Periodic g-xTB therefore provides a fast and accurate approach to modeling extended systems beyond the semi-local approximation.

References

[1] T. Froitzheim, M. Müller, A. Hansen, S. Grimme, *ChemRxiv* (2025), DOI: `10.26434/chemrxiv-2025-bjxvt`.

P154

From Single α-Helices to Helical Bundles: Electronic Properties and Stability

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α-Helices and their higher-order assemblies are fundamental structural motifs in biology and promising building blocks for bioelectronic devices. Their intrinsic polarity and chirality make them particularly interesting for understanding charge transport and chiral-induced spin selectivity (CISS), yet the relationship between molecular sequence, helix length, and higher-order organization remains incompletely understood. In this work, we use density-functional tight-binding (DFTB+) calculations to investigate peptide systems across two structural levels, from individual α-helices to helical bundles. For single helices, we examine polyalanine and sequence-modified peptide models to investigate how helix length and side-chain chemistry influence their electronic structure. We find systematic changes in molecular orbital energies with increasing helix length, consistent with the additive macrodipole arising from the aligned peptide-bond dipoles and terminal groups. Analysis of the electronic structure further reveals how side chains and termini influence the spatial distribution of electronic states, providing insight into possible charge-transport pathways along the helix. These results demonstrate how molecular design can be used to tune the electronic properties of peptide-based molecular wires. We then extend this molecular picture to helical bundles, in which multiple α-helices assemble into supercoiled, chiral architectures. Using DFTB+, we investigate the energetic factors governing different bundling motifs and assess the contribution of dispersion interactions and solvation to their stability. This provides a connection between the intrinsic properties of individual helices and the structural organization of their higher-order assemblies. By combining electronic-structure analysis of individual helices with energetic and structural analysis of helical bundles, this work establishes a multiscale framework for understanding how molecular polarity, chirality, and intermolecular interactions shape peptide architectures. The results provide design principles for peptide-based electronic and chiral materials, with potential implications for bioelectronics, electronic hyperpolarization, and CISS.

P155

Conversion of Light Alkanes into Value–Added Products Over Boron–Based Catalysts

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The oxidative dehydrogenation (ODH) of light alkanes over metal–free boron–based catalysts has recently emerged as a promising strategy for the selective production of value–added chemicals. Among these catalysts, boron oxide (B_2O_3) [1] and borocarbonitride nanotubes (BCNNTs) [3] have attracted considerable attention for their remarkable catalytic performance. In 2023 [2], *ab initio* molecular dynamics (AIMD) simulations identified catalytically active structural motifs in molten B_2O_3 at 850 K. However, the catalytic roles of these active sites in the activation of methane and propane, as well as the mechanistic pathways governing product selectivity, remain poorly understood. In this work, we employ a combination of *ab initio* CASSCF/NEVPT2 and DFT calculations to investigate the reaction mechanisms of methane [4] and propane [5] over the liquid B_2O_3 active sites in the presence of singlet ($^1O_2$) and triplet ($^3O_2$) oxygen.
Our calculations reveal that B–B motifs are the most catalytically active sites, enabling efficient methane conversion into formaldehyde (HCHO) and propane into propylene and ethylene. In contrast, cyclic B–O–B motifs exhibit substantially higher activation barriers, indicating significantly lower catalytic activity. Orbital analyses further demonstrate distinct activation mechanisms: oxygen lone–pair donation governs reactivity at B–O–B motifs, whereas σ(B–B) bond donation drives bond activation at B–B sites. In addition, mechanistic investigations of methanol oxidation over BCNNTs provide detailed insights into the formation pathways of HCHO and the undesired over–oxidation product CO_2. These findings provide fundamental mechanistic insights for the rational design of efficient catalysts for selective light–alkane oxidation.

References

[1] J. Tian, J. Tan, Z. Zhang, P. Han, M. Yin, S. Wan, J. Lin, S. Wang, Y. Wang, *Nat. Commun.* **2020**, *11*, 5693.
[2] J. Tian, G. Collinge, S. F. Yuk, J. Lin, V.–A. Glezakou, M.–S. Lee, Y. Wang, R. Rousseau, *ACS Catal.* **2023**, *13*, 8219–8236.
[3] X. Zhang, P. Yan, J. Xu, F. Li, F. Herold, B. J. M. Etzold, P. Wang, D. S. Su, S. Lin, W. Qi, Z. Xie, *Sci. Adv.* **2020**, *6*, eaba5778.
[4] A. Sharma, P. Rawal, P. Gupta, *Chem. Eur. J.* **2026**, *32*, e202500000.
[5] A. Sharma, S. D. Choudhury, P. Gupta, *Chem. Eur. J.* (under review).

P156

Prokyon – An Open-Source Library for On-the-Fly Machine Learning of Interatomic Potentials

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Machine-learned interatomic potentials (MLIPs) have emerged as a transformative tool in computational chemistry, offering a bridge between the high accuracy of ab initio electronic structure methods and the efficiency of classical force fields. By learning the potential energy surface (PES) from quantum mechanical (commonly DFT) reference data, these models enable the simulation of complex systems at time and length scales previously deemed inaccessible.

A significant challenge remains in the reliability of state-of-the-art models when they encounter under-represented regions of chemical and configurational space in their reference data. Static ML models often struggle with extrapolation; when a simulation explores configurations far from the training set, the model may produce unphysical results without warning.

We present Prokyon, an open-source library to provide on-the-fly machine learning to existing electronic structure packages. Prokyon mitigates the risks of extrapolation by implementing active learning workflows based on Bayesian uncertainty quantification (UQ). The library monitors the model's predictive variance in real-time. Whenever the model's uncertainty exceeds a specified confidence threshold, the library triggers an automatic fallback to the underlying electronic structure method. This new data point is then used to refine the model on-the-fly, ensuring that the simulation remains physically grounded even in novel regions of the PES.

Our initial implementation targets CP2K as a primary host package, chosen for its high efficiency in generating reference data. Prokyon is designed for broad interoperability, with future integrations planned for other major codes including ORCA, CPMD, and Quantum Espresso. By providing a standardized interface for adaptive, Bayesian-guided learning, Prokyon aims to make robust, high-fidelity ML-based atomistic simulations more accessible to the broader computational chemistry community.

P157

Transcorrelation in Solid-State Matter

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Accurate ab initio electronic structure theory for solids remains a long-standing challenge: the most reliable correlated methods of quantum chemistry are limited by bottlenecks in system size, correlation treatment, and basis-set requirements. Pushing these methods toward larger, more strongly correlated systems while reaching the complete-basis-set limit at tractable cost is among the central challenges in the field today.
Transcorrelation (TC) is an explicitly correlated framework that addresses these problems. The second-quantized Hamiltonian is similarity-transformed with respect to a Jastrow factor — a real-space correlator borrowed from variational Monte Carlo that imposes the electron-electron cusp — folding correlation into an effective Hamiltonian with modified two- and three-body interactions. Since this transformation preserves the total energy while lowering the reference energy, less correlation remains to be recovered from virtual-space excitations. This compactification makes lower-level methods more accurate, while the explicit real-space cusp greatly accelerates basis-set convergence. A recently developed pseudopotential framework further accelerates the TC methods, particularly for large systems with heavy atoms [1-2].
Together, these features tackle all three bottlenecks for solid-state simulations: triple-zeta basis sets already suffice for convergence in bulk silicon; CCSD(T) reaches quantitative accuracy for point defects; and long-range Jastrow factors with TC embedding shrink the orbital space required to capture correlation.
In this talk I will present the current state of TC theory in solids, covering both method and applications: the periodic TC formalism and pseudopotentials in TC, followed by basis-set and method convergence in bulk silicon, recent formation energies of silicon self-interstitial defects [3], and magnetic interactions in cuprate and nickelate superconductors.

References
[1] Simula, Kristoffer, et al. JCTC 21.10 (2025): 5155-5170.
[2] Simula, Kristoffer, et al. Physical Review A 112.3 (2025): 032805.
[3] Simula, Kristoffer, et al. Physical Review B 113.19 (2026): 195125.

P158

Pressure induced luminescence shift in coordination compounds

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The phoshorescence of square-planar Pt (II) complexes is known to red-shift upon aggregation [1,2]. This effect can be exploited to control the emission wavelength of Pt (II) complexes by external pressure [3]. In this work, we investigate the pressure-dependent phosphorescence of a new square-planar Pt (II) complex both in solution and in MOFs by a combination of periodic DFT, QM/MM and classical molecular dynamics.
The monomeric Pt(II) complex shows a large red shift of 0.27 eV, when subjected to hydrostatic pressures ranging from 100 kbar to 170 kbar using DCM as solvent. The reason being shortening of Pt-ligand bonds with increasing pressure and interaction of monomeric Pt(II) complex with the solvent. The Pt(II) complex also shows a red shift of 0.7 eV, when embedded inside the ZIF-8 MOF upon isotropic compression from 0 bar to 50 kbar, again due to shortening of Pt-ligand bonds with increasing pressure and the electronic interaction of monomeric Pt(II) complex with the MOF.
The properties observed during these simulations make Pt (II) complexes promising candidates for pressure sensing and other stimuli-responsive photonic applications.

References

[1] I. Maisuls, C. Wang, M. E. Gutierrez Suburu, S. Wilde, C.-G. Daniliuc, D. Brünink, N. L. Doltsinis, S. Ostendorp, G. Wilde, J. Kösters, U. Resch-Genger, C. A. Strassert, Chem. Sci., 2021, 12, 3270.
[2] T. Theiss, S. Buss, I. Maisuls, R. Lopez-Arteaga, D. Brünink, J. Kösters, A. Hepp, N. L. Doltsinis, E. Weiss, C. A. Strassert, J. Am. Chem. Soc., 2023, 145, 3937.
[3] P. Steeger, T. Theiss, D. Schwab, I. Maisuls, V. Senthiappan Vellaiappan Uthayasurian, R. Schmidt, I. Kupenko, C. Sanchez-Valle, S. Michaelis de Vasconcellos, N. Doltsinis, C. Strassert, R. Bratschitsch, Nano Lett., 2025, 25, 2628.

P159

Constructing systematically convergent Gaussian basis sets for solids

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In Gaussian basis-set calculations for solids, the increasing condition number of the overlap matrix can lead to severe numerical difficulties and, in some cases, prevent calculations for heavier elements. This issue is particularly challenging for correlated relativistic calculations of solids using Gaussian orbitals.
In our approach, we systematically optimize atomic basis sets using even-tempered basis functions while introducing an additional penalty for excessive interatomic overlap. This strategy has the potential to provide a systematically improvable basis-set construction with a variable number of basis functions, enabling controlled convergence toward the basis-set limit and, potentially, basis-set extrapolation.
We will investigate this construction for both molecular and solid-state systems and assess its convergence, numerical stability, and transferability across different physical systems.

P160

DFT investigation of the effects of hydration on the reactivity of the low-valency distant binuclear vanadium species balanced in zeolites

A. Stepan Sklenak (Heyrovsky Institute/CZ)

The distant binuclear cationic sites were firstly identified using theoretical modeling in the context of the study of the N2O decomposition over the Fe(II) cation exchanged zeolites: ferrierite, the beta zeolite, and ZSM–5 [1]. We devised that the presence of the active sites formed by the distant binuclear Fe(II) centers explained the exceptional activity of Fe–ferrierite in comparison with the Fe–beta and Fe–ZSM–5 catalysts [1]. The first chemical step of the N2O decomposition is the formation of the Alpha–oxygen species [1-2] [i.e., (Fe(IV)=O)2+] which exhibits unique oxidation properties reflected in an outstanding activity in the oxidation of methane to methanol at room temperature [2]. Furthermore, we firstly predicted using the power of periodic DFT calculations that these distant binuclear cationic sites were able to split dioxygen to yield pairs of the distant Alpha–oxygen species. Subsequently, experiments were performed at room temperature and the theoretical prediction of a cleavage of dioxygen to give a pair of the temperature and the theoretical prediction of a cleavage of dioxygen to give a pair of the distant Alpha–oxygen atoms was confirmed experimentally and thus splitting dioxygen was discovered [3]. The effects of various transition metals [4] and zeolite topologies were investigated [5] employing periodic DFT were studied. In this contribution, we report on our periodic DFT investigation regarding the effects of hydration on the reactivity of low-valency early transition metal cations, namely vanadium.

References

[1] S. Sklenak, P. C. Andrikopoulos, B. Boekfa, B. Jansang, J. Nováková, L. Benco, T. Bucko, J. Hafner, J. Dedecek, Z. Sobalík, J. Catal., 272 (2010) 262.
[2]E. Tabor, M. Lemishka, Z. Sobalik, K. Mlekodaj, P. C. Andrikopoulos, J. Dedecek, S. Sklenak, Commun. Chem., 2 (2019) 71.
[3] E. Tabor, J. Dedecek, K. Mlekodaj, Z. Sobalik, P. C. Andrikopoulos, S. Sklenak, Sci. Adv., 6 (2020) eaaz9776.
[4] J. Dedecek, E. Tabor, P. C. Andrikopoulos, S. Sklenak, Int. J. Quantum Chem., 121 (2021) e26611.
[5] E. Tabor, M. Lemishka, J. E. Olszowka, K. Mlekodaj, J. Dedecek, P. C. Andrikopoulos, S. Sklenak, ACS Catal., 11 (2021) 2340.

P161

Neural Network Potentials of 2D Nanoconfined Water: Efficient Training Set Construction and Calculation of Dielectric Properties

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The structure and dynamics of water's hydrogen bonding network are still not fully understood, which is why water is the focus of many theoretical studies utilizing Molecular Dynamics (MD) simulations to investigate its properties. One way to compute the energies and forces to propagate the simulated trajectories is the usage of Neural Network Potentials (NNPs) which enable the representation of the potential energy surface with ab initio accuracy at a fraction of the computational cost. In nanoconfinement, a lot of properties of water change, e.g. dielectric properties. [1] Theoretical evaluations of water's dielectric properties often involve MD simulations in the pico- to nanosecond range – time scales which NNPs make feasible. Numerous approaches to train NNPs of nanoconfined water can be found in the literature;[2-5] however, they do not include a systematic evaluation of which confined structures need to be included in the training set to generate a NNP with the ability to extrapolate to arbitrary degrees of confinement. In this work, committee NNPs in combination with an Active Learning (AL) procedure are used to determine which water film thicknesses in 2D nanoconfinement and which water densities included in the dataset dominate the NNP performance and accuracy. Furthermore, an outlook on the usage of the constructed datasets to train multi-valued machine-learned dipolar models [6] is given which makes the calculation of static dielectric properties of both bulk and nanoconfined water possible: The multi-valued dipolar models enable MD simulations at fixed values of the dielectric displacement, reducing the simulation times needed in order for the dielectric properties to converge.

References

[1] T. Dufils et al., Chem. Sci. 2024,15, 516-527.
[2] W. Zhao et al., J. Phys. Chem. C 2022, 126, 25, 10546–10553.
[3] C. Schran et al., Proc. Natl. Acad. Sci. U.S.A. 2021, 118, 38, e2110077118.
[4] X. R. Advincula et al., ACS Nano 2025, 19, 18, 17728–17737.
[5] K. D. Fong et al., Nano Lett. 2024, 24, 16, 5024–5030.
[6] E. Stocco et al., npj Comput. Mater. 2025, 11, 304.

P162

Enhancing the Transferability of Machine Learning Potentials by Training on Atomic Properties

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Machine Learning Potentials (MLPs) have become an established tool for describing potential energy surfaces of complex systems, since they combine ab initio accuracy with reduced and linear-scaling computational costs. In most MLPs, linear scaling is achieved by leveraging the nearsightedness of electronic matter. Instead of directly predicting the total energy, these models yield atomic contributions that sum up to the total energy [1]. In practice, trained MLPs may exploit this freedom for error compensation. Using High-dimensional neural network potentials (HDNNPs) as an example, we show that the atomic energies predicted by these models are mathematically not unique, since the training process can introduce systematic offsets between different local structural motifs, which cancel out to yield the correct total energy. This compensation mechanism is robust as long as the relative abundance of different motifs in the application set closely matches that of the training set.

However, because the systematic offsets of the model depend on the statistical distribution of motifs encountered during training, systematic deviations—such as variations in overall composition or shifting structural characteristics—can prevent the cancellation of these offsets. This breakdown in error compensation can lead to shifts in the total energy, independent of the potential's intrinsic accuracy.

To remedy these uncertainties, we analyze the mechanisms of error compensation and propagation in detail, demonstrating results on various systems. We show that utilizing physically motivated atomic energy partitioning schemes during the training process can effectively mitigate these artificial errors and enhance the transferability and reliability of the resulting models.

References

[1] Behler, J.; Parrinello, M. Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces. Phys. Rev. Lett. 2007, 98 (14), 146401.

P163

A new Molecular Dynamics approach for treating spin-phonon effects in solids

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The most common working assumption in quantum chemistry is that the system being studied is stationary, which may not always be true. While for most systems, motion can just be a frustrating smear around the average behaviour, there are systems where dynamical nature and electronic structure are invariably entangled. One class of such problems is the spin-phonon systems where the spin-spin interactions modify the potential energy surface and the motion of atoms affects the spin-spin interactions. In particular we are studying spin-Peierls distortions in the antiferromagnetic isotropic Heisenberg model on one- and two-dimensional lattices where the coupling constants are position dependent. For a long time people have approached this problem with various Monte Carlo schemes, which required numerous assumptions and approximations. These approximations would severely limit the applicability of the method and quality of the resulting description. We present an alternative approach rooted in Molecular Dynamics. By expanding and combining Stochastic Series Expansion method to discrete imaginary time path integral formalism, we can use path integral molecular dynamics to calculate the statistical observables of these systems accounting for the nuclear quantum effects faster and while retaining the quality of the description. By understanding the decorrelation effect that the spin-phonon coupling has on the spin-spin interactions, we can use machine-learned forces trained on Monte Carlo data to break the curse of the exponential scaling and study the phase diagrams and different solid phases arising from the spin-phonon interactions. We showcase the method capturing the thermal and quantum effects on one dimensional lattices and the rich phase behaviour of two dimensional systems, with a particular focus on the honeycomb lattice.

P164

Fluorine-Specific π-Stacking Accelerates Ring-Opening Copolymerisation

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The copolymerisation reaction of fluorinated and non-fluorinated styrene oxide with phthalic anhydride yields degradable polymers with high molecular weights and recoverable fluorine. These polymers represent an important step towards a closed fluorine economy. [1-3] To understand the effect of fluorine substituents on reaction kinetics, we calculated Gibbs free energies of intermediates and transition states using a carefully evaluated computational prototol: Initial conformer sampling with the CREST/CENSO approach, PBEh-3c optimised geometric structures, single-point energies at the ωB97M-V/def2-TZVPPD level, and the COSMO-RS model (BP/TZVPD) to quantify the effect of solvation. The conformer sampling revealed π-stacking interactions, with the catalyst, additional monomers and the polymer chain. [1,2] Our results indicate that fluorine-specific π-stacking stabilises key intermediates and provides a molecular explanation for the experimentally observed rate acceleration, which occurs exclusively for heterofluorinated monomer combinations. Additionally we studied the degradation of these polymers as they allow to fully recover fluorine by methanolysis. [3] Fluorinated phthalic acid becomes defluorinated by methanolate, attacking as nucleophiles in an aromatic substitution reaction. Furthermore, methanolate attacks the backbone of the copolymer. Both corresponds to a very fast reaction, according to the surprisingly low barrier of 31.7 kJ/mol.

P165

Configurational Entropy of Mixing as a Spatially Resolved, Time-Dependent Observable

Tom Erik Steinkopf (Martin-Luther-Universität Halle-Wittenberg), Daniel Sebastiani (Martin-Luther-Universität Halle-Wittenberg)

Mixing and demixing govern the efficiency of many processes in chemistry, process engineering and materials science, yet their thermodynamic description rests predominantly on averaged models that capture local heterogeneity and non-equilibrium contributions only indirectly. This work determines the configurational entropy of mixing explicitly and with spatial resolution from molecular dynamics trajectories. Locally smoothed particle densities yield local mole fractions and hence an entropy density $s_{mix}(r)$, whose volume integral $\Delta S_{mix}(t)$ is a genuine time-dependent observable rather than an equilibrium average. Fitting a saturation model separates two physically distinct quantities for every component combination and temperature: the equilibrium plateau as the thermodynamic target, and a relaxation time characterising interdiffusion. Alkane/perfluoroalkane mixtures serve as the reference system, since their pronounced non-ideality exposes the deviation of the entropy of mixing while keeping enthalpic and entropic contributions separable.

Work to date has converted a functioning pipeline into a defined measurement protocol: separately equilibrated pure slabs docked into a cubic box with a sharp interface, a consistent equation of state throughout, and a production length that follows the mixing kinetics. These changes make the fitted parameters interpretable as converged physical quantities.

The continuing work targets the method itself and how molecular properties shape its output: the response to the smearing width σ and broadening kernel, toward a transferable calibration; the implicit role of molecular size and molar volume; the disproportionate weight of terminal ($CH_3 \rightarrow CF_3$) over interior ($CH_2 \rightarrow CF_2$) substitution; and the effect of fluorination architecture on non-ideality — validated against independent entropy routes.

P166

Reliable Redox-Potential Simulations of Proteins

M. Stier (University of Stuttgart/DE), J. Kästner (University of Stuttgart/DE)

The simulation of protein redox potentials is crucial for understanding various biological processes. However, such simulations remain challenging due to the complexity of these systems, arising from the electronic structure of the redox center and the dynamics of the surrounding protein and solvent environment. Moreover, the large system sizes preclude a full quantum mechanical (QM) treatment.
Commonly applied hybrid quantum mechanical/molecular mechanics (QM/MM) methods are often unsuitable due to nonpolarizable force fields, making pure QM calculations on cluster models a more reliable choice. However, truncating a solvated protein with several thousand atoms to a small cluster model introduces errors. Therefore, we recently proposed and tested a methodology for the simulation of redox potentials in proteins, as outlined below [1].
First, redox properties were computed using density functional theory (DFT) for a diverse set of ten proteins with cluster models of varying sizes, ranging from 200 to 1500 atoms, including full protein calculations. Our results show that cluster models containing approximately 500 atoms are sufficient, as truncation errors become smaller than the typical DFT errors beyond that size.
Second, the standard redox potentials of ten proteins were computed using the 500-atom cluster models within a snapshot-based approach grounded in Marcus theory. In our approach, QM/MM geometry optimizations were performed on snapshots from a classical molecular dynamics trajectory. The resulting QM/MM geometries were truncated to 500-atom cluster models, and redox potentials were computed from vertical reduction energies using DFT.
Comparison with experimental redox potentials revealed a mean absolute error (MAE) of 0.12 V, achieved without any reference calculations, which is only slightly larger than the previously estimated intrinsic DFT error. These findings are not only significant for improving redox potential predictions but are also broadly relevant for electronic structure calculations in complex systems involving charge changes, such as protonation/deprotonation, electron transfer, and electrocatalysis.

References
[1] Stier, M.; Kästner, J. J. Chem. Theory Comput. 2026, 22, 1, 334–357.

P167

Breaking Harmonics Walls: The Anharmonic Vibronic Coupling Approach in Trajectory Surface Hopping Simulations

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In this contribution, we present anharmonic vibronic coupling (AVC) Hamiltonians in combination with trajectory surface hopping (TSH) dynamics, providing a more flexible description of potential energy surfaces (PESs) than standard linear vibronic coupling (LVC) models. Furthermore, the reparametrization procedure has been automated, considerably reducing the manual effort traditionally associated with the reconstruction and fitting of multiple vibrational modes. LVC models constitute a widely employed framework for nonadiabatic quantum dynamics simulations due to their efficient parametrization of the PESs involved in excited-state relaxation processes. However, standard LVC models rely on the harmonic approximation and on a local expansion around a reference geometry, limiting their applicability in systems displaying large-amplitude motions, conformational flexibility, or strongly anharmonic vibrational modes governing the nonadiabatic dynamics. To overcome these limitations, some quantum dynamics studies have previously explored reparametrized vibronic Hamiltonians capable of reconstructing problematic regions of the PES beyond the harmonic approximation [1]. TSH provides a computationally cheaper alternative to fully quantum dynamical propagation. Nevertheless, standard on-the-fly TSH simulations remain computationally demanding due to the need to evaluate electronic energies, gradients, and couplings at every time step of the dynamics. For this reason, previous studies introduced the combination of standard LVC Hamiltonians with TSH dynamics as an efficient alternative to computationally demanding on-the-fly simulations [2–5]. However, these approaches still relied on the harmonic LVC approximation, thus inheriting the same limitations associated with anharmonic and flexible systems. Combining AVC Hamiltonians with TSH therefore preserves the computational efficiency characteristic of the standard LVC/TSH framework while considerably extending its applicability. Our results show that the proposed methodology is capable of reproducing the main features of photoinduced relaxation dynamics, including intersystem crossing, in systems where the standard LVC approach fails, while maintaining a very low computational cost.

References

[1] Lehr A. et al., Phys. Chem. Chem. Phys., 22, 25272-25283 (2020).
2 Plasser F. et al., Phys. Chem. Chem. Phys., 21, 57-69 (2019).
[3] Zobel J. P. et al., Acc. Chem. Res., 54, 3760-3771 (2021).
[4] Polonius, S. et al., J. Chem. Theory Comput., 19, 7171-7186 (2023).
[5] Farkhutdinova, D. et al., J. Phys. Chem. A, 129, 2655-2666 (2025).

P168

Coordination Corrected Enthalpies for the Thermodynamics of Ionic Materials

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Thermodynamic stability parameters, such as the formation enthalpy and energy above the convex hull, are fundamental descriptors for the discovery and design of new materials. Density Functional Theory (DFT) generally provides reliable thermodynamic predictions for systems in which compounds and their elemental reference phases have a similar chemical bonding nature, e.g., in metallic alloys. However, DFT accuracy worsens for chemically dissimilar systems such as ionic compounds. In these cases, the calculated formation enthalpies can deviate significantly from experiment, with mean absolute errors (MAE) of at least 100 meV/atom, leading to unreliable stability predictions. While advanced methods, such as hybrid functionals, may improve the accuracy, they are computationally expensive and still do not generally reach experimental accuracy. To overcome these limitations, empirical correction schemes such as the Fitted Elemental-phase Reference Energies (FERE) [1] and the Coordination-Corrected Enthalpy (CCE) [2,3] methods have been developed. FERE shifts the elemental reference energies but does not generally achieve experimental accuracy. In contrast, the CCE approach explicitly accounts for the local bonding environment through oxidation-state- and coordination-dependent corrections. Such an approach substantially improves the accuracy of formation enthalpies while maintaining the computational efficiency of standard DFT. In this work, we extend the CCE methodology within the AFLOW [4] framework to halides, chalcogenides, and pnictides, intending to significantly broaden the applicability of CCE beyond oxides. These classes of compounds cover a wide range of technologically relevant materials for energy storage, electronics, catalysis, and optoelectronics, and also exhibit diverse bonding environments. By deriving systematic bond-based correction parameters for these anion chemistries, we achieve more accurate formation enthalpies and more reliable predictions of thermodynamic stability. This extension enhances the robustness of high-throughput computational materials screening and provides a more general framework for accelerated materials discovery across chemically diverse inorganic compounds.

References

[1] V. Stevanović, et al., Phys. Rev. B. 2012, 85, 115104.
[2] R. Friedrich, et al., npj Comput. Mater., 2019,5,59.
[3] R. Friedrich, et al., Phys. Rev. Mater.,2021, 5, 043803.
[4] S. Divilov, et al., High Entropy Alloys and Mater., 2025, 3, 178.

P169

Interactions in N-Heterocyclic Carbene Monolayers on Cu(111) and Au(111) from Periodic Energy Decomposition Analysis and NOCV

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Understanding why an adsorbate binds where it does, and how tightly a self–assembled monolayer can pack, requires more than adsorption energies. Energy decomposition analysis for extended systems (pEDA) [1] combined with natural orbitals for chemical valence (NOCV) quantifies the driving forces of molecule–surface bonding under periodic boundary conditions. We apply it to *N*–heterocyclic carbenes (NHCs) on Cu(111), relevant to area–selective atomic layer deposition [2], using PBE–D4, periodic pEDA/NOCV in AMS–BAND, and scanning tunneling microscopy (STM).

I_{iPr}^{NHC}, the isopropyl–substituted carbene, chemisorbs by extracting a Cu terrace atom and binding to the resulting adatom ($\Delta E_{bond} = -289$ kJ/mol). Unusually for an adsorption, the bond is electronic rather than dispersive (dispersion 24%), with electrostatics contributing 72% of the attractive terms, while diffusion barriers of 3.6–4.7 kJ/mol leave the surface complex highly mobile. NOCV resolves σ–donation (65% of ΔE_{orb}) and π–backdonation into the NHC π^* (16%), consistent with a charge transfer of 0.25 e. Comparison with Au(111) shows identical bonding character but larger NOCV channels (σ–donation -256 on Au versus -176 on Cu, π–backdonation -56 versus -44 kJ/mol), following from the carbene–metal distance relative to the bulk nearest–neighbor spacing rather than its absolute value.

Periodic fragmentation models separate adsorbate–surface from lateral interactions. Lateral interactions are attractive ($\Delta E_{int} = -63$ kJ/mol) and 87% dispersion–driven, with only a small electronic contribution (-9 kJ/mol). The periodic analysis uncovers a cooperative, non–additive effect inaccessible to single–molecule models: direct Pauli repulsion between neighbors is small (29 kJ/mol), yet their presence raises the adsorbate–surface Pauli repulsion from 778 to 871 kJ/mol, setting the packing limit. Simulated STM images reproduce the experimental pattern and identify the wingtip rotamer. Resolving bonding channels and lateral interactions in one periodic framework thus provides a quantitative basis for tailoring carbene monolayers on coinage–metal surfaces.

References

[1] M. Raupach, R. Tonner, *J. Chem. Phys.* **2015**, *142*, 194105.
[2] A. Furlan et al., *Chem. Mater.* **2025**, *37*, 5753.

P170

Atomistic Insights into Si(100) Surface Passivation by Water: ReaxFF Molecular Dynamics and Fine-Tuned Machine Learning Interatomic Potential Simulations.

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The passivation of silicon surfaces is important in several industrial settings, such as for semiconductor device fabrication and photovoltaic applications. In this study, we use ReaxFF reactive molecular dynamics simulations combined with fine-tuned Machine Learning Interatomic Potential (MLIP) to extend our understanding of mechanism of Si(100) surface passivation in presence of water at various temperatures (300K,350K,383K). Simulation results indicate that the surface termination occurs predominantly through dissociative adsorption leading to the formation of silanol-terminated silica networks on the surface. At higher temperatures the atoms possess sufficient kinetic energy to reorganize into thermodynamically more stable structures as compared to room temperature where the hydroxylation is more disordered. The fine-tuned MLIP model confirms the key structural features obtained from ReaxFF, validating the latter as an approach for capturing essential passivation behavior. These findings provide atomistic insights into water-driven silicon passivation and may guide room-temperature passivation strategies. Our strategy also highlights a computationally efficient approach to the passivation of reactive surfaces, as a preliminary and necessary step in numerous theoretical studies. This protocol can be also utilized for wider varieties of solid electrolyte interphase system, to prepare a realistic, passivated surfaces for advance theoretical studies.

P171

Benchmarking Tensor Hypercontraction Techniques for Molecular Systems

Niklas Paulicks (University of Hamburg), Johannes Tölle (University of Hamburg)

Exploiting sparsity in the two-electron integral (ERI) tensor is a crucial step toward reducing the computational cost of many-body electronic structure methods. In recent years, a range of tensor factorization techniques, such as the tensor hypercontraction (THC) approach [1], have been developed to achieve this goal. In particular, linear optimization techniques based on least-squares (LS) fitting are of special interest because of their algorithmic simplicity, which may enable their application to large-scale systems.
Unfortunately, the landscape of these THC techniques remains highly fragmented, and many closely related methods have been developed independently in the literature, including LS-THC [1], interpolative separable density fitting (ISDF) [2], and RI-RS [3]. In this talk, I will present a unifying perspective on these approaches, which has enabled their efficient implementation in a newly developed library, pyTHC [4].
This new library enables, for the first time, a systematic comparison of the various linear THC techniques on an equal footing. To this end, I will present an in-depth assessment of these methods for the GMTKN55 benchmark database [5].

References
[1] R. M. Parrish, E. G. Hohenstein, T. J. Martínez, and C. D. Sherrill, J. Chem. Phys. 137, 224106 (2012)
[2] J. Lu, L. Ying, J. Comput. Phys. 302, 329 (2015)
[3] I. Duchemin, X. Blase, J. Chem. Phys. 150, 174120 (2019)
[4] N. Paulicks, J. Tölle, in preparation
[5] L. Goerigk, A. Hansen, C. Bauer, S. Ehrlich, A. Najibi, S. Grimme, Phys. Chem. Chem. Phys. 19, 32184 (2017)

P172

Machine-Learning Supported Reaction Mechanism Exploration of cis-trans Isomerisation in Retinal

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Rare events such as chemical reaction are transitions across an energy barrier. To rationalize the mechanism of such rare events and calculating their reaction rates, the search for an optimal reaction coordinates has long been very active field of research.[1] Using the minimum energy path (MEP) as reaction coordinate simply neglects entropic contributions and dynamic effects. Alternatively, the committor function offers a complementary perspective in which the transition state is defined as an isocommittor surface at which trajectories reach the reactant or product with equal probability. I will present our findings on the reaction mechanism of the cis-trans isomerization of retinal in gas phase and various solvation environments.[2] We apply the recently developed AIMMD algorithm,[3] where the committor of isomerization reaction is learned by a Neural Network (NN) function from short, dynamically unbiased trajectories that form a transition path ensemble (TPE) and are generated by two-way shooting molecular dynamics (MD) simulations. MD simulations are performed using self-developed machine-learned interaction potentials (MLIPs) and adaptively trained universal MLIPs to provide ab initio accuracy for comparatively low computational costs.[4] The low-dimensional approximation of committor function is obtained through symbolic regression, which yields a human-interpretable reaction mechanism in terms of a small set of internal degrees of freedom.
We observe a significant deviation of the TPE from underdamped Langevin dynamics with time scales in the range of hundreds of femtoseconds compared to the MEP obtained from overdamped simulations in the nanoseconds regime. Overdamped simulations do not show the asymmetric distribution of kinetic energy along different internal coordinates at various stages of the isomerization reaction, as seen in the TPE. Such differences must be taken into account when calculating accurate reaction rates with respect to experimental results, e.g., transition state theory.[5]

References

[1] Chen, H. et al., J. Chem. Theory Comput. 2023, 19, 4414–4426.
[2] Ghysbrecht, S. et al., J. Comput. Chem. 2025, 46, e27529.
[3] Jung, H. et al., Nat. Comput. Sci. 2023, 3, 334–345.
[4] Käser, S. et al., Digit. Discovery 2023, 2, 28–58.
[5] Ghysbrecht, S. et al., J. Comput. Chem. 2024, 45, 1390–1403.

P173

Implementations of Molecular Integral-Equation Solvation Models: From 6D-MOZ to RISM-SCF

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Solution-phase property computations need to balance physical rigor with computational efficiency. On one hand, explicit-solvent simulations provide a microscopic description of the solvent structure but require extensive sampling of the configurational space. On the other hand, continuum models are very efficient but most often do not describe solvent structure directly. Molecular integral-equation theories based on the Ornstein–Zernike equation [3] and approximate closure relations provide an intermediate approach, yielding equilibrium solvent distributions and thermodynamic quantities without trajectory-based sampling. We present the Modular and Open-source Implicit Solvation Toolbox (MOIST) [1,2], which provides implementations of molecular integral-equation methods including six-dimensional molecular Ornstein–Zernike theory (6D-MOZ) [3], as well as fast and robust implementations of one- and three-dimensional reference interaction site models (1D- and 3D-RISM) [4,5]. 6D-MOZ retains the relative translational and orientational dependence of molecular correlations and is systematically improvable through the intermolecular interaction potential and orientational resolution, but at substantial computational cost. For less computationally demanding applications, 1D-RISM provides bulk site–site solvent correlations, while 3D-RISM resolves solvent-site distributions around a solute [4,5]. MOIST also supports self-consistent coupling of RISM to electronic-structure methods, providing a molecularly resolved extension of continuum reaction-field models in which equilibrium solvent-site distributions generate a reaction-field contribution to the solute's electronic Hamiltonian. The coupling is available through embedded-cluster RISM-SCF (EC-RISM) [6] and the more efficient constrained-spatial-electron-density formulation, RISM-SCF-cSED [7]. MOIST itself is open-source [1,2] and will be available within the next major releases of ORCA and TURBOMOLE.

References

[1] L. Wittmann, A. Trajkovski, S. Grimme, manuscript in preparation (2026).
[2] MOIST, GitHub repository, 2026, https://github.com/lukaswittmann/moist
[3] L. Blum, A. J. Torruella, J. Chem. Phys. 1972, 56, 303–310.
[4] D. Chandler, H. C. Andersen, J. Chem. Phys. 1972, 57, 1930–1937.
[5] D. Beglov, B. Roux, J. Phys. Chem. B 1997, 101, 7821–7826.
[6] T. Kloss, J. Heil, S. M. Kast, J. Phys. Chem. B 2008, 112, 4337–4343.
[7] D. Yokogawa, Bull. Chem. Soc. Jpn. 2018, 91, 1540–1545.

P174

Excited-State Pair Natural Orbitals for Shallow System-Adapted Quantum Circuits

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Pair natural orbitals (PNO) offer an efficient way to represent molecular correlated wave functions. In prior works [1,2] it was shown that pair natural orbitals can be determined directly within a multiresolution analysis (MRA) framework and subsequently be employed for general many-electron methods. In this work, we extend the use of MRA-MP2-PNOs from ground- state calculations to excited states, in particular with respect to the design of shallow system-adapted quantum circuits. However, direct application showed that MRA-MP2-PNOs do not provide sufficient accuracy for excited states unless computational expensive orbital refinement [3,4] is performed in a state-averaged manner. To overcome these limitations, we extend the MP2- PNO basis by including configuration interaction singles (CIS) [5] vectors and the corresponding configuration interaction singles and doubles (CIS(D)) pair natural orbitals (CIS(D)-PNOs). We demonstrate the approach using the separable pair approximation (SPA) [2], which approximates the separable pair wave function as a tensor product of electron-pair functions. Since the SPA ansatz is formulated in terms of electron pairs, single excitations cannot be described directly. To correctly capture the excited states, we introduce suitable orbital rotations to keep the hardcore-bosonic character of the qubit state. The accuracy and performance of this approach are compared to the full configuration interaction (FCI) calculations for different molecules.

References

[1] J. S. Kottmann, F. A. Bischoff, and E. F. Valeev (2020).
J. Chem. Phys., doi.org/10.1063/1.5141880
[2] J. S. Kottmann and A. Aspuru-Guzik (2022).
Physical Review A, doi.org/10.1103/PhysRevA.105.032449
[3] E. F. Valeev, R. J. Harrison, A. A. Holmes, C. C. Peterson, and D. A. Penchoff (2023).
J. Chem. Theory Comput., doi.org/10.1021/acs.jctc.3c00732
[4] F. Langkabel, S. Knecht, and J. S. Kottmann (2026). APL Comput. Phys.,
doi.org/10.1063/5.0316008
[5] J. S. Kottmann, H. Sebastian, and F. A. Bischoff (2015). Physical Chemistry Chemical Physics, doi.org/10.1039/c5cp00345h

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Accurate treatment of hyperfine coupling in the diabatic potential model of HI

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The accurate treatment of relativistic couplings like spin–orbit (SO) coupling into diabatic potential models is an important but very difficult task. To this end, the Effective Relativistic Coupling by Asymptotic Representation (ERCAR) approach has been developed over the past years [1]. The central idea of ERCAR is the representation of the system's operators in an asymptotic diabatic basis consisting of direct products of atomic and fragment states. This allows to treat relativistic coupling operators like SO coupling analytically within the atomic basis states. This idea is extended to the incorporation of hyperfine coupling into the diabatic potential model.
Hyperfine coupling is due to the magnetic dipole and the contact interaction as well as the electric quadrupole interaction. The corresponding operators can be expressed in the angular moment operators $\hat{I}$ for nuclear spin and $\hat{J}$ for total angular momentum of the atomic fine structure states. The magnetic dipole and contact interactions both are proportional to $\hat{I} \cdot \hat{J}$ while the electric quadrupole operator is more complicated. The diabatic basis of an existing ERCAR model can be complemented by nuclear spinors and the hyperfine coupling operators are easily evaluated in that basis. Diagonalization of the resulting full diabatic ERCAR model yields the hyperfine structure energies and states for any molecular geometry of interest.
The new method is demonstrated using an existing accurate diabatic potential model for hydrogen iodide (HI) [2] to see the effects of hyperfine coupling. As a proof of principle study, the hyperfine coupling effect of the $^2P_{3/2}$ ground state and spin–orbit excited $^2P_{1/2}$ state of Iodine combined with the $^2S_{1/2}$ ground state of hydrogen are added to the ERCAR Hamiltonian and the effect of hyperfine coupling is investigated. Since both atoms have a non-vanishing nuclear spin, the hyperfine coupling needs to be treated in analogy to the multi-atom variant of the ERCAR approach [3].

References

[1] H. Ndome, R. Welsch, W. Eisfeld, *J. Chem. Phys.* **136**, 034103 (2012).
[2] N. Weike, A. Viel, W. Eisfeld, *J. Chem. Phys.* **159**, 244119 (2023).
[3] N. Weike, W. Eisfeld, *J. Chem. Phys.* **160**, 064104 (2024).

P176

Accelerating Large-Scale GW Calculations using Real-Space Resolution of Identity

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The G_0W_0 approximation provides accurate bandgaps and quasiparticle energies compared to standard density functional methods, but its $O(N^4)$ scaling and huge memory costs severely restrict its applicability. In recent years, various low-scaling formulations have reduced this computational cost to the cubic or even quadratic regime [1–4]. While many of these approaches have extended the accessible system size, they often suffer from large computational prefactors, numerical noise, uncontrolled truncation errors, or are restricted to non-periodic systems. A promising approach is the real-space resolution-of-identity (RI-RS) framework introduced by Duchemin et al. for non-periodic systems [2,3]. Unlike conventional Gaussian-based formulations, RI-RS projects orbital-pair densities onto a real-space grid, thereby eliminating the need for three- and four-center integral tensors while retaining sub-meV accuracy in quasiparticle energies.

Motivated by this, we extended the formalism and developed a massively parallel RI-RS G_0W_0 implementation in CP2K [5] capable of treating systems with more than 10,000 atoms. Built on the distributed block-compressed sparse row-matrix (DBCSR) library [6], this method aggressively exploits the inherent block-sparsity of atomic orbitals, polarizability matrices, and self-energy operators ensuring efficient memory usage and excellent parallel scalability on modern HPC architectures.

We demonstrate this approach on nanoscale non-periodic systems, including nanographene flakes and silicon clusters, reaching sizes previously inaccessible for deterministic G_0W_0. Compared to CP2K's existing low-scaling GW implementation using truncated RI [1], our RI-RS approach achieves speedups of up to 100× while maintaining sub-meV accuracy. Finally, we outline the extension of the RI-RS formalism to fully periodic boundaries for Γ-point calculations in large unit cells. This brings accurate quasiparticle calculations to unprecedented length scales and paves the way for large-scale Bethe–Salpeter equation (BSE) calculations for optical absorption spectra of large, disordered cells and associated excited-state gradients.

References

[1] M. Graml et al., *J. Chem. Theory Comput.* **20**, 2202 (2024).
[2] I. Duchemin et al., *J. Chem. Theory Comput.* **17**, 2383 (2021).
[3] F. A. Delesma et al., *J. Chem. Phys.* **160**, 024118 (2024).
[4] S. Phillip et al., *J. Chem. Theory Comput.* **22**, 3960 (2026).
[5] M. Iannuzzi et al., *J. Phys. Chem. B* **130**, 1237 (2026).
[6] U. Borštnik et al., *Parallel Comput.* **40**, 47 (2014).

P177

Molecular switches and machines

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Molecular switches and machines underlie many essential biological processes, including muscle contraction and peptide synthesis. Incorporating such motifs into synthetic materials is therefore a promising design strategy for the development of responsive functional materials. Mechanically interlocked molecules (MIMs) in combination with metal–organic frameworks (MOFs) are particularly well suited. The restricted configurational phase space of the MIM confines motion to well-defined pathways, while the MOF offers a tuneable scaffold that arranges the MIMs into a periodic lattice.
Our previous work has shown that neighbouring mechanically bonded rings (MBRs) within these frameworks can move in a coupled, cooperative fashion.(1) However, additional control of the dynamics by external stimuli has so far not been investigated. Here, we focus on deciphering the influence of mechanical compression and decompression of the framework on the coupled dynamics. Such an external stress can arise for example by gas uptake and release, or changes in environmental conditions.
To elucidate how the external mechanical force modulates the potential energy surface and thus influences the molecular-level dynamics, including the translational and rotational pathway, we performed molecular dynamics simulations. The mechano-responsivity of the shuttling and rotational dynamics of individual and coupled MBRs in UWDM-4(2) was studied by employing a machine-learned force field trained specifically for this system based on DFT calculations(3). Moreover, mechano-responsivity was studied at varying temperatures to reveal the coupling of both external stimuli.
The MD simulations demonstrate that slight compression of 1-3% increases the MBR shuttling rate and gives rise to a previously unobserved intermediate state. In this case the MBR resides between its two shuttling stations rather than moving directly between them. Detailed analysis of the trajectories indicates that compression of the framework introduces an altered translational pathway not accessible in the unperturbed framework. These atomistic simulations underscore the potential of mechano-responsivity for externally controlling and tuning the dynamics of molecular machines in functional materials.

References
[1] E. Kolodzeiski, S. Amirjalayer, Sci.Adv. 2022, 8(26)
[2] K. Zhu, C.A. O'Keefe, V.N. Vukotic, R.W. Schurko, S.J. Loeb, Nature Chem. 2015, 7(6), 514–9
[3] E. Kolodzeiski, S. Amirjalayer, J. Chem. Theory Comput. 2021, 17 (11), 7010– 7020

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Philicity at Play: Entropy, Enthalpy and Miscibility in Alkane–Perfluoroalkane Mixtures

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What drives molecular mixing and demixing at the atomic level remains a simple yet unresolved question. In a computational study of the elementary atomic interactions that govern phase behavior, we address this challenge using molecular dynamics simulations. Specifically, we investigate how the miscibility of an alkane–perfluoroalkane mixture responds to changes in the Lennard-Jones interaction potential parameters, that tune their lipophilic and fluorophilic characters. One particular challenge is measuring the instantaneous degree of mixing. We demonstrate that an approach based on a state function for the configurational entropy of mixing enables direct access to the system's state of mixing from the simulation trajectory.[1] Furthermore, we provide an enthalpic explanation of this specific mixing state by decomposing the total energy of mixing into effective interaction strengths.[2] Thereby, we elucidate how the complex interplay of direct and indirect enthalpic effects and entropic contributions induced by changes in the atomic interaction parameters determines the free energy of mixing, leading to either mixing or demixing.

References

[1] A. L. Upterworth, T. E. Steinkopf, D. Sebastiani, ChemPhysChem 2026, 27, e202500875.
[2] A. L. Upterworth, D. Sebastiani, J. Phys. Chem. A 2026, 130 (26), 5045–5056.

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Low-Spin Co(III) Stability in Cobalamin Derivatives Under Hydrostatic Pressure: A Computational Investigation

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The deep sea represents a challenging environment where life is subjected to hydrostatic pressure reaching up to 0.1 GPa at regular seafloor conditions and over 1.0 GPa near hydrothermal vents. Direct measurements are extremely difficult, so there is a genuine and important gap in our knowledge of how biomolecules behave under these conditions. Cobalamin (Vitamin B12) is a universal cobalt–corrin cofactor, but only a subset of bacteria and archaea can produce it. At ambient pressure, cobalt sits in a low-spin Co(III) d^6 singlet state, and while one crystal structure at 1.0 GPa has been reported, no spin-state study of cobalamin under pressure exists. Besides temperature and light, pressure can induce spin crossover (SCO) in transition metal complexes. Our goal is to detail the pressure response of its metal–ligand coordination sphere (Co–Ligand and Co–Imidazole), which are hypothesized to be the most sensitive to compression, by simulating the behavior of these systems under pressure.

For Co(III) d^6, 3D compression could drive a LS singlet ($t_{2g}^6 e_g^0$) $\rightarrow$ HS triplet or quintet ($t_{2g}^5 e_g^1$ / $t_{2g}^4 e_g^2$) transition, populating antibonding e_g orbitals and expecting longer, weaker metal–ligand bonds. Geometry optimizations and zero-point corrections were obtained at TPSSh(D3BJ)/def2-TZVP using the X-HCFF distortion model in Q-Chem 6.3, simulating pressures from 0.1 to 4.0 GPa, with energy corrections from the PV method. Multireference validation was performed at HCI-CASSCF(14,12)/def2-SVP using the HUMMR program.

The Co(III) low-spin singlet remained the ground state across all ligand variants tested up to 1.0 GPa, with no SCO detected until 4.0 GPa. The z-axis coordination bonds with Co are responsible for the main stress dissipation, while the corrin ring remains rigid. The triplet state is consistently more compressible than the singlet, as population of the antibonding e_g orbital directly weakens axial bonds. CN^- and Me-cobalamins show superior pressure resilience. Multireference calculations revealed that although DFT gives a good energy description, it misassigns spin density in high-spin states. For instance, while DFT delocalizes unpaired density onto the corrin ring at 2.5 GPa, HCI-CASSCF localizes it on cobalt d-orbitals (d_{xy}, d_{xz}, d_{yz}), highlighting the necessity of multireference methods for accurate spin-state descriptions in corrinoids in deep-sea environments.

P180

Simulations of Tricalcium Silicate-Water Interfaces using High-Dimensional Neural Network Potentials

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Calcium silicates are widely used as binding materials in cementitious systems where the hydration process is essential for developing the desired mechanical properties. Upon contact with water these minerals undergo a complex series of reactions of which the actual mechanisms remain mostly unclear. Insights into the behavior of water at calcium silicate surfaces at an atomistic level are therefore crucial to reach a better understanding of general hydration reactions of this class of materials. Tricalcium silicate is the most prominent species in context of cementitious systems and is characterized by a particularly high reactivity with water. These characteristics make it a good model system and particularly interesting to study. Molecular dynamics (MD) have been a key tool to examine processes along solid-liquid interfaces. However, conventional approaches faced a trade-off between accuracy and simulation length. Force fields struggle with description of the chemical reactions while ab initio methods are restricted to small systems. The advent of machine learning potentials (MLP), which allow to represent the energies and forces of electronic structure calculations at lower cost, offered a solution to these problems. In this study, we use High-Dimensional Neural Network Potentials (HDNNP),[1,2] a computational efficient MLP method, to investigate interfaces of liquid water with tricalcium silicate. We present high-accuracy, large-scale MD simulations of the (001) surface of M1 alite, one of the two major polymorphs, interacting with water at 300 K in nanosecond scale.

References

[1] J. Behler and M. Parrinello, Phys. Rev. Lett. 98, 146401 (2007).
[2] J. Behler, Chem. Rev. 121, 10037 (2021).

P181

Quantum Chemical Study of the Reactivity of a Platinum(II)nitrene with 1,4-Benzoquinone

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Nitrenes (R–N) are subvalent nitrogen compounds, therefore exhibiting remarkably high reactivity toward different substrates. Depending on the electronic environment they can feature a triplet or (multireference) singlet ground state. By substituting the organic rest R with a (heavy) transition metal like Pt, spin–orbit coupling (SOC) effects are introduced, potentially promoting multi–state reactivity [1–3].
We herein report a quantum–chemical mechanistic study of the reaction of a triplet platinum(II) nitrene with 1,4–benzoquinone (BQ) toward an aziridine as well as the rearrangement to an enamine, both of which were observed experimentally. The nitrene was generated *in situ* by photochemical dissociation of an azide precursor in toluene. The reaction with BQ proceeds through an electrophilic attack at one of the C=C double bonds ($\Delta^{\ddagger}G_{195} = 17$ kcal mol^{-1}). Subsequent transition to the singlet surface leads to the barrierless formation of the aziridine. Alternative reaction pathways were ruled out by kinetic arguments.
The subsequent rearrangement to the enamine was observed experimentally over the course of three days in toluene-d_8 at room temperature. Quantum–chemical results rule out a thermal process ($\Delta^{\ddagger}G_{298} = 47$ kcal mol^{-1}). Instead, the reaction occurs in the first excited state [4], in which an electron is excited from the nitrogen π–lone pair into the first π^*–orbital of the BQ scaffold. This enables a low–energy C–N bond dissociation and subsequent $1,2$–H shift ($\Delta^{\ddagger}G_{298} = 18$ kcal mol^{-1}) to the enamine.

References

[1] J. Sun, J. Abbenseth, H. Verplancke, M. Diefenbach, B. de Bruin, D. Hunger, C. Würtele, J. van Slageren, M. C. Holthausen, S. Schneider, *Nat. Chem.* **2020**, *12*, 1054–1059.
[2] M. Soleilhavoup, G. Bertrand, *Chem* **2020**, *6*, 1275–1282.
[3] M. C. Neben, N. Wegerich, T. A. Al Said, R. R. Thompson, S. Demeshko, K. Dollberg, I. Tkach, G. P. Van Trieste III, H. Verplancke, C. von Hänisch, M. C. Holthausen, D. C. Powers, A. Schnegg, S. Schneider, *J. Am. Chem. Soc.* **2025**, *147*, 5330–5339.
[4] S. Zheng, X. Chen, S. Liu, J. Lin, W. Chen, S. Ye, C.-Y. Su, Z. Li, *J. Am. Chem. Soc.* **2026**.

P182

Absorption spectra of N-heteroacenes

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Acenes are a class of organic molecules that have attracted considerable interest in both experimental and theoretical research due to their unique electronic structure and broad range of applications. Among these, their use as organic materials in electronic devices is particularly promising, where the properties of their low-lying excited states largely determine their suitability for specific applications. Chemical functionalization provides an effective means of tuning these optical properties, with nitrogen substitution representing a widely employed strategy. To systematically investigate the effect of nitrogen substitution on the excited states of N-heteroacenes, absorption spectra are simulated using time-dependent density functional theory (TD-DFT) at the TDA/CAM-B3LYP/6-311G* level of theory. Excitation energies, oscillator strengths and transition densities are computed to establish relationships between the excited states of N-heteroacenes and their unsubstituted acene counterparts, as well as to identify excitations unique to the substituted systems. In addition, the influence of the increasing acene length on the absorption spectra and the emergence of diradical character is examined.

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Spin-State Switching in Trinuclear Exchange-Coupled Systems

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Coordination compounds that exhibit switchable spin states are of great interest due to their potential applications in molecular spintronics. While spin-crossover (SCO) phenomena, which are reversible transitions between low-spin (LS) and high-spin (HS) states, have been extensively studied in mononuclear systems, the cooperative dynamics of these phenomena in polynuclear complexes remain largely unexplored.
This study focuses on trinuclear iron(II) complexes featuring a triangular arrangement of exchange-coupled metal centres with C_3 symmetry.[1] We employ a multi-method computational approach to analyse the electronic structure and spin state characteristics of the complexes. Geometry optimisations and vibrational frequency calculations are performed using density functional theory (DFT)[2]. The electronic structure and spin state behaviour are investigated, including the effects of spin-orbit coupling, using active space self-consistent field (CASSCF) and complete active space spin-orbit configuration interaction (CASOCI)[3] methods. By diamagnetic substitution, spin Hamiltonian parameters can be extracted for zero-field splittings and magnetic exchange couplings.
A Z/E isomerism of an imine bond within the ligand framework introduces structural variability, enabling a comparative analysis of the influence of the structure on the spin-state behaviour. Furthermore, the theoretical analysis suggests that E- and Z-isomers can be distinguished by their characteristic vibrational signatures in IR spectra. This provides a simple experimental method for differentiating between the two isomers. The calculations reveal that the exchange coupling between the iron centres is remarkably weak, whereas spin-orbit coupling is significant and must be explicitly accounted for in computational models.

References
[1] SPP 2491 Interactive Spin-State Switching: Spin-State Switching in Trinuclear Exchange-Coupled Systems, K. Fink, W. Plass, R. Röhlsberger, DFG project number: 561223321 (https://gepris.dfg.de/project/561223321?lang=en)
[2] TURBOMOLE V7.8 2023, a development of University of Karlsruhe and Forschungszentrum Karlsruhe GmbH, 1989-2007, TURBOMOLE GmbH, since 2007 (https://www.turbomole.org)
[3] T. Bodenstein, A. Heimermann, K. Fink, C. van Wüllen, ChemPhysChem 2022, 23, 1439–4235 (https://doi.org/10.1002/cphc.202100648)

P184

Extending an Automated Ensemble-Averaged Kinetics Framework for Reverse Intersystem Crossing in Flexible TADF Emitters

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Thermally activated delayed fluorescence (TADF) enables metal-free OLEDs with up to 100% internal quantum efficiency through reverse intersystem crossing (RISC) from dark triplet states to emissive singlet states.[1] In flexible donor-acceptor emitters, RISC rates are especially sensitive to geometry-dependent fluctuations of singlet-triplet gaps and spin-orbit couplings (SOCs). Rate models built on single optimized geometries can substantially misrepresent the kinetics of such flexible emitters by ignoring the conformational ensemble and the involvement of higher-lying triplet states.[2] We have developed an automated quantum-chemical workflow that restores both effects:[3] A conformational search on the triplet surface identifies representative macrostates that are subsequently expanded into Wigner-sampled nuclear ensembles. For each geometry, singlet-triplet gaps and SOC matrix elements are then calculated at the TD-DFT level. The global RISC rate follows from Fermi's golden rule as a Boltzmann-weighted sum over the thermally populated triplet channels. Because the couplings are evaluated between adiabatic states, each channel also implicitly carries second-order spin-vibronic coupling borrowed from the respective higher triplets. This recovers over an order of magnitude of the rate for the flexible emitter 3DPA2FBN while accurately reproducing the rate of the rigid benchmark emitter 4CzIPN. We are currently extending this framework along its two dominant error sources: First, TD-DFT overestimates the singlet-triplet gap in 3DPA2FBN, so we introduce a correction of the ensemble gaps against spin-component-scaled post-HF references through a Δ-machine learning approach that propagates the correlated correction across all sampled geometries at a fraction of the cost. Second, the harmonic Wigner approximation poorly represents the large-amplitude, anharmonic torsional modes that mediate RISC in flexible emitters. We therefore replace it with anharmonic sampling from various molecular dynamics approaches based on semiempirical, ab initio, and machine learning methods, while ensuring the sampled geometries remain compatible with the subsequent excited state calculations.

References

[1] C. Adachi, Jpn. J. Appl. Phys. 2014, 53, 060101.
[2] D. Beer, J. Weiser, T. Gabler et al., arXiv preprint 2026, arXiv:2604.08483.
[3] J. Weiser, C. Wiebeler, in preparation.

P185

Interactive JEDI Strain Analysis as a Pre-Screening Tool for Mechanophore Design

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Predicting which bond in a mechanophore activates under a given pulling geometry is central to their design, and is typically approached by elongating a structure along a chosen coordinate and relaxing at each step until rupture. We present an interactive implementation of the JEDI (Judgement of Energy DIstribution) strain-energy decomposition,[1,2] intended as an inexpensive pre-screening instrument for the design of mechanophores. JEDI partitions the harmonic deformation energy over redundant internal coordinates, giving a per-bond strain measure rendered directly as a color map on the molecular structure while the user deforms it. Porting the analysis to a C++ core exposed to Python, together with targeted optimization of the per-frame path, reduces the cost of a single strain evaluation to well under a millisecond, comfortably within a 60 Hz frame budget. The engine requires only a relaxed reference geometry, its Hessian, and a stream of distorted structures to evaluate against them; it is intended to be attached to any interactive chemistry environment. We demonstrate it within SCINE Heron,[3] using Sparrow as the semiempirical driver and with optional haptic force feedback. Emulating a CoGEF (Constrained Geometries simulate External Force) distortion of trans-1,2-diethylbenzocyclobutene and 1,1-Dichloro-2,3-diethylcyclopropane at the PM6 level, the redistribution of strain can be followed while the structure is manipulated, up to the rupture of the bond reported in published calculations.[4] Strain analysis is no longer the limiting factor; the electronic structure calculations now determine the achievable frame rate. Development is ongoing, with user-defined pulling directions and faster electronic-structure backends as the immediate next steps.

References

[1] T. Stauch, A. Dreuw, J. Chem. Phys. 2014, 140, 134107.
[2] H. Wang, S. Benter, W. Dononelli, T. Neudecker, J. Chem. Phys. 2024, 160, 152501.
[3] T. Weymuth et al., J. Chem. Phys. 2024, 160, 222501.
[4] I. M. Klein et al., J. Am. Chem. Soc. 2020, 142, 16364.

P186

Molecular Dynamics and Computational Spectroscopy for Systems in Soft Condensed Phase

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Molecular dynamics is a wide-spread simulation technique, which (i) we have employed to elucidate photophysical processes based on potentials from electronic structure calculations [1] and (ii) can be used to obtain structural insights into biological macromolecules like proteins and DNA using classical force fields [2, 3]. Complementary to this, computational molecular spectroscopy allows to establish structure-property relationships. For example, our calculations helped to assign chemical shifts and hyperfine couplings to atoms of chromophores [4]. We have also combined both in the framework of quantum mechanics/molecular mechanics (QM/MM) simulations to understand the molecular origin of the difference in optical absorption between the photoproduct (Pg) and dark state (Pr) of the photoreceptor protein Slr1393 [5]. Furthermore, our QM/MM-optimized structures provided further details of structural rearrangements in the binding pocket [6]. Currently, we are working on unraveling the mechanisms leading to thermally activated delayed fluorescence in donor-acceptor cyanoarenes [7] and on the parametrization of metal ions [8], which is a prerequisite for classical MD simulations.

References

[1] C. Wiebeler et al., J. Phys. Chem. Lett. 8, 1086–1092 (2017).
[2] S. Dürauer et al., Nat. Commun. 16, 5422 (2025).
[3] K. R. Storm et al., Nucl. Acids Res. 54, 5, gkag192 (2026).
[4] P. Kurle-Tucholski et al., J. Magn. Reson. 353, 107497 (2023).
[5] C. Wiebeler et al., Angew. Chemie Int. Ed. 58, 1934–1938 (2019).
[6] L. Köhler et al., Chem. Sci. 14, 6295–6308 (2023).
[7] A. Morgenstern et al., Adv. Opt. Mater. 14, 2, e01504 (2026).
[8] C. Wiebeler et al., doi: https://doi.org/10.64898/2026.03.31.715553 (Preprint).

P187

Automated Reaction detection with different Bond Order Schemes

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Automated reaction discovery using ab initio molecular dynamics (AIMD) is a field in computational and theoretical chemistry. It aims to identify various reaction pathways originating from a defined molecular system and to build comprehensive reaction networks. A central and crucial requirement for constructing these networks is the reliable automated reaction detection from resulting trajectories. Conventional distance-based criteria and simple bond order schemes are often insufficient to accurately identify bond breaking and bond forming events, as both neglect the underlying electronic structure. This limitation becomes even more pronounced in transition metal systems, where the complex and flexible nature of metal ligand interactions further challenges such simplified approaches. A more sophisticated description of bonding is therefore essential for reliable reaction discovery. In this study, we present a systematic comparison of various bond order analysis methods to evaluate their performance in automated reaction detection. We investigate Mayer, Wiberg, Mulliken, Fuzzy and Laplacian bond orders, as well as approaches derived from Intrinsic Bond Orbitals (IBOs) and Natural Bond Orbitals (NBOs). The main focus is on evaluating the consistency, sensitivity, and interpretability of each bond order scheme in identifying bond breaking and bond formation events. Special attention is given to challenging systems involving transition metal complexes. The results illuminate the strengths and inherent limitations of the respective approaches, they serve as a well-founded guideline for selecting the optimal bond order scheme for robust, automated reaction discovery workflows in organic and organometallic chemistry.

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Improving the scaling of AC0 with laplace-THC

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This work presents an efficient approach to capture dynamic electron correlation for a complete active space self-consistent field (CASSCF) wave function as a reference. The basis for this work is the linearized variant of the adiabatic connection approximation (AC0) [1]. This method only involves the use of one- and two-electron density matrices, resulting in a more favorable scaling with active space size than for example NEVPT2. To enhance the scaling of AC0 with basis set size, the Tensor Hypercontraction (THC) [2] method is used in combination with a Laplace Transform of the energy denominator [3]. Laplace-THC-AC0 is an approach that can efficiently calculate dynamic electron correlation for large systems and big active spaces, making it a promising complement to our recently developed CFG-HCI approach [4].

References

[1] Y. Guo, K. Pernal, Faraday Discuss. 2024, 254, 332–358.
[2] a) E. G. Hohenstein, R. M. Parrish, T. J. Martínez, J. Chem. Phys., 2012,137(4), 044103, b) R. M. Parrish, E. G. Hohenstein,, T. J. Martínez, J. Chem. Phys., 2012,137(22): 224106.
[3] a) M. Häser, J. Almlöf, J. Chem. Phys., 1992, 96(1), 489–494. b) M. Kobayashi, H. Nakai, Chem. Phys. Lett., 2006, 420(1-3), 250-255.
[4] M. Ugandi, M. Roemelt, J. Comput. Chem., 2023, 44(31), 2374-2390.

P189

A Minimally Empirical and Self-Consistent Implicit Solvation Model for Robust Quantum Chemistry in Solution

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Implicit solvation models are often the workhorse in computational quantum chemistry, but their reliability can deteriorate for more complex systems [1]. We present GEMS, a fully self-consistent and general implicit solvation model that aims to enable robust quantum-chemical computations in solution and is constructed from physically motivated and interpretable components. The solvent is represented by a simple density model derived from basic quantum-mechanical properties of the solvent [2]. Its overlap with the solute electron density defines the solvent–solute boundary, allowing the cavity to adapt self-consistently to changes in the electronic structure, state, and molecular geometry [3,4]. This density-based boundary is used to evaluate nonlocal electrostatics, continuum-integrated DFT-D5 dispersion, explicit cavitation, and translational/rotational entropy losses upon transfer to solution.

For practical applications, GEMS is fully differentiable [4,5], allowing efficient geometry optimizations and Hessian computations. The model's computational cost is typically less than twice that of the corresponding gas-phase calculation, and extension to new solvents requires only the solvent geometry, mass density, and dielectric permittivity. We evaluate GEMS on a compiled set with more than 12,000 experimental data points, including diverse solvents and properties such as solvation free energies and acid dissociation constants. On these benchmarks, GEMS reaches the accuracy of SMD and COSMO-RS for simpler neutral organic molecules, while showing particularly promising results for more challenging cases such as larger molecules and zwitterions [1].

The model will be available in ORCA and in the new Modular and Open-source Implicit Solvation Toolbox (MOIST) [6], providing an accessible route toward robust quantum-chemical modeling in solution.

References

[1] L. Wittmann, C. S. Selzer, S. Grimme, Chem. Sci. 2025, 16, 22976.
[2] R. Sundararaman, W. A. Goddard, J. Chem. Phys. 2015, 142, 064107.
[3] L. Wittmann, S. Grimme, Manuscript in preparation.
[4] L. Wittmann, A. Pausch, ChemRxiv 2026, 10.26434/chemrxiv.15003893/v2.
[5] L. Wittmann, M. Garcia-Ratés, C. Riplinger, J. Comput. Chem. 2025, 46, e70099.
[6] L. Wittmann, MOIST, GitHub repository, 2026, https://github.com/lukaswittmann/moist

P190

Simulating a bimetallic rotaxane with classical molecular dynamics

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Structural confinement is ubiquitous in nature and can lead to strong alteration of the physicochemical properties of the confined molecules. Previously, we have reported a photocatalytically active rotaxane, which is the product of confining a bimetallic, Ru-Rh, dyad inside the cavity of a cucurbit[7]uril ring. [1] This design principle leads to steric protection that overcomes degradation during catalytic conditions.
Our initial studies involved density functional theory (DFT) calculations in combination with continuum solvation models, to account for solvent effects. Our approach was also limited to the study of a few conformations. The limitations concerning solvation and geometry sampling can in principle be overcome by classical molecular dynamics (MD) simulations, where solvent molecules are treated explicitly and a vast number of geometries are sampled. However, MD simulations on our system were not possible due to the absence of force-field (FF) parameters for the description of the Rh-diimine moiety, which is part of the dyad.
Herein, we present such tailored FF parameters [2] and perform MD simulations by combining them with already existing ones for the other parts of the molecule. The FF is validated against DFT results, and the effects of confinement are studied, by comparing the behavior of the rotaxane against the 'naked' dyad. Moreover, we study the effect the different solvents exert on the dynamics of the dyad and the rotaxane, by performing the MD simulations in water, acetonitrile, and diethylene glycol. Key dihedral angles are identified, which enable a structural classification of the system. Our classification leads to a quick determination of a conformer's ability to allow for charge-transfer between the metal centers. We also determine lifetimes and corresponding energy barriers associated with important structural motions.

References
[1] M. Müßler, M. Semwal, K. P. Zois, L. Blechschmidt, B. M. W. Roberts, A. K. Mengele, L. Zedler, P. A. Hoffmann, L. D. Kapp, M. von Delius, L. González, B. Dietzek-Ivanšić and S. Rau, ChemRxiv, 2025
[2] R. Jacobi, K. P. Zois, A. K. Mengele, S. Rau and L. González, The Journal of Physical Chemistry B, 2025, 130, 472–485.

P191

Small molecular ions as probes for variations of fundamental constants

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Molecules provide a well-established framework for investigating the laws of nature at low-energies [1]. One such test is a hypothetical variation of fundamental physical constants, such as the fine-structure constant or the electron-to-proton mass ratio [2]. Many theories that attempt to unify gravity with other known forces assume such variations. Accompanying energy-shifts can, in principle, be detected in laboratory experiments when clock transitions in atoms and molecules tend to drift [3]. Thus, progress in research on variations of fundamental constants also represents, indirectly, advances in time measurement.

One way of testing such variations is to measure two transitions starting from a common initial state. The influence of a potential variation in the fine-structure constant is then obtained from the difference between the sensitivity factors of these transitions, divided by the difference in energy. Transitions to quasi-degenerate vibronic states are therefore particularly well suited. The sensitivity factors are usually calculated using electronic structure theory by numerically varying the speed of light in relativistic electronic structure programs.

In this contribution, we discuss an analytical approach [4] for calculating the sensitivity factors. The formulas are derived both within the four-component Dirac–Coulomb framework and the two-component Zeroth Order Regular Approximation [5-7]. On the level of complex generalized Hartree–Fock, we investigate several small molecular ions for possible large energy-shifts due to variations in the fine-structure constant and the electron-to-proton mass ratio. Small molecules with a large charge have the appeal of a dense level structure [8], which gives rise to many quasi-degenerate levels and enhances thereby relative shifts of transitions energies.

References

[1] G. Arrowsmith-Kron, et al., Rep. Prog. Phys. (2024), 87, 084301.
[2] J.-P Uzan, Rev. Mod. Phys. (2003), 75, 403.
[3] M. S. Safronova, Ann. Phys. (2019), 531, 1800364.
[4] C. Zülch, K. Gaul, R. Berger, arXiv (2025), 2511.10791, physics.chem-ph.
[5] C. Chang, M. Pelissier, P. Durand, Phys. Scr. (1986), 34, 384.
[6] E. van Lenthe, E. J. Baerends, J. G. Snijders, J. Chem. Phys. (1993), 99, 4597.
[7] C. Zülch, K. Gaul, S. M. Giesen, R. F. Garcia Ruiz, R. Berger, arXiv (2022), 2203.10333, physics.chem-ph.

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Grid-Based Description of Molecular Association

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We develop a sampling-free, grid-based approach as an alternative to Markov State Models (MSMs) to model molecular association processes. We use rigid body approximation to simplify association of two molecules down to a six-dimensional space of relative translation and orientation, where a grid-based discretization of space is computationally manageable and physically meaningful. Considering the total exchange of probability density between neighboring grid cells as the approximation of rate in the short-time limit, we provide expressions for rates that do not rely on sampling frequency; instead, a single energy evaluation per grid cell is required as well as geometric properties of the grid: cell surface area, distance between gridpoints and cell volumes. The resulting rate matrix is closely related to the transition matrix, offering insights into metastable states and association kinetics. The reduced number of energy evaluations compared to sampling approaches makes the method accessible not only to MD, but also DFT-based methods. We demonstrate our methodology on several systems of interests, including hydrogen bonding of water molecules, pi-pi stacking of guanidinium groups and diffusion of host-molecules through metal-organic frameworks. The software is made available as a python package MolGri.

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Accelerating Conformer Reranking with Machine Learning Interatomic Potentials

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The foundation for most computational chemistry workflows is thorough conformational sampling. As the number of low-energy conformers grows rapidly with molecular flexibility, tools like CREST [1] and GOAT [2] are usually restricted to inexpensive molecular mechanics (GFN-FF [3]) or semi-empirical (GFN2-xTB [4]) methods. To make up for their rather inaccurate conformational energies, large energy windows need to be chosen to avoid discarding relevant structures. This leads to ensembles of hundreds of conformers, which makes reranking at DFT level the computational bottleneck.
In this context, we investigate whether machine learning interatomic potentials including MACE-POLAR-1 [5] and UMA [6] provide more accurate conformers, including structures that might be absent in GFN-FF- or GFN2-xTB-based ensembles. Furthermore, we evaluate to what extent MLIPs as well as the next-generation SQM method g-xTB [7] may accelerate the reranking workflow. Here, we tested the respective methods in the CENSO [8] multi-level framework.
For conformer search, using uma-s-1p2 with GOAT shows only marginal improvement in the final DFT ensembles compared to GFN2-xTB-based sampling, indicating limited practical benefit at a significantly higher cost. In contrast, for the subsequent reranking, optimizing conformer ensembles at MLIP level allows for much smaller energy windows, decreasing the number of candidate structures, and hence accelerating the reanking procedure significantly. In our benchmarks, MACE-POLAR-1 offers the best balance of accuracy and cost.

References
[1] P. Pracht, S. Grimme, C. Bannwarth, et al., J. Chem. Phys. 2024, 160, 114110.
[2] B. de Souza, Angew. Chem. Int. Ed. 2025, 64, e202500558.
[3] S. Spicher, S. Grimme, Angew. Chem. Int. Ed. 2020, 59, 15665–15673.
[4] C. Bannwarth, S. Ehlert, S. Grimme, J. Chem. Theory Comput. 2019, 15, 1652–1671.
[5] I. Batatia, W. J. Baldwin, D. Kuryla, et al., 2026, arXiv preprint,
DOI: 10.48550/ARXIV.2602.19411.
[6] B. M. Wood, M. Dzamba, X. Fu, et al., 2025, arXiv preprint,
DOI: 10.48550/ARXIV.2506.23971.
[7] T. Froitzheim, M. Müller, A. Hansen, et al., 2025, DOI: 10.26434/chemrxiv-2025-bjxvt.
[8] S. Grimme, F. Bohle, A. Hansen, et al., J. Phys. Chem. A 2021, 125, 4039–4054.

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Computational discovery of the initial formation processes of the solid-electrolyte interface and the role of novel additives

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Solid electrolyte interphase (SEI) formation in lithium-ion batteries is governed by the earliest reductive decomposition events of electrolyte components. Yet these events occur on different timescales, involve multiple competing and often multi-electron pathways, and are difficult to probe experimentally. Conventional mechanistic studies compound this problem: a pathway is typically proposed by chemical intuition and then characterized in isolation by density functional theory (DFT), an approach that can only ever describe the pathway it set out to test and therefore cannot detect competing, unanticipated chemistry. Our approach combines ab initio nanoreactor molecular dynamics with electron addition. Periodic mechanical compression drives the system into densely interacting configurations that accelerate reactive events, but SEI formation is inherently a reductive process, and mechanical driving forces alone cannot simulate the electron-transfer steps required to initiate it. Sequential electron injection therefore supplies the missing reducing environment, approximating the potential experienced at the electrode surface and allowing reduction-initiated SEI chemistry to be simulated alongside the compression-driven reactions. This approach grants unbiased access to both barrierless and activation-limited decomposition pathways beyond the reach of conventional AIMD. In ethylene carbonate (EC) and EC-based mixtures with vinylene carbonate (VC) or 1,3-propane sultone (PS), ab initio nanoreactor MD in combination with electron addition maps in a dense network of reductive decomposition pathways, revealing that EC, VC, and PS share a common kinetic bottleneck in EC ring-opening, while VC and PS become reducible at less negative potentials, allowing them to intercept electron flux and redirect decomposition before EC-dominated pathways can proceed. Building on this validated framework, we then turn the electron-addition nanoreactor on an actively debated additive, 1,3,6-hexanetricarbonitrile (HTCN), whose decomposition mechanism, reductive cyanide CN- release, has so far been examined only along this single proposed pathway. The resulting network is resolved into distinct mechanistic classes, bimolecular EC-addition, CN- release, and oligomerization, each kinetically and thermodynamically characterized.

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Automated Assembly and Characterization of Coordination Cages

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Coordination cages are self-assembling metal-ligand structures that feature nanoscale cavities utilized for sensing, encapsulation, and functional materials.[1] Expanding upon our previous frameworks for the automated rational design and assembly modelling of metal-organic polyhedra (MOPs),[2,3] we present the "Cageinator", a computational Python pipeline designed for the fast generation and preliminary screening of coordination cages. The "Cageinator" builds geometries directly from linker coordinates by positioning metal nodes across predefined topologies (such as octahedral, cuboctahedral, or lantern) and optimizing the orientation of the linkers so that donor atoms (N/O/S) align with these nodes. To enable quick screening of structures and properties, candidate architectures are initially optimized using force fields and subsequently refined via GFN2-xTB.[4] For validation purposes, we reconstruct a previously reported photoswitchable cage, successfully replicating the characteristic UV-Vis signatures of both its open- and closed-ring isomers.[5] Current efforts are focused on expanding this methodology to encompass a wider variety of cage families, alongside the implementation of automated property ranking.[6]

References

[1] A. Tarzia, K. E. Jelfs. Chem. Commun. 2022, 58, 3717–3730. DOI: 10.1039/D2CC00532H.
[2] A. Kondinski et al. J. Am. Chem. Soc. 2022, 144, 11713–11728. DOI: 10.1021/jacs.2c03402.
[3] A. Kondinski et al. Eur. J. Inorg. Chem. 2025, 28(26), e202500115. DOI: 10.1002/ejic.202500115.
[4] C. Bannwarth, S. Ehlert, S. Grimme. J. Chem. Theory Comput. 2019, 15(3), 1652–1671. DOI: 10.1021/acs.jctc.8b01176. [5] S. J. Wezenberg. Chem. Lett. 2020, 49(6), 609–615. DOI: 10.1246/cl.200076. [6] J. Leodolter, A. Kondinski. Manuscript in preparation.

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Thermodynamic Properties of Various Polymorphs of 2-Thiobarbituric Acid via Quasi-Harmonic Calculations

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Polymorphism is a well-known phenonomen in molecular crystal systems and important for the development of active pharmaceutical ingredients (APIs). A particular variant, tautomeric polymorphism, occurs when the crystalline phases of a system contain different tautomeric forms. Although many APIs exhibit tautomerism, tautomeric polymorphs are rarely observed [1].
A prominent example of a system with a rich collection of tautomeric polymorphs is 2-thiobarbituric acid (TBA) as it is reported to exist in up to six different crystalline forms: one containing only the enol tautomer (II), four composed of the keto form (I, III, V and VI), and one containing a 50:50 mixture of both tautomers (IV) [2,3].
Here, we explore the thermodynamic properties of six polymorphs (II, III, IV, V, VI and VI_{trans}) of TBA by using a subtractive multimer embedding scheme — ME3(PBE0+MBD:PBE+MBD) — which approximates a full periodic density functional theory (DFT) calculation at the PBE0+MBD level by embedding multimers up to trimers into DFT calculations at the PBE+MDB level [4]. This approach was used for lattice optimizations and phonon calculations using the light species default settings in FHI-aims, with additional electronic lattice energies obtained using tight settings. All calculations were performed utilizing the MEmbed python package. To account for thermal effects, the quasi-harmonic approximation was used in conjunction with a fit to the Murnaghan equation of state.
Overall, polymorphs of the same tautomeric composition show similar values for the relative Helmholtz (ΔF) and Gibbs (ΔG) free energy per molecule both at 0 K and 300 K, allowing for a clear separation of all investigated polymorphs based on thermodynamic stability: form II is the thermodynamically most stable polymorph, followed by form IV, with the least stable group comprising the forms III, V, VI and VI_{trans}. Values for the heat capacity at constant pressure C_p at 300 K lie in the range of 128.1 to 132.0 J/mol·K per molecule for all polymorphs except form V, which exhibits a value of 123.2 J/mol·K per molecule. Thermal expansion coefficients β range from 0.5×10^{-5} K^{-1} per molecule (form IV) to 4.8×10^{-5} K^{-1} per molecule (form III).

References

[1] A. J. Cruz-Cabeza, C. R. Groom, CrystEngComm 13, 93–98 (2011).
[2] M. R. Chierotti, L. Ferrero, N. Garino, R. Gobetto, L. Pellegrino, D. Braga, F. Grepioni, L. Maini, Chem. Eur. J. 16, 4347–4358 (2010).
[3] A. Woods-Ryan, C. L. Doherty, A. J. Cruz-Cabeza, CrystEngComm 25, 2845–2858 (2023).
[4] J. Hoja, A. List, A. D. Boese, J. Chem. Theory Comput. 20, 357–367 (2024).

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Detailed Pathways of Chemical Reactions in Molecular Crystals Using MLFFs

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Machine-learned force fields (MLFFs), which provide density functional theory (DFT)-level accuracy for nudged elastic band (NEB) and frequency calculations, enable the simulation of supercells that are otherwise computationally prohibitive [1]. Reaction pathways in molecular crystals frequently assume that all constituents within a unit cell react simultaneously, neglecting how sequential reaction orders impact energy barriers and internal mechanical stress. Building upon the Diels-Alder reaction in a bis(N-allylimino)-1,4-dithiin and 9-bromoanthracene cocrystal [2], we decouple these individual reactions and investigate a detailed sequential pathway. Although the minimum energy path (MEP) profiles yield comparable energies for both simultaneous and sequential mechanisms, the sequential route is physically more probable. Expanding this analysis from a single unit cell to 2x2x2 and 3x3x3 supercells further demonstrates that surrounding unreacted lattice environments significantly influence the overall MEP energy barrier.

References

[1] N. Goncharova, O. Schweighofer, J. Hoja, in preparation.
[2] N. Goncharova, J. Hoja, J. Chem. Phys. 163, 164127 (2025).