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P51 **Szabolcs Goger** — Universität des Saarlandes, Germany
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P57 **Eric Heller** — University of Freiburg, Germany
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P1

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

T. Abdurakhmonov (University of Rostock, DE), C. Rehhagen (University of Rostock, DE), M. Frank (University of Rostock, DE), O. Kühn (University of Rostock, DE), S. Lochbrunner (University of Rostock, DE)

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

Z. Aigner (University of Innsbruck/AT), M. Hütter (University of Innsbruck/AT), M. Ončák (University of Innsbruck/AT)

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.

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P3

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

Ardhmeri Alija (Helmholtz Institute Mainz, Johannes Gutenberg-Universität Mainz, DE), Konstantin Gaul (Helmholtz Institute Mainz, Johannes Gutenberg-Universität Mainz, DE)

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.

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P4

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

V. Alizadeh (CASUS, Germany), T. D. Kühne (CASUS, Germany)

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)

V.K. Ambasta (FAU, Erlangen/GER), S. Maier (RWTH, Aachen/GER), C. Müller (FAU, Erlangen/GER)

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].

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P6

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

A. Mrinal Arandhara (Ruhr University Bochum/DE) B. Laura Duran Caballero (Ruhr University Bochum/DE) C. Harald Forbert (Ruhr University Bochum/DE) D. Dominik Marx (Ruhr University Bochum/DE)

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

A. Balbi (University of Warsaw/PL), D. Zgid (University of Warsaw/PL)

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).
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P9

Optimizing Input vs Correcting Output in Semiempirical Machine Learning

P. Baltruschat (University of Hamburg), C. Herrmann (University of Hamburg)

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.

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P11

Exploiting Hard-Core Boson Structure in Quantum Chemistry

L. Barta (University of Augsburg), J. S. Kottmann (University of Augsburg)

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.

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P12

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

Marco Bauer (KTH/SE); Patrick Norman (KTH/SE); Andreas Dreuw (Heidelberg University/DE); Anthony Dutoi (University of the Pacific/US)

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

Saman Behjou, Iulia Emilia Brumboiu, Katharina Boguslawski, (Nicolaus Copernicus University in Toruń, Poland)

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.

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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.

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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.

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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.

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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

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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.

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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)

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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.

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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.

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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.

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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.

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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

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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.

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P29

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

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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

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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

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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

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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

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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.

P36

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.

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P37

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.

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P38

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.

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P39

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.

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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.

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P41

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.

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P42

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.

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P43

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.

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P44

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

R. Friedrich (TU Dresden)

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.

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P45

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.

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P46

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.

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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].

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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.

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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]

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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.

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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.

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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.

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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.

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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.

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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]

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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.

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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.

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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.

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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.

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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.

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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.

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Multimer Embedding for Predictions of Polymorph Stability

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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).

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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.

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