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

Andreas Grüneis[1]

[1] *TU Wien, Austria*

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

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