O 17: Focus Session: Frontiers of Electronic-Structure Theory II (joint session O/HL)

Time: Monday 15:00–17:15

Location: TRE Ma

Topical Talk   O 17.1   Mon 15:00   TRE Ma
Coupled-cluster theory for complex solids made ready —
•Andreas Grüneis — TU Wien, Institute for Theoretical Physics,
Wiedner Hauptstraße 8-10/136, 1040 Vienna, Austria

This talk will review recent progress in applying periodic coupled-cluster theory, which has the potential to achieve a systematically improvable level of accuracy, to solids and surfaces. We will discuss novel techniques that reduce the computational cost by accelerating the convergence of calculated properties towards the complete basis set as well as the thermodynamic limit. The newly developed techniques have been implemented in the free and open source simulation software Cc4s that is interfaced to a growing number of widely-used electronic structure theory codes. These developments have enabled an increasing number of ab initio studies and allowed for assessing the level of accuracy of coupled-cluster theory by comparing to experimental findings as well as quantum Monte Carlo results. The presented applications will cover a wide range of materials science problems including the study of phase diagrams, molecule-surface interactions and properties of defects.

O 17.2   Mon 15:30   TRE Ma
**Speedup of structural optimisations using hybrid functionals:
Case studies for energy materials** — •Daniel Fritsch — Weierstrass Institute for Applied Analysis and Stochastics, Mohrenstr. 39,
10117 Berlin, Germany — Helmholtz-Zentrum Berlin für Materialien
und Energie, Hahn-Meitner-Platz 1, Berlin, 14109, Germany

First-principles calculations based on density functional theory have been established as de facto standard for computational materials investigations. Depending on the size of the unit cell, for every material of interest a suitable choice for the unknown exchange and correlation functional has to be made; taking not only into account the desired accuracy, but also the available computational resources.

In recent years, a promising combination of two approaches emerged, starting from a structural relaxation based on a simpler (semi)local functional, supplemented by a single shot hybrid functional calculation. Here, we propose a new method for combining different levels of exchange and correlation functional for structural relaxations.

In a first benchmarking step, this new method will be applied to various sets of promising energy materials, where full hybrid functional calculations are available, with a main focus on the performance of this new approach on the structural properties and the required computational resources. In a second step, this new approach will be applied to materials, which up-to-now have not been accessible to hybrid functional calculations due to the required computational resources. All the presented results on the structural, electronic, and optical properties will be critically discussed alongside experimental findings.

O 17.3   Mon 15:45   TRE Ma
All-Electron Large-Scale Hybrid Density Functional Simulations — Florian Merz[1], Andreas Marek[2], •Sebastian Kokott[3],
Christian Carbogno[3], Yi Yao[3], Mariana Rossi[4], Markus
Rampp[2], Matthias Scheffler[3], and Volker Blum[5] — [1]Lenovo
HPC Innovation Center, Stuttgart — [2]Max Planck Computing and
Data Facility, Garching — [3]The NOMAD Laboratory at the FHI-
MPG and IRIS-Adlershof HU, Berlin — [4]MPI for the Structure and
Dynamics of Matter, Hamburg — [5]Duke University, North Carolina,
USA

The localized resolution-of-identity approach [1] enables $O(N)$ hybrid density functional simulations and, thus, the computation of accurate electronic properties of large scale atomistic models in the range of ten thousands of atoms in *FHI-aims* [2]. In this range, parallelization and memory requirements of the exact exchange part, and the evaluation of the Hartree potential remain challenging. The solution of the generalized eigenvalue problem with direct eigensolvers like ELPA [3] naturally becomes a bottleneck due to $O(N^3)$ scaling. In this work, we present recent algorithmic advances for the exact exchange part and the evaluation of the Hartree potential, as well as optimizations of the ELPA library. We systematically perform benchmark tests on CPU and GPU-accelerated architectures covering inorganic solids, large molecules, and organic crystals with up to 50,000 atoms.
[1] Ihrig *et al.*, *New J. Phys.* **17**, (2015).
[2] Levchenko *et al.*, *Comp. Phys. Commun.* **192**, (2015).

[3] Marek *et al.*, *J. Phys. Condens. Matter* **26**, (2014).

15 min. break

O 17.4   Mon 16:15   TRE Ma
**A Koopman's compliant exchange correlation potential for
semiconductors** — •Michael Lorke[1], Peter Deak[2], and Thomas
Frauenheim[2] — [1]Institute for Theoretical Physics, University of Bremen, Otto-Hahn-Allee 1, Bremen, 28359, Germany — [2]BCCMS, University of Bremen, Germany

Density functional theory is the workhorse of theoretical materials investigations. Due to the shortcoming of (semi-)local exchange correlation potentials, hybrid functionals have been established for practical calculations to describe surfaces, molecular adsorption, and defects. These functionals operate by mixing between semi-local and Hartree-Fock exchange semi-emprically. However, their parameters have to be optimized for every material separately. To treat materials with a more physics driven approach and without the need of parameter optimization is possible with many-body approaches like GW, but at an immense increase in computational costs and without the access to total energies and hence geometry optimization. We propose a novel exchange correlation potential[1] for semiconductor materials, that is based on physical properties of the underlying microscopic screening. We demonstrate that it reproduces the low temperature band gap of several materials. Moreover it respects the required linearity condition of the total energy with the fractional occupation number, as expressed by the generalized Koopman*s theorem. We also show that this novel functional can be used as a kernel in linear response TDDFT to reproduce excitonic effects in optical spectra [1] Physical Review B 102 (23), 235168 (2020)

O 17.5   Mon 16:30   TRE Ma
**Accurate and efficient treatment of spin-orbit coupling via
second variation employing local orbital basis functions** —
•Hannah Kleine[1], Andris Gulans[2], Sven Lubeck[1], Cecilia
Vona[1], and Claudia Draxl[1] — [1]Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin — [2]Department of Physics, University of Latvia

Spin-orbit-coupling (SOC) effects can significantly change the properties of materials containing heavy elements, mostly by introducing shifts and splittings in the band structure. Including SOC effects in density-functional-theory (DFT) calculations can be challenging. In the linearized augmented planewaves plus local orbitals (LAPW+LO) method, SOC is treated as a perturbation and solved by a second variational (SV) scheme where eigenvectors of the scalar-relativistic Kohn-Sham Hamiltonian are used as basis functions for the SO-coupled problem. For certain materials, especially those with strong SOC effects, many SV basis functions are required which leads to high computational costs. By adding LOs to the SV basis, we are able to drastically reduce the basis-set size and thus the computational cost. We implement this approach in the all-electron full-potential computer package exciting [1] and combine it with the use of relativistic LOs to achieve high accuracy results for a variety of different materials.
[1] A. Gulans et al., J. Phys. Condens. Matter 26, 363202 (2014).

O 17.6   Mon 16:45   TRE Ma
**Exact sum-rule approach to polarizability and asymptotic van
der Waals functionals – derivation of exact benchmarks** —
•Alberto Ambrosetti[1], John Dobson[2], Matteo Ricci[1], and Pier
Luigi Silvestrelli[1] — [1]Dipartimento di Fisica e Astronomia, Università degli Studi di Padova, via Marzolo 8, 35131, Padova, Italy
— [2]Nanoscale Science and Technology Centre, Griffith University,
Nathan, Queensland 4111, Australia

Using a sum-rule approach we develop an exact theoretical framework for polarizability and asymptotic van der Waals correlation energy functionals of isolated objects[1]. The functionals require only monomer groundstate properties as input. Functional evaluation proceeds via solution of a single position-space differential equation, without the usual summations over excited states or frequency integrations. Explicit functional forms are reported for reference physical systems, including atomic hydrogen and single electrons subject to harmonic confinement, and immersed in a spherical-well potential. Direct com-

parison with the popular Vydrov-van Voorhis density functional shows that this performs best when density decay occurs at atomic scales. The adopted sum-rule approach implies general validity of our theory, enabling exact benchmarking of van der Waals density functionals, and direct inspection of the subtle long-range correlation effects that constitute a major challenge for approximate (semi-)local density functionals.

[1] M Ricci, PL Silvestrelli, JF Dobson, A Ambrosetti J. Phys. Chem. Lett. 13, 8298-8304

O 17.7   Mon 17:00   TRE Ma

Interacting electrons and bosons in the doubly screened $G\widetilde{W}$ approximation — ●Yaroslav Pavlyukh — Department of Theoretical Physics, Wroclaw University of Science and Technology

In Ref. [1] we built on the Generalized Kadanoff-Baym Ansatz for electrons and bosons to map a broad class of nonequilibrium Green's function theories onto a coupled system of ordinary differential equations with linear time-scaling. Available methods to treat e-e correlations include GW [2], T-matrix and Faddeev, while e-b correlations are described by Ehrenfest and second-order diagrams in the e-b coupling [3].

In this work we present a substantial advance in the treatment of correlations, requiring no extra computational cost and preserving all conserving properties. Specifically, we include the effects of dynamical screening due to *both e-e* and *e-b* interactions ($G\widetilde{W}$ approximation). The $G\widetilde{W}$ extension opens the door to a wealth of phenomena ranging from carrier relaxation and exciton recombination to molecular charge migration and transfer in optical or plasmonic cavities.

[1] Y. Pavlyukh, E. Perfetto, D. Karlsson, R. van Leeuwen, and G. Stefanucci, Phys. Rev. B **105**, 125134 (2022).

[2] E. Perfetto, Y. Pavlyukh, and G. Stefanucci, Phys. Rev. Lett. **128**, 016801 (2022).

[3] D. Karlsson, R. van Leeuwen, Y. Pavlyukh, E. Perfetto, and G. Stefanucci, Phys. Rev. Lett. **127**, 036402 (2021).