MM 12: Computational Materials Modelling: Physics of Ensembles 1

Time: Tuesday 10:15–13:00   Location: H44

MM 12.1 Tue 10:15 H44

Pinpointing Hubbard corrections to tackle inhomogenous nl subshells: The DFT+U(m) method — •Eric Macke[1], Iurii Timrov[2], Lucio Colombi Ciacchi[1], and Nicola Marzari[2] — [1]Hybrid Materials Interfaces Group and Bremen Center for Computational Materials Science, MAPEX, University of Bremen, Am Fallturm 1, 28359 Bremen, Germany — [2]Theory and Simulation of Materials (THEOS) and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), Ecole Polytechnique Federale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

DFT+U remains a key tool in computational material science as it mitigates the DFT self-interaction error. While this simplistic approach often provides electronic and magnetic properties in fair agreement with literature, recent investigations revealed that DFT+U massively over-stabilizes high spin configurations of transition metal elements surrounded by strong ligand fields. In such compounds, the projection of occupied Kohn-Sham states onto the atomic basis frequently yields occupation numbers $n_m^\sigma \approx 0.5$ for certain magnetic quantum numbers m, so that the penalizing Hubbard term $E_U = \sum_{m,\sigma} \frac{U}{2}[n_m^\sigma(1 - n_m^\sigma)]$ is maximized. We argue that if the occupation of a magnetic (spin-) orbital is dominated by the effect of hybridization with a ligand, Hubbard corrections should not be applied in the same way as for unaffected orbitals. To account for such inhomogeneous bonding regimes, we propose a more flexible scheme that enables the use of distinct Hubbard parameters $U(m)$ for the same species, computable by means of *ab initio* methods.

MM 12.2 Tue 10:30 H44

Efficient computation of optical properties of large-scale heterogeneous systems — •Joseph C. A. Prentice[1] and Arash A. Mostofi[2] — [1]Department of Materials, University of Oxford, UK — [2]Departments of Physics and Materials, Imperial College London, UK

The optical properties of large-scale (>1000 atoms) heterogeneous systems are of interest in several fields, from photovoltaics to biological systems. Computing such properties accurately from first principles, however, is challenging; even if only a small region is optically active, quantum mechanical environmental effects must often be included, and the cost of applying a quantitatively accurate level of theory is prohibitive. Here, I present recent work demonstrating how such calculations can be performed efficiently from first principles via two methods: an extension of the spectral warping method of Ge et al., and a novel combination of quantum embedding (specifically embedded mean-field theory) and linear-scaling (time-dependent) density functional theory. The accuracy and utility of these methods is demonstrated by applying them to systems including the molecular crystal ROY, chromophores in solution, and pentacene-doped p-terphenyl. The results pave the way for quantitatively accurate calculations to be performed on previously inaccessible large-scale systems.

MM 12.3 Tue 10:45 H44

Converging tetrahedron method calculations for non-dissipative parts of spectral functions — •Minsu Ghim[1,2,3] and Cheol-Hwan Park[1,2,3] — [1]Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, Korea — [2]Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea — [3]Center for Theoretical Physics, Seoul National University, Seoul 08826, Korea

Integrations in k space are used to calculate many of the physical quantities in solid-state physics. Examples include various static or dynamical conductivity, self-energy of an electron, and electric polarizability. The integral usually takes the form of a product of proper matrix elements and $1/[\hbar\omega - (\epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}}) + i\eta]$, which is decomposed into the real part and the imaginary part, $P\{1/[\hbar\omega - (\epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}})]\}$ and $-i\pi\delta[\hbar\omega - (\epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}})]$, respectively. Here, ω is the frequency, $\epsilon_{m\mathbf{k}}$ and $\epsilon_{n\mathbf{k}}$ are the energies of the valence and conduction electronic bands with Bloch wavevector $\mathbf{k}$, respectively, and $\eta = 0^+$. Although the delta-function part has been widely calculated by the tetrahedron method, the non-dissipative principal value integral part has not. Tools to obtain matrix elements and energy eigenvalues from first principles have been actively developed, but there are technical difficulties in the tetrahedron method for the non-dissipative part. In this talk, we introduce an easy-to-implement, stable method to overcome those obstacles. Furthermore, our method is tested by calculating the spin Hall conductivity of fcc platinum.

MM 12.4 Tue 11:00 H44

Self-consistency in GWΓ formalism leading to quasiparticle-quasiparticle couplings — •Carlos Mejuto-Zaera[1] and Vojtěch Vlček[2] — [1]Scuola Internazionale Superiore di Studi Avanzati SISSA, Trieste, Italy — [2]University of California Santa Barbara UCSB, USA

Many-body perturbation theory (MBPT) has become a tool of choice for the description of materials. Traditionally tied to the weakly-interacting limit, e.g. through the GW approximation, its expansion towards higher interactions is a fundamental goal in the field. Existing attempts can rely on selected diagram resummation or downfolding, which involve some choice of expansion parameters typically reducing the approach's generality and flexibility. Despite this apparently intrinsic price, MBPT provides through Hedin's equations a formally exact path to describe any dynamically correlated system, regardless of interaction strength. We revisit this framework to disentangle how to systematically extend its practical validity beyond weak interactions, focusing on the structure of the interaction vertex Γ within self-consistent implementations. Using the Hubbard dimer at half-filling, we unveil the role of Γ's functional derivative with respect to the Green's function, hitherto typically neglected, in generating diagrams that can extend the reliability of MBPT towards high interactions [1].

[1] arXiv:2203.05029

MM 12.5 Tue 11:15 H44

Automated Corrections for Materials Design of Ionic Systems: AFLOW-CCE — •Rico Friedrich[1,2], Marco Esters[1], Corey Oses[1], Stuart Ki[1], Maxwell J. Brenner[1], David Hicks[1], Michael J. Mehl[1], Cormac Toher[1], and Stefano Curtarolo[1,3] — [1]Center for Autonomous Materials Design, Duke University, USA — [2]Helmholtz-Zentrum Dresden-Rossendorf, Dresden — [3]Materials Science, Electrical Engineering, and Physics, Duke University, USA

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] — our implementation of CCE into the AFLOW framework. It provides a tool where users can input a structure file and receive the CCE corrections, or even the CCE formation enthalpies if pre-calculated LDA, PBE or SCAN values are provided. The implementation features a command line tool, a web interface, and a Python environment.

[1] S. Curtarolo *et al.*, Comput. Mater. Sci. **58**, 218 (2012).
[2] V. Stevanović *et al.*, Phys. Rev. B **85**, 115104 (2012).
[3] R. Friedrich *et al.*, npj Comput. Mater. **5**, 59 (2019).
[4] R. Friedrich *et al.*, Phys. Rev. Mater. **5**, 043803 (2021).

15 min. break

MM 12.6 Tue 11:45 H44

Revealing the ambient and high-pressure phases of group IV monochalcogenides and monoxides with an evolutionary algorithm — •Long Nguyen and Guy Makov — Department of Materials Engineering, Ben-Gurion University of the Negev, Beer-Sheva, Israel

Group IV chalcogenides and oxides are candidates for incorporation into optoelectronic and thermoelectric conversion devices. These functional materials are mostly p-type semiconductors that provide low-cost, environmentally friendly, abundant, and high-efficiency. To expand the possible phases and study their stability upon compression, a combination of DFT modelling with evolutionary algorithm (EA) is applied to extensively investigate across the group IV monochalcogenides and oxides. We will report our results from the implementation of the EA to search for stable phases. In group IV monoxides, our results include multiple structural transitions of SnO and PbO upon

compression. Two new high-pressure polymorphs are predicted, the orthorhombic Pbcm SnO and the octahedral monoclinic C2/m PbO. Weakening of the lone pairs and elastic instability are the main drivers for the structural transitions. Among the monochalcogenides, SnS and GeS phase families are examined together with their electronic and optical properties. At ambient conditions, GeS and SnS can present several polymorphs with bandgaps in the range from 0.3 to 1.6 eV, which can be attractive for a variety of optoelectronics applications. Upon compression, we found a similar convergence of SnS and GeS into the Cmcm and later the Pm3m phase. The transition mechanism is discussed through the lone pairs' influence on the structural distortion.

MM 12.7 Tue 12:00 H44

An Open-source Interface to MP2 and Coupled-Cluster Methods for Solids for Localized Basis Set Codes — •Evgeny Moerman[1], Felix Hummel[2], Andreas Irmler[2], Andreas Grüneis[2], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the FHI-MPG, Berlin — [2]Inst. for Th. Phys., TU Wien, Vienna

Coupled cluster (CC) theory is often considered the gold standard of quantum-chemistry. For solids, however, the available software is scarce ([1] and references therein). We present CC-aims[2], which can interface ab initio codes with localized atomic orbitals and the CC for solids (CC4S) code by the group of A. Grüneis. CC4S features a continuously growing selection of wave function-based methods including perturbation and CC theory. The CC-aims interface was developed for the FHI-aims code[2] but is implemented such that other codes may use it as a starting point for corresponding interfaces. As CC4S offers treatment of both molecular and periodic systems, the CC-aims interface is a valuable tool, where DFT is either too inaccurate or too unreliable, in theoretical chemistry and materials science alike. In this talk we describe the CC-aims interface, the CC4S code, and demonstrate the application of CC and MP2 by investigating the relative stability of boron nitride phases. - This work received support from the European Union's Horizon 2020 research and innovation program under Grant Agreement No. 951786 (The NOMAD CoE).

[1] Q. Sun *et al.*, J. Chem. Phys. 153:024109 (2020)
[2] E. Moerman *et al.* to be published, J. Open Source Software
[3] The FHI-aims web page: https://fhi-aims.org

MM 12.8 Tue 12:15 H44

Wannier function perturbation theory: localized representation and interpolation of wavefunction perturbation — •Jae-Mo Lihm and Cheol-Hwan Park — Department of Physics, Seoul National University, Seoul, Korea

Wannier functions provide a localized representation of the electronic structure of solids and are thus finds wide application in condensed matter physics. However, the Wanneier function method is limited in that it cannot be used to represent the change of the wavefunctions due to perturbations. In this work, we introduce Wannier function perturbation theory (WFPT) [1], which overcomes this limitation by providing a localized representation of wavefunction perturbation which we term "Wannier function perturbation." Among the diverse possible applications of WFPT, we provide three examples. First, we calculate the temperature dependence of the indirect optical absorption spectra of silicon. Our calculation differentiates phonon-absorption and phononemission processes and includes the band-gap renormalization without arbitrary temperature-dependent shifts in energy at the same time,

which is beyond reach of existing methods. Second, we develop a theory for calculating the shift spin current without any band-truncation error and apply it to monolayer WTe_2. Third, we apply WFPT to calculate the spin Hall conductivity of the same material, again without any band-truncation error. WFPT will open up a new way for studying the response properties of the electron states in real materials.

[1] J.-M. Lihm and C.-H. Park, Phys. Rev. X, 11, 041053 (2021)

MM 12.9 Tue 12:30 H44

Boosting first-principles molecular dynamics with orbital-free density functional theory — •Lenz Fiedler[1], Zhandos A. Moldabekov[1], Xuecheng Shao[2], Kaili Jiang[2], Tobias Dornheim[1], Michele Pavanello[2], and Attila Cangi[1] — [1]Helmholtz-Zentrum Dresden-Rossendorf / CASUS — [2]Rutgers University Newark

Kohn-Sham density functional theory (KS-DFT) is one of the most important simulation methods in materials science and quantum chemistry. Yet, standard DFT codes exhibit scaling behaviors in terms of system size and temperature that prohibit extended dynamical investigations of materials using DFT driven molecular dynamics simulations (MD), especially towards the warm dense matter regime (WDM). We present a practical hybrid approach that combines orbital-free density functional theory (DFT) with Kohn-Sham DFT for speeding up first-principles molecular dynamics simulations. Equilibrated atomic configurations are generated using orbital-free DFT for subsequent Kohn-Sham DFT molecular dynamics. This leads to a massive reduction of the simulation time without any sacrifice in accuracy. We show results across systems of different sizes and temperature, up to the warm dense matter regime. To that end, we use the cosine distance between the time series of radial distribution functions representing the ionic configurations. Likewise, we show that the equilibrated ionic configurations from this hybrid approach significantly enhance the accuracy of machine-learning models that replace Kohn-Sham DFT.

MM 12.10 Tue 12:45 H44

First-principles calculations of plasma frequency in an all-electron full-potential framework — •Maria K. Pogodaeva, Sergey V. Levchenko, and Vladimir P. Drachev — Moscow, Russia

We present a first-principles methodology to calculate plasma frequency of using density-functional theory in an all-electron full-potential framework implemented in FHI-aims. The results are compared to the results obtained with pseudopotential approaches and experimental data from photo-emission, absorption, and electron energy loss spectra. We test the approach on seven elemental metals: gold, silver, sodium, copper, palladium, platinum, and aluminum. Our results reproduce experimental values for plasma frequency within 0.1-0.3 eV accuracy compared to experiment for all metals except sodium. Calculated plasma frequency for sodium is consistently overestimated by 0.5 eV by VASP and FHI-aims. For other metals except gold FHI-aims and VASP are in a perfect agreement with each other and with experiment. For gold VASP underestimates plasma frequency by 0.6 eV compared to experiment, but FHI-aims calculates the value within 0.2 eV. Overall, the agreement between the two codes and the experiment is good. This validates our all-electron implementation and allows for further applications of the methodology to more complex systems