O 50: Electronic structure theory II

Time: Tuesday 14:30–16:15 Location: TRE/PHYS

O 50.1 Tue 14:30 TRE/PHYS

Efficient DFPT for 2D Films in the FLAPW Code FLEUR — •Thomas Bornhake[1,2], Alexander Neukirchen[1], Gregor Michalicek[1], Daniel Wortmann[1], Gustav Bihlmayer[1], and Stefan Blügel[1,2] — [1]Peter Grünberg Institut, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany — [2]Institute for Theoretical Physics, RWTH Aachen University, 52074 Aachen, Germany

Interest in structural instabilities, magnetism, transport, and superconductivity in 2D films is rapidly increasing, and all of these phenomena involve phonons. However, *ab initio* phonon calculations for films typically require large supercells, which include substantial vacuum. As a result, they become computationally expensive. In this talk, we present an efficient and unique DFPT implementation in the context of the all-electron FLAPW code FLEUR [1,2] that treats films as single slabs in a truly 2D geometry perfectly embedded in semi-infinite vacuum [3]. We outline the key features of the method, benchmark it against the commonly employed supercell approach, and show first results on how a homogeneous external electric field modifies the phonon properties.

This work was supported by the CoE-MaX (EuroHPC Joint Undertaking, Grant 101093374) and DFG through CRC 1238 (Project C01).

[1] D. Wortmann *et al.*, 10.5281/zenodo.7576163; www.flapw.de
[2] C.-R. Gerhorst *et al.*, Electron. Struct. **6**, 017001 (2024).
[3] M. Posternak *et al.*, Phys. Rev. B **21** 5601 (1980).

O 50.2 Tue 14:45 TRE/PHYS

Valence only FLOSIC with pseudo core electrons — •Simon Liebing and Jens Kortus — Institute of Theoretical Physics, TU Bergakademie Freiberg, 09599 Freiberg, Germany

Over the past decade, the FLOSIC method [1, 2] has demonstrated its versatility and applicability to a wide range of systems. However, it has been observed that this method can require significant computational time, particularly when adjusting the core electrons in the case of heavier elements. Previous work has shown that FLOSIC calculations can be effectively carried out using pseudopotentials [3]. In the case of heavier atoms, however it is possible that the valence orbital descriptors may shift too close to the core region due to the limitations of pseudopotentials. In this work, the authors introduce a novel approach to also account for the repulsive interactions between the core and valence electron FLOSIC descriptors in case of pseudopotentials. This method has the potential to significantly reduce the computational cost typically associated with SIC calculations compared to standard DFT methods. References: [1] M. R. Pederson et al., J. Chem. Phys., 140, 121103 (2014) [2] S. Schwalbe, et al., J. Chem. Phys., 153, 8, 2020. [3] S. Liebing et al., DPG Verhandlungen Berlin, O, Session 82, Contribution 8, 2018.

O 50.3 Tue 15:00 TRE/PHYS

Spectroscopy of chemisorped systems from a first-principles local-orbital representation — •Simiam Ghan and Jens Nørskov — Technical University of Denmark DTU

We present a local-orbital representation of density functional theory (DFT) which elucidates the properties of chemisorped systems. A direct calculation of orbital overlap and coupling matrix elements between the adsorbate and surface allows us to construct the hybridization function, familiar from e.g. Anderson-Newns-Grimley models, which encodes rich orbital phase interactions[1]. Beyond a qualitative tool, we demonstrate the quantitative correspondence of this object to observables in ultrafast spectroscopy and scanning-tunneling spectroscopy. The scheme is generally useful to assess the coupling between a local state and its environment from first principles.

[1] S. Ghan *et al.*, J. Chem. Phys. 158, 234103 (2023).

O 50.4 Tue 15:15 TRE/PHYS

Quantum-mechanical analysis of the 4H-SiC surface — •Kateřina Dočkalová[1,2], Martin Friák[2], and Jana Pavlů[1] — [1]Dept. Chem., Masaryk Uni., Brno, Czech Rep. — [2]Inst. Phys. Mater., Czech Acad. Sci., Brno, Czech Rep.

Silicon carbide (SiC) is a key wide-bandgap semiconductor employed in high-power and high-voltage electronic devices. Among its polytypes, 4H-SiC is particularly promising for next-generation power electronics. We investigated the atomic and electronic structure of the 4H-SiC (0001) surface with a specific focus on the challenges of modelling polar orientations. To accurately capture the intrinsic surface properties, we evaluate the planar-averaged electrostatic potential and the electron localisation function (ELF) using various models (differing in bottom-surface termination, passivation with pseudohydrogens, and vacuum thickness) and applying electrostatic corrections. These strategies suppress artificial dipole moments and eliminate spurious internal electric fields that often compromise surface calculations. We find that either carbon termination at the bottom surface or pseudohydrogen passivation can individually introduce a spurious internal electric field manifested in the vacuum potential. Interestingly, applying electrostatic corrections does not improve the electrostatic-potential profile for models in which the vacuum region is already sufficiently thick. Financial support from the Czech Academy of Sciences (Praemium Academiae, M.F.) and Masaryk University (MUNI/A/1691/2024) is acknowledged. Computational resources were provided via e-INFRA CZ (ID:90254).

O 50.5 Tue 15:30 TRE/PHYS

Quantifying conductance variations in single molecule junctions using machine learning — •Hector Vazquez — Inst. of Physics, Czech Academy of Sciences

In single molecule circuits, conductance (the inverse of resistance) is strongly dependent on the junction geometry. Break-junction experiments are often carried out at room temperature, where many molecular conformations are sampled during the measurements. In contrast, the computational cost of DFT-NEGF calculations restricts them to only a few geometries.

We recently developed a computationally efficient method to calculate molecular conductance within DFT for thousands of geometries, based on small Au-molecule-Au clusters [1,2]. Their geometry is taken from MD simulations of the junction at room temperature. We can thus compute within DFT the conductance for tens of thousands of thermally-accessible molecular geometries.

We study typical conjugated and alkane molecules and interpret these large conductance datasets with machine learning methods including regression models, feature importance techniques, and SHAP analysis. Our work identifies which of the bond lengths, angles, or dihedral angles in the molecule, all of which are changing continuously and simultaneously, have a larger impact on conductance.

[1] H. Vazquez, J. Phys. Chem. Lett. 13 9326 (2022)
[2] E. Montes, W.Y. Rojas and H. Vázquez, J. Phys. Chem. C 129, 9947 (2025)

O 50.6 Tue 15:45 TRE/PHYS

Towards Accurate Surface Adsorption Through a Numeric-Orbital FDET Framework — •Danjo De Chavez[1] and Reinhard J. Maurer[1,2,3] — [1]Department of Chemistry, University of Warwick, UK — [2]Department of Physics, University of Warwick, UK — [3]Faculty of Physics, University of Vienna, AT

Multiscale embedding methods balance accuracy and computational efficiency in modeling extended materials by combining high-level treatments of active regions with lower-level descriptions of their environments. We implemented Frozen Density Embedding Theory (FDET) with Freeze-and-Thaw (F&T) cycles in the numeric atomic orbital code FHI-aims, enabling consistent cluster and periodic calculations. Initial electron densities are constructed from free-atom superpositions, with one subsystem kept frozen while the other is optimized. The implementation leverages the efficient parallelization of FHI-aims. Embedding potential and subsystem density are decomposed using an auxiliary basis set via an RI framework, facilitating reconstruction in the real space. This method enables accurate predictions with high-level theory for adsorption, bond dissociation, optical excitation, and charge-transfer phenomena relevant to surface catalysis and spectroscopy.

O 50.7 Tue 16:00 TRE/PHYS

Brillouin zone sampling in ONETEP — •Chengcheng Xiao, Peter Haynes, and Arash Mostofi — Department of Material, Imperial College London, London SW7 2AZ, United Kingdom

ONETEP is a parallelised linear-scaling density-functional theory (DFT) simulation software package designed for large-scale calculations of properties of materials. It was engineered under the premise

that, for self-consistent calculations of systems with thousands of atoms, the Brillouin zone only needs to be sampled at its centre - the Gamma-point.

Whilst this is a suitable approximation for large systems, it often leads to the unnecessary use of large supercells of structures where periodicity can be exploited. For those systems, a more suitable method is to sample the Brillouin zone along the periodic directions.

In this work, we introduce a new Brillouin zone sampling technique in ONETEP where the localisation constraint imposed on the basis functions is selectively lifted along the periodicity directions, making it possible to perform a "hybrid" calculation where the best scaling can be achieved by incorporating both Brillouin zone sampling and linear-scaling techniques. Furthermore, if the unit cell is small along all directions, all localisation constraints can be lifted, which effectively turns ONETEP into a plane-wave code. This enables us to compare the algorithmic accuracy of ONETEP to other DFT codes on the same footing. These developments enable much more efficient calculations on short and intermediate-scale periodic systems, such as bulk crystals, nanowires, surfaces, and interfaces.