

DS 41: Frontiers in Electronic-Structure Theory - Focus on Electron-Phonon Interactions V (joint session O/HL/DS/ CPP)

Time: Thursday 15:00–17:30

Location: GER 38

Invited Talk DS 41.1 Thu 15:00 GER 38

Huge quantum effects on the 250 K superconducting lanthanum hydride — ●ION ERREA — University of the Basque Country, Donostia/San Sebastián, Spain

The discovery of superconductivity at 200 K in the hydrogen sulfide system at large pressures was a clear demonstration that hydrogen-rich materials can be high-temperature superconductors. The recent synthesis of LaH₁₀ with a superconducting critical temperature (T_c) of 250 K place these materials at the verge of reaching the long-dreamed room-temperature superconductivity. Here we show that quantum atomic fluctuations stabilize in the superconducting pressure range a high-symmetry Fm-3m crystal structure consistent with experiments, which has a colossal electron-phonon coupling of 3.5. Even if *ab initio* classical calculations predict this structure to distort below 230 GPa yielding a complex energy landscape, the inclusion of quantum effects evidences the Fm-3m as the true ground state. The agreement between the calculated and experimental T_c values further supports this phase as responsible for the 250 K superconductivity. The relevance of quantum fluctuations questions many of the crystal structure predictions made for hydrides within a classical approach that at the moment guide the experimental quest for room-temperature superconductivity. Furthermore, quantum effects are revealed to be crucial to stabilize solids with extraordinary electron-phonon coupling, which may otherwise be destabilized by the large electron-phonon interaction, reducing the pressures needed for their synthesis.

DS 41.2 Thu 15:30 GER 38

Self-Interaction Corrected SCAN for Solids: All-Electron Implementation with Numeric Atom-Centered Basis Functions — ●SHENG BI¹, IGOR YING ZHANG², CHRISTIAN CARBOGNO¹, and MATTHIAS SCHEFFLER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — ²Fudan University, Shanghai, China

For all semi-local density-functional approximations (DFAs), electronic self-interaction errors lead to an erroneous description of charge-transfer processes, a systematic underestimation of band gaps in semiconductors, and incorrect total energies [1]. These errors can be alleviated via localized-orbital scaling corrections [2] or via self-interaction corrections (SIC) [3]. In this work, we have implemented a reciprocal-space formulation of self-consistent SIC in the all-electron, numeric atomic-orbitals code *FHI-aims*, which is applicable for all semi-local DFAs, including the promising meta-GGA “strongly constrained and appropriately normed” (SCAN) functional [4]. We validate our implementation by inspecting charge transfer, cohesive energies, and band gaps for a test set of molecules and solids, showing that SIC considerably improves SCAN calculations and yields results on par with standard *GW* calculations at a fraction of the computational cost. This allows us to use SCAN-SIC for studying the adsorption of organic molecules on the H-Si(111) surface.

[1] A. J. Cohen *et al.*, *Chem. Rev.* **112**, 289 (2011).

[2] N. Q. Su *et al.*, *Proc. Natl. Acad. Sci.* **115**, 9678 (2018).

[3] Z. Yan *et al.*, *Phys. Rev. A* **95**, 052505 (2017).

[4] J. Sun *et al.*, *Phys. Rev. Lett.* **115**, 036402 (2015).

DS 41.3 Thu 15:45 GER 38

Understanding the lattice dynamics of 3D hexagonal boron nitride (h-BN): beyond the LDA approach — ●LUIGI CIGARINI, MICHAL NOVOTNÝ, and FRANTIŠEK KARLICKÝ — Department of Physics, Faculty of Science, University of Ostrava, Czech Republic

It is fundamental to achieve a clear depiction of the lattice dynamics of 3D h-BN in order to understand the experimental outcomes. Five different stacking conformations are possible for 3D h-BN and at least two or three of them are systematically present in samples in variable amounts [1-2], resulting as a source of irreproducibility for experiments, such as the infrared optical response [3-4].

The lattice dynamics of h-BN is particularly tough to describe, stated the different nature of the forces participating in it: covalent bonds and Van der Waals interactions. The LDA approach seemed to be the most effective compromise, at the DFT level [1,5-6].

In this work we explain the surprisingly good performance of LDA. We also show that it is possible to achieve better results, in comparison with experimental IR spectra, by using, instead, the GGA approach to

DFT and treating separately the two parts of the dynamical matrix. Besides, we found that IR spectroscopy is able to give some information about stacking composition. [1]. Liu, L. *et al.*, *Phys. Rev. B*, **68**(10), 104102 (2003). [2]. Constantinescu, G. *et al.*, *Phys. Rev. Lett.*, **111**(3), 036104 (2013). [3]. Çamurlu, H.E. *et al.*, *Ceram. Int.*, **42**(5), 6312-6318 (2016). [4]. Mukheem, A. *et al.*, *Nanomaterials*, **9**(4), 645 (2019). [5]. Cuscó, R. *et al.*, *Phys. Rev. B*, **97**(15), 155435 (2018). [6]. Serrano, J. *et al.*, *Phys. rev. Lett.*, **98**(9), 095503 (2007).

DS 41.4 Thu 16:00 GER 38

Error Estimation of Energy-per-Atom of Semiconductor Compounds Using Statistical Learning — ●DANIEL T. SPECKHARD^{1,2}, SVEN LUBECK², CHRISTIAN CARBOGNO¹, LUCA GHIRINGHELLI¹, CLAUDIA DRAXL^{1,2}, and MATTHIAS SCHEFFLER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — ²Humboldt-Universität zu Berlin, Institut für Physik and IRIS Adlershof, Berlin, Germany

Material databases such as NOMAD give researchers the ability to work with millions of material simulation results [1]. However, it is typically unclear to which extent calculations performed with different numerical settings and computer codes can be trusted and related to each other. This project presents statistical learning strategies to model errors in energies for two all-electron DFT codes, *FHI-aims* and *exciting*, for different basis-set sizes and k -point densities. Specifically, we use mutual information scores to select features that are able to capture the energy-per-atom errors. With respect to several metrics, random forest regression on the selected features shows the most promising results. This work lays the foundation for estimating errors in DFT data in NOMAD and helps to save computing resources by *a priori* predicting the DFT simulation settings required to achieve a desired level of precision. This also enables us to estimate the basis-set and k -point converged results of not fully converged calculations.

[1] C. Draxl and M. Scheffler, *J. Phys. Mat.*, **2** 036001 (2019). <https://nomad-coe.eu>

DS 41.5 Thu 16:15 GER 38

Force balance approach for advanced approximations in density functional theories — ●MARY LEENA TCHENKOUÉ¹, MARKUS PENZ¹, IRIS THEOPHILOU¹, MICHAEL RUGGENTHALER¹, and ANGEL RUBIO^{1,2} — ¹Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — ²Center for Computational Quantum Physics (CCQ), The Flatiron Institute, New York NY 10010, USA

We propose a systematic and constructive way to determine the exchange-correlation potentials of density-functional theories including vector potentials. The approach does not rely on energy or action functionals. Instead it is based on equations of motion of current quantities (force balance equations) and is feasible both in the ground-state and the time-dependent setting. This avoids, besides differentiability and causality issues, the optimized-effective-potential procedure of orbital-dependent functionals. We provide straightforward exchange-type approximations for different density functional theories that for a homogeneous system and no external vector potential reduce to the exchange-only local-density and Slater $X\alpha$ approximations.

DS 41.6 Thu 16:30 GER 38

Combining embedded mean field theory with linear-scaling density functional theory — ●JOSEPH PRENTICE^{1,2}, ROBERT CHARLTON², ARASH MOSTOFI², and PETER HAYNES² — ¹St Edmund Hall and Department of Materials, University of Oxford, Oxford, UK — ²Department of Materials, Department of Physics and the Thomas Young Centre, Imperial College London, London, UK

We demonstrate the capability of embedded mean field theory (EMFT) within the linear-scaling density-functional theory code ONETEP, which enables DFT-in-DFT quantum embedding calculations on systems containing thousands of atoms at a fraction of the cost of a full calculation. We perform simulations on a wide range of systems from molecules to complex nanostructures to demonstrate the performance of our implementation with respect to accuracy and efficiency. This work paves the way for the application of this class of quantum embedding method to large-scale systems that are beyond the reach of existing implementations.

DS 41.7 Thu 16:45 GER 38

Topological semimetallic phase in PbO_2 promoted by temperature — ●BO PENG¹, IVONA BRAVIĆ¹, JUDITH L. MACMANUS-DRISCOLL², and BARTOMEU MONSERRAT¹ — ¹Cavendish Laboratory, University of Cambridge, United Kingdom — ²Department of Materials Science and Metallurgy, University of Cambridge, United Kingdom

Materials exhibiting topological order host exotic phenomena that could form the basis for novel developments in areas ranging from low-power electronics to quantum computers. The past decade has witnessed multiple experimental realization and thousands of predictions of topological materials. However, it has been determined that increasing temperature destroys topological order, restricting many topological materials to very low temperatures and thus hampering practical applications. Here, we propose the first material realization of temperature promoted topological order. We show that a semi-conducting oxide that has been widely used in lead-acid batteries, β - PbO_2 , hosts a topological semimetallic phase driven by both thermal expansion and electron-phonon coupling upon increasing temperature. We identify the interplay between the quasi-two-dimensional nature of the charge distribution of the valence band with the three-dimensional nature of the charge distribution of the conduction band as the microscopic mechanism driving this unconventional temperature dependence. Thus, we propose a general principle to search for and design novel topological materials whose topological order is stabilized by increasing temperature. This provides a clear roadmap for taking topological materials from the laboratory to technological devices.

DS 41.8 Thu 17:00 GER 38

How Electric Fields Affect Intermolecular van der Waals Interactions — ●MOHAMMAD REZA KARIMPOUR, DMITRY FEDOROV, and ALEXANDRE TKATCHENKO — University of Luxembourg, 1511 Luxembourg, Luxembourg

van der Waals (vdW) dispersion interactions between atoms or molecules originate from electromagnetic forces caused by the zero-point quantum-mechanical fluctuations of electronic charge densities. They are ubiquitous in nature and present in many areas of physics, chemistry, biology, and nanotechnology. Recently, it has been shown that the strength of vdW interactions can be controlled and tailored

by external electric charges [1]. In addition, an external field strongly modifies the dispersion interaction between two hydrogen atoms and can change both its spatial dependence and its attractive or repulsive character [2]. To describe such important phenomena in large molecular systems, we employ the Many-Body Dispersion (MBD) method [3] based on the quantum Drude oscillator model. Since the conventional MBD method includes only dipole-dipole coupling, it does not capture the effects of external fields on vdW interactions. Therefore, we first extend the approach to dipole-quadrupole and quadrupole-quadrupole couplings. Then, the developed formalism is applied to calculate the MBD energy in the presence of an external electric field for low-dimensional systems including bilayer graphene.

[1] Kleshchonok and Tkatchenko, *Nat. Commun.* **9**, 3017 (2018)[2] Fiscelli *et al.* arXiv:1909.03517 (2019)[3] Tkatchenko *et al.* *Phys. Rev. Lett.* **108**, 236402 (2012)

DS 41.9 Thu 17:15 GER 38

Electronic structure of β -SiAlON: effect of Al/O doping and of finite temperature — ●SALEEM AYAZ KHAN¹, ONDREJ ŠÍPR², JIŘÍ VACKÁŘ², ROBIN NIKLAUS³, WOLFGANG SCHNICK³, and JAN MINÁR¹ — ¹University of West Bohemia, Plzen, Czech Republic — ²Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic — ³LMU Munich, Germany

Electronic structure of a series of ordered and disordered β -Si_{6-z}Al_{z}O_{z}N_{8-z} systems is investigated by means of ab initio calculations, using the FLAPW method as implemented in the wien2k code and Green function KKR method as implemented in the SPR-KKR code. Finite temperature effects are included within the alloy analogy model. We found that the trends with the Al/O doping are similar for ordered and disordered structures. The electronic band gap decreases with increasing z by about 1 eV when going from z=0 to z=2. The optical gap decreases analogously as the electronic band gap. The changes in the density of states (DOS) at Si and N atoms introduced by doping β -Si_{3}N_{4} with Al and O are comparable to the DOS at Al and O atoms themselves. The bottom of the conduction band in β -Si_{6-z}Al_{z}O_{z}N_{8-z} is formed by extended states residing on all atomic types. Increasing the temperature leads to a shift of the bottom of the conduction band to lower energies. The amount of this shift increases with increasing doping z.