O 82: Poster Focus Session: Frontiers of Electronic-Structure Theory - Correlated Electron Materials

Time: Wednesday 18:15–20:30 Location: Poster A

O 82.1 Wed 18:15 Poster A

Implementation of the Strongly Constrained and Appropriately Normed Semilocal Density Functional (SCAN) with Self Interaction Correction (SIC) — •Lenz Fiedler, Jens Kortus, Torsten Hahn, and Sebastian Schwalbe — TU Freiberg, Institute for Theoretical Physics, Freiberg, Germany

Density Functional Theory (DFT) has become the most important computational method for the calculation of electronic structure. In order to calculate the exchange and correlation energy of a given system, there exists a wide variety of functionals. One of the recent developments in that area is the introduction of the SCAN functional [1]. Even such modern exchange-correlation functionals like SCAN are still subject to the self-interaction error [2]. The recently proposed Fermi-Löwdin orbital method for self-interaction correction (FLO-SIC)[3-4] seeks to correct this self-interaction error.
We therefore present the results of an implementation of the SCAN functional in the Naval Research Laboratory Molecular Orbital Library (NRLMOL). This implementation is used to verify the computational properties of the SCAN functional as well as to enable the combination of SCAN and FLO-SIC in order to further improve the results of DFT calculations.
[1] J. Sun, A. Ruzsinszky, J. Perdew, PRL, vol. 115, 036402 (2015)
[2] J. Perdew, A. Zunger, Phys. Rev. B 23, 5048 (1981)
[3] M. R. Pederson et al., J. Chem. Phys., vol. 140, 121103 (2014)
[4] M. R. Pederson, J. Chem. Phys., vol. 142, 064112 (2015)

O 82.2 Wed 18:15 Poster A

Fermi-Löwdin orbital self-interaction corrected DFT for molecules — •Jakob Kraus, Sebastian Schwalbe, Torsten Hahn, and Jens Kortus — TU Bergakademie Freiberg, Institute for Theoretical Physics, Germany

One of the more recent methods of addressing the self-interaction error (SIE) in density functional theory (DFT), namely the Fermi-Löwdin orbital self-interaction correction (FLO-SIC [1]), provides significant improvements in total energies, level ordering and ionization potentials [2]. Notably, the generated Fermi-Löwdin orbitals are suspected to hold actual chemical meaning. Furthermore, there is evidence implying that FLO-SIC results allow for an interpretation in terms of the Lewis formulae of common molecules. In this contribution, we deliver FLO-SIC structural formulae of neutral and charged diatomic molecules which, from a chemist's point of view, do not possess an adequate Lewis formula. Given these results, we propose a FLO-SIC-based bond order scheme.

[1] M. R. Pederson et al., JCP, vol. 140, 121103 (2014)
[2] T. Hahn et al., JCP, vol. 143, 224104 (2015)

O 82.3 Wed 18:15 Poster A

Force balance equation and exchange-correlation potentials in time-dependent current density functional theory(tdCDFT) — •Mary-Leena Martine Tchenkoue Djouom, Markus Penz, Michael Ruggenthaler, and Angel Rubio — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

The Runge-Gross theorem as the conceptual basis of tdCDFT is based on a force balance equation from which the existence of a unique mapping between potentials (including a vector potential) on the one side and one-particle densities and currents on the other side is derived. Since tdCDFT is expected to solve important problems that persist in tdDFT (without regarding vector potentials and currents) the study of this force equation is of special interest. Here we use the force balance equation to derive approximate exchange-correlation potentials that obey basic conditions such as zero-force and zero-torque. Such approximations will be useful in the calculation of dynamical properties of many-particle systems.

O 82.4 Wed 18:15 Poster A

Reliable magnetic properties for spin-crossover complexes with Fermi-Löwdin orbital self-interaction corrected DFT ? — •Julia Richter, Sebastian Schwalbe, Jens Kortus, and Torsten Hahn — TU Freiberg, Institute for Theoretical Physics, Germany

Octahedral Fe^{2+} molecules are particularly interesting as they often exhibit a spin-crossover transition. The existing semilocal exchange-correlation functionals used in density functional theory (DFT) calculations are known to fail badly in predicting the energy differences between the various spin states of these molecules. We apply the recently developed Fermi-Löwdin orbital self-interaction correction (FLO-SIC) [1,2] to $[Fe(H_2O)_6]^{2+}$, $[Fe(NH_3)_6]^{2+}$, $[Fe(NCH)_6]^{2+}$ and $[Fe(CO)_6]^{2+}$ complexes and we benchmark against non-SIC corrected DFT and diffusion Monte Carlo results [3]. We expect, that the correct treatment of the exchange energy within the FLO-SIC method leads to improved description of spin states.
[1] M. R. Pederson et al., J. Chem. Phys., vol. 140, 121103 (2014)
[2] M. R. Pederson, J. Chem. Phys., vol. 142, 064112 (2015)
[3] Droghetti, A. et. al., J. Chem. Phys., vol. 137, 124303 (2012).

O 82.5 Wed 18:15 Poster A

Towards chemical accuracy of FLO-SIC — •Sebastian Schwalbe[1], Torsten Hahn[1], Simon Liebing[1], Kai Trepte[2], and Jens Kortus[1] — [1]TU Bergakademie Freiberg, Institute for Theoretical Physics, Germany — [2]Central Michigan University, Department of Physics, USA

Fermi-Löwdin orbital self-interaction correction (FLO-SIC) [1,2,3] is applied to atoms and molecules as defined in the standard G2-1 test set. Within this systematic study calculated ionization potentials and binding energies are compared with experimental values. It will be shown that the FLO-SIC method provides significant improvements towards chemical accuracy by using the framework with the LSDA functional, but noteworthy worse performance in combination with the PBE-GGA or other GGA functionals.

[1] M. R. Pederson et al., JCP, vol. 140, 121103 (2014)
[2] T. Hahn et al., JCP, vol. 143, 224104 (2015)
[3] T. Hahn et al., JCTC (2017)

O 82.6 Wed 18:15 Poster A

Improvements for FLO-SIC: Electron force field (eFF) and Frozen-Core Approximation — •Sebastian Schwalbe[1], Simon Liebing[1], Kai Trepte[2], Torsten Hahn[1], and Jens Kortus[1] — [1]TU Bergakademie Freiberg, Institute for Theoretical Physics, Germany — [2]Central Michigan University, Department of Physics, USA

The Fermi-Löwdin orbital self-interaction correction (FLO-SIC) [1,2,3] is computational limited by the time needed for the optimization of reference positions in real space, namely the Fermi-Orbital-descriptors (FODs). Using the electron force field (eFF) for pre-optimization of these positions enables a significant speed-up of any FLO-SIC calculation. Further, the authors give a short review on other speed-up possibilities. In particular a so called Frozen-Core FLO-SIC method is discussed. Finally, a general speed-up methodology combining all the aforementioned techniques is proposed.

[1] M. R. Pederson et al., JCP, vol. 140, 121103 (2014)
[2] T. Hahn et al., JCP, vol. 143, 224104 (2015)
[3] T. Hahn et al., JCTC (2017)

O 82.7 Wed 18:15 Poster A

Ab-initio structural relaxation of molecular crystals under pressure — •Eric Pierschel — Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, Germany

The structural relaxation of molecular crystals presents serious difficulties. In principle, forces acting on the atoms, calculated as energy gradients, allows for the search of the molecular configuration with the minimum possible energy. The drawback of this method is that the forces between atoms belonging to a single molecule are significantly stronger than the ones between atoms in different molecules. This makes the numerical determination of the orientation of molecules with respect to each other very difficult. To avoid this problem, we use a cluster algorithm which, first, detects individual molecules. Then, independent translations and rotations for each molecule are added to the list of the standard $3N$ degrees of freedom, corresponding to the N

atoms in the unit cell. The structure optimization is performed in this overdetermined parameter space by using the BFGS-algorithm. Optionally, our procedure allows for performing the optimization of the molecular degrees of freedom only, by considering each molecule as a rigid rotator. We use this approach for relaxing molecular crystals, like the oligo-acens, under pressure. *Ab-initio* total energies are calculated using density-functional theory including van-der Waals interactions as implemented in the full-potential all-electron code `exciting`.

O 82.8 Wed 18:15 Poster A

FLO-SIC DFT applied to pseudopotentials — •Simon Liebing, Sebastian Schwalbe, Torsten Hahn, and Jens Kortus — TU Bergakademie Freiberg, Institute for Theoretical Physics, Germany

A method bases on Fermi-Löwdin orbitals to correct the self-interaction error within DFT (FLO-SIC) was developed by Pederson and co-workers. [1,2,3] The authors show how the FLO-SIC method can be used within the pseudopotential methodology. The calculation speedup and accuracy in comparison with all-electron calculations are discussed in detail. The FLO-SIC pseudopotential scheme leads to a similar improvement of the ionization potentials, eigenvalues and bond energies compared to the all-electron variant.

[1] M. R. Pederson et al., JCP, vol. 140, 121103 (2014)
[2] T. Hahn et al., JCP, vol. 143, 224104 (2015)
[3] T. Hahn et al., JCTC (2017)

O 82.9 Wed 18:15 Poster A

Self-energy self-consistency within density functional theory + dynamical mean field theory — •Sumanta Bhandary[1] and Karsten Held[2] — [1]Centre de Physique Thèorique, Ecole Polytechnique, 91128 Palaiseau, France — [2]Institute of Solid State Physics, TU Wien, 1040 Wien, Austria

We study the effects of self-energy self-consistency within a framework combining density functional theory (DFT; Wien2k) and dynamical mean field theory (DMFT; w2dynamics) in a basis of maximally localized Wannier orbitals. In addition to the charge density correction in DFT+DMFT charge self-consistency [1], in this scheme, we re-write the DFT exchange correlation potential in a form of a local self-energy, obtained in DMFT. The method is applied to transition metal oxides, involving a so called $d + p$ calculations. The overestimated $d - p$ hybridization in DFT is greatly improved, correcting the relative energy positions of the d and the p orbitals. Importantly, in this methodology, the explicit form of the exchange correlation potential is known and hence the ambiguity of the troublesome 'double-counting' term in regular DFT+DMFT formalism is avoided.

[1] S. Bhandary, E. Assmann, M. Aichhorn, and K. Held, Phys. Rev. B **94**, 155131 (2016).

O 82.10 Wed 18:15 Poster A

Vacancy charged defects in two-dimensional GaN: An ab-initio study — •Rafael Gonzalez-Hernandez[1], William Lopez-Perez[1], Alvaro Gonzalez-Garcia[1], and Maria Guadalupe Moreno Armenta[2] — [1]Depament of Physics, Universidad del Norte, Barranquilla, Colombia — [2]Centro de Nanociencias y Nanotecnología-UNAM, Km 107 carretera Tijuana-Ensenada, Ensenada, México

In this work, we have studied the structural and electronic properties of vacancy charged defects in the graphene phase (honeycomb type) of gallium nitride (g-GaN) by using first-principles calculations within the framework of the Density Functional Theory [1]. It is found that the vacancies introduce defect levels in the band gap, and these generate a total magnetization in the g-GaN system. The formation energy with different charge states for the vacancies of gallium and nitrogen were calculated, obtaining higher energies than the GaN wurtzite phase (w-GaN). Furthermore, nitrogen vacancies were found to be more stable than gallium vacancies in a whole range of electronic chemical potential. Gallium and nitrogen vacancies produce a nonzero magnetic moment in g-GaN, making it a potential candidate for future spintronics applications. In addition, free-standing g-GaN has been overcome by Al Bolushi et al. [2] using a migration-enhanced growth (MMEG) method in which two- dimensional GaN layers are created beneath the graphene layer generated by the Si sublimation form SiC(0001) sample.

References [1] Giannozzi et al., J. Phys.:" Condens. Matter 21 (2009) 395502 [2] Z. Al Balushi et al, "Two-dimensional gallium nitride realized via graphene

O 82.11 Wed 18:15 Poster A

Convergence of Kohn-Sham construction in lattice tdDFT — •Benedikt Mehmel, Michael Ruggenthaler, Markus Penz, and Angel Rubio — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg

In this master project we use the simple model of a two-site lattice system with two interacting particles to study the mathematical properties of the Kohn-Sham system in time-dependent density function theory. The necessary functional analytic tools include Fréchet differentiability and they are employed to show convergence of the self-consistent Kohn-Sham construction as a fixed point scheme. Such tools are also used to study the possibilty of a Runge-Gross theorem via the inverse function theorem applied to the potential-density map. As a side product exact conditions on possible approximations to the exchange-correlation potential in the scope of this simple model are established.

O 82.12 Wed 18:15 Poster A

Density Response to a Phonon Perturbation from DFPT Using the All-Electron FLAPW Method — •Christian-Roman Gerhorst, Markus Betzinger, Gustav Bihlmayer, Daniel Aaron Klüppelberg, and Stefan Blügel — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

A significant milestone in the calculation of phonon dispersions within Density Functional Perturbation Theory (DFPT) is the self-consistent solution of the Sternheimer equation. The resulting expansion coefficients of the response of the wavefunctions describe the linear variation of the density induced by a phonon perturbation. Using the all-electron Full-potential Linearized Augmented Plane-Wave (FLAPW) method, as implemented in the FLEUR code, enables an accurate treatment of complex materials without approximations to the 1/r potential or to the core electrons, but at the expense of additional conceptual challenges due to atom-positioned basis functions. In this contribution, we resolve computational challenges and show results of the self-consistently iterated Sternheimer equation reporting especially on the first variation of the density due to a phonon perturbation. In addition, we briefly shed light on the second-order quantities required for the dynamical matrix and focus on how the choice of the FLAPW method influences the shape of their equations.

O 82.13 Wed 18:15 Poster A

Calculating temperature-dependent resistivities in the KKR formalism: Implementation and application to simple metals — Carsten Eberhard Mahr, •Michael Czerner, and Christian Heiliger — Justus-Liebig-University, Giessen, Germany

Electron-phonon coupling is one of the main incoherent scattering mechanisms in a wide variety of crystalline material systems at room temperature and above. Therefore, it is necessary to incorporate those effects in any realistic calculations of electronic transport properties. To do so, we have recently reported [Phys. Rev. B 96, 165121 (2017)] an extension to our density functional theory (DFT) based Korringa-Kohn-Rostocker (KKR) Green's function formalism code.

By approximating the Fröhlich-type interaction by a suitably wavevector-averaged energy-dependent self-energy Σ_{eph} we are able to compute the dressed propagator $G = G_{\mathrm{ref}} + G_{\mathrm{ref}} \cdot \left(\Delta V + \Sigma_{\mathrm{eph}}\right) \cdot G$ by solving Dyson's equation, where G_{ref} is the Green's function of an arbitrary (though typically repulsive) reference system. The averaged self-energy is extracted from first principles electron linewidth calculations employing Quantum ESPRESSO and EPW.

We demonstrate the physical validity of the beforementioned calculational scheme for non-equilibrium properties by comparing evaluated temperature-dependent resistivities of copper and aluminum to experimental data. Further, technical details of the implementation in the KKR basis set are presented.

O 82.14 Wed 18:15 Poster A

Developing Density Functional Based Tight-Binding Methods within a Self-Consistent Field Density Functional Theory Framework — •Frederick Stein, Jens Kunstmann, and Gotthard Seifert — TU Dresden, Germany

The Density-Functional Tight-Binding (DFTB) method is an approximate density functional theory scheme that is routinely used in computer simulations with large system sizes or long simulation times. The method traditionally exists in a non-selfconsistent (DFTB0) and a charge-selfconsistent (SCC-DFTB) flavor. Here we develop a self-consistent field DFTB (SCF-DFTB) scheme that is very general and directly allows improvements of traditional DFTB methods. We demonstrate how the SCC-DFTB formalism can be derived from SCF-DFTB without the use of a second-order expansion of the total energy. This leads to a new interpretation of SCC-DFTB and the involved Hubbard parameter in terms of a flexible basis set. Furthermore we use small molecules to study SCF-DFTB variants with spherical and non-spherical corrections of the DFTB0 Hamiltonian. Our results indicate that the success of DFTB is based on multiple, well-balanced approximations. Attempts to only correct individual approximations are not leading to better results.

O 82.15 Wed 18:15 Poster A

Considerations About the Gauge Factor in TDDFT-Ehrenfest MD with Non-Local Pseudopotentials — •Lukas Deuchler and Eckhard Pehlke — Institut für Theoretische Physik und Astrophysik, CAU Kiel

It is well known that non-local pseudopotentials (PPs) require a gauge factor when a vector potential is introduced into the Kohn-Sham equations [1]. In PP-based TDDFT-Ehrenfest molecular dynamics simulations, the motion of the ions leads to a similar issue [2,3]. In case of norm-conserving PPs, which are applied in the fully separable Kleinman-Bylander form, Avendaño-Franco has stated in his thesis [4] that the non-local PP has to be multiplied by a phase factor with a phase proportional to the ion velocity to derive the dynamic ground state for TDDFT-MD. Here we argue that Galilei invariance of the TDDFT equation of motion requires this gauge factor to be applied to the non-local PP during the time propagation of the Kohn-Sham states. Quantitatively, the effect of omitting the gauge factor during propagation (while still correctly including the initial boost of the Kohn Sham states) appears to be small in case of the semilocal Troullier-Martins PPs we have tested, consistent with [4]. The TDDFT-MD calculations have been carried out with the code Octopus [5].

[1] Bertsch et al., Phys. Rev. B 62, 7998 (2000).
[2] Qian et al., Phys. Rev. B 73, 035408 (2006).
[3] Ojanperä et al., J. Chem. Phys. 136, 144103 (2012).
[4] Avendaño-Franco, PhD Thesis, UC Louvain (2013).
[5] Andrade et al., Phys. Chem. Chem. Phys. 17, 31371 (2015).