SYEC 3: Exact-exchange and hybrid functionals meet quasiparticle energy calculations III - Poster (joined by SYMS posters)

Time: Thursday 18:30–19:30 Location: Poster F

SYEC 3.1 Thu 18:30 Poster F

Implementation of exact exchange in a real-space DFT code using the projector augmented wave method — •Carsten Rostgaard and Jens Jørgen Mortensen — Center for Atomic-scale Materials Design, Technical University of Denmark, Denmark

The projector augmented wave (PAW) method for density-functional theory (DFT) calculations, offers a formally well defined linear transformation from the sharply featured all-electron wave function to the smooth pseudo wave function, thus enabling a computationally efficient approach to solving the Kohn-Sham equations, while retaining the accuracy of a full potential (frozen core) calculation.

As DFT calculations with orbital dependent exchange-correlation functionals probe the core region of the wave function, the PAW scheme is ideally suited for such functionals. For exact exchange calculations in PAW, one needs to determine the action of the non-local Fock operator on the smooth pseudo wavefunctions. The Fock operator can then be applied directly, as is done in most hybrid functionals, of it can be 'localized' (i.e. made multiplicative) by some appropriate scheme, like OEP, LHF, KLI, etc.

We have implemented the Fock operator in the real-space PAW-DFT code gpaw, and a benchmark test of the atomization energies and HOMO-LUMO gaps of a number of small molecules has been performed, and compared to calculations done in VASP and Gaussian, using the PBE0 hybrid functional. We find that the effect of the valence-core exchange interaction, available in the PAW framework, can be substantial.

SYEC 3.2 Thu 18:30 Poster F

Study of electronic properties and point defects in UO_2 using local hybrid exchange-correlation functionals — •Doru Torumba[1], Jean-Paul Crocombette[1], and François Jollet[2] — [1]CEA Saclay, DEN/DMN/SRMP, 91191 Gif-Sur-Yvette, France — [2]Département de Physique Théorique et Appliquée, CEA-DAM, Bruyères-le-Châtel, France

The new computational methods, as the local hybrid exchange-correlation functionals, require a careful benchmarking of their performance on many different systems in order to asses their reliability.

In the present contribution the local version of the hybrid PBE0 exchange-correlation functional is used for calculating the structural and magnetic properties of uranium dioxide. The results are compared to the ones obtained using the more common functionals: GGA and LDA+U. Additionally, the results of an ongoing study on the charge and the structure of point defects in uranium dioxide will be presented.

SYEC 3.3 Thu 18:30 Poster F

Role of emerging methods in advanced DFT study of novel compounds with high-efficiency photovoltaic applications — •Pablo Palacios[1,2], Perla Wahnon[1], Kefren Sanchez[1], Irene Aguilera[1], Julio Juan Fernandez[1], and Jose Carlos Conesa[2] — [1]Universidad Politecnica de Madrid, Madrid, Spain — [2]Consejo Superior de Investigaciones Cientificas, Cantoblanco, Spain

An exhaustive study of compounds with unusual electronic structure used as new efficient photovoltaic materials is presented. These intermediate band materials obtained by selective substitution of atoms of a host semiconductor by transition metals present a narrow, partially-filled band inside the semiconductor band-gap, separated from host valence and conduction bands. Therefore additional current carriers can be obtained from the absorption of low energy photons enabling electrons to get the conduction band through the intermediate band.

Although standard DFT is a suitable tool for our purposes, in order to enhance prediction capabilities for these new materials there are some features, i.e. host semiconductor gap, which can be improved with advanced methods let be GW, exact exchange or hybrid functionals.

Exact exchange studies have been done in several candidates for intermediate band material as well as advanced analysis of the correlation effect. Exact exchange method was implemented in our group within SIESTA. For the moment these studies have contributed to support the reliability of the prediction of intermediate band materials to such an extent that experimental samples are being grown. State-of-the-art techniques could complement the results obtained to date.

SYEC 3.4 Thu 18:30 Poster F

Implementation of the non-local exchange potential within the FLAPW method — •M. Betzinger, C. Friedrich, G. Bihlmayer, and S. Blügel — Institut für Festkörperforschung, Forschungszentrum Jülich, 52425 Jülich, Germany

Hybrid functionals (e.g. PBE0, B3LYP) are currently explored as a practical approximation to the exchange-correlation functional in density-functional theory (DFT). They yield accurate results for molecules, improve the band gap of semiconductors and insulators as well as the description of strongly correlated systems. So far most of the implementations for periodic systems employ pseudopotentials with a plane-wave basis set, which restricts their range of applicability. We develop an implementation within the full-potential linearized augmented plane-wave method (FLAPW), with which a much wider range of materials can be treated. The crucial step is the evaluation of the non-local exchange potential. Our approach uses a mixed basis-set. After representing the Coulomb interaction in this basis, the exchange matrix elements can be written as a Brillouin-zone (BZ) integral over vector-matrix-vector products. The Coulomb interaction goes to infinity in the center of the BZ leading to a divergence in the integrand. The divergent part is separated from the numerical integration and treated analytically. We show convergence properties of the mixed basis-set and first results for prototype semiconductors and insulators.
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SYEC 3.5 Thu 18:30 Poster F

Two-particle excitations within the Hubbard model — Berlinson Dominikus Napitu[1,2] and •Jamal Berakdar[2] — [1]Martin-Luther-Universität, Halle-Wittenberg,Germany — [2]Max-Planck-Institut für MikrostrukturPhysik, Halle

Remarkable progress has been achieved recently in studying experimentally the two-particle spectrum of surfaces via angular and energy-resolved two-electron emission upon [1,2] a single-photon absorption. We show in this theoretical contribution how these measurements correlate with the particle-particle Green's function and contrast with related spectroscopies such as core-valence-valence (CVV) Auger spectra and appearance potential spectroscopy (APS). Based on the single band Hubbard model [3] we developed a method for the calculations of the corresponding two-particle spectra while the single particle properties are being evaluated within dynamical-mean field theory (DMFT)scheme [4].

[1] Schumann, F.O., Winkler, C., Kirschner, J., Phys. Rev. Lett. 98, 257604(2007); [2] Schumann, F.O., Winkler, C., Kirschner, J., New. Jour. Phys. 9.372/1-14 (2007); [3] Hubbard, J.I., Proc. Roy. Soc. London 276, 238 (1963); [4] Georges, A., et al., Rev. Mod. Phys.68, 13 (1996).

SYEC 3.6 Thu 18:30 Poster F

Optical Excitation of the F Center in Calcium Fluoride Within Many-Body Perturbation Theory — •Yuchen Ma and Michael Rohlfing — Fachbereich Physik, Universität Osnabrück, Osnabrück, Germany

As a prototype for defects in insulators, we discuss the optical properties of the F center in calcium fluoride (CaF_2), which constitutes a prominent defect of the material. The F center of CaF_2 exhibits a defect state deep in the band gap. Its excitation by light (at 3.3 eV excitation energy) is described by *ab-initio* many-body perturbation theory (GW approximation and Bethe-Salpeter equation), including electronic exchange, correlation, and electron-hole interaction effects. The excitation can be regarded as a $1s \rightarrow 2p$ transition. The exciton of the absorption band is strongly localized around the vacancy (within about 2 Å for the hole and 4 Å for the electron), but they induce prominent lattice distrotion around the defect. Constrained density-functional theory can predict reliable geometric relaxation for the localized excitons in ionic insulators. The excitation causes strong relaxation of the defect geometry, leading to significant broadening of the optical spectrum by 0.5 eV and to an large Stokes shift of 1.5 eV.

SYEC 3.7 Thu 18:30 Poster F

Physisorption of Organic Molecules on Noble-Metal Surfaces — •Michael Rohlfing[1], Thomas Bredow[2], and Florian Janetzko[2] — [1]Fachbereich Physik, Universität Osnabrück, Germany — [2]Institute for Physical and Theoretical Chemistry, Universität Bonn, Germany

We discuss the physisorption of organic molecules to noble-metal surfaces due to Van-der-Waals interaction. Different from density-functional theory, we treat the exchange energy explicitly and calculate the correlation part within the random-phase approximation. In here, the response of the metal surface is described by a combination of a jellium surface and local polarizability of the d electrons, while the response of the adsorbate is obtained from a DFT calculation. We discuss results for the adsorption of Xe, benzene, and PTCDA on the Ag(111) surface. The obtained adsorption energies and distances are between those of DFT-LDA (which overbinds) and DFT-PBE (which does not bind at all), thus constituting a significant improvement beyond DFT. The correlation energy obtained with RPA is compared to classical R^{-6} correction terms. Although the empirical approach gives qualitatively similar potential curves, RPA better reproduces experimental adsorption energies and distances.