

O 21: Methods: Electronic structure theory I

Time: Tuesday 10:30–12:00

Location: SCH A316

O 21.1 Tue 10:30 SCH A316

Embedding based order-N implementation of the FLAPW method — ●FRANK FREIMUTH^{1,2}, DANIEL WORTMANN^{1,2}, and STEFAN BLÜGEL^{1,2} — ¹Institut für Festkörperforschung, Forschungszentrum Jülich, 52425 Jülich, Germany — ²Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany

The Full-potential Linearized Augmented Plane Wave (FLAPW) method is known to set the standard for precision among the *ab initio* density functional theory codes. On the other hand, the computational effort of the FLAPW method is high and increases cubically with system size, making the applicability to large systems difficult.

In this talk we will discuss a Green-function (GF) based implementation of the FLAPW method suitable for the calculation of thick layered systems. Using the embedding method the layers may be calculated individually and joined together subsequently. The overall computational cost increases linearly with the number of layers. Our order-N GF implementation of the FLAPW method is able to fully replace the standard Bloch-function based implementation of the FLAPW method. Furthermore, the embedding concept allows to extend the applicability of the FLAPW framework to non-periodic infinite systems, e.g. surfaces and interfaces. Due to the high flexibility of the GF method general boundary conditions are easily imposed, e.g. the non-equilibrium situation arising in tunnel junctions under finite bias.

Besides the theoretical description of the method we give proof of evidence by presenting a set of applications for film, surface and transport calculations.

O 21.2 Tue 10:45 SCH A316

Scalar relativistic schemes for all-electron DFT with atom-centered basis functions — PAULA HAVU, ●VOLKER BLUM, and MATTHIAS SCHEFFLER — Fritz-Haber-Institut der Max-Planck-Gesellschaft, D-14195 Berlin, Germany

Numeric atom-centered orbitals are an efficient, accurate basis choice for all-electron electronic structure theory [1]. For seamless efficiency and accuracy, a one-component (two with spin) Schrödinger-like equation is computationally most convenient, but for most elements ($Z \gtrsim 30$), relativistic effects arising near the nucleus cannot be ignored. Dirac's equation can simply be rewritten in a "scalar-relativistic" (one-component) form, but with a separate Hamiltonian for each eigenstate. For some paradigm test systems [e.g., the Au dimer; CO adsorption on Pt(111)], we here benchmark the accuracy of a hierarchy of scalar-relativistic schemes that circumvent the state dependence: (i) the unsatisfactory "zero-order regular approximation" (ZORA), which simply neglects the state dependence; (ii) a restriction of ZORA to only the atomic center of each basis function ("atomic ZORA") and (iii) a perturbative rescaling of all ZORA eigenvalues ("scaled ZORA" [2]), which both recover geometries and binding energies within a few 10 meV of benchmark full-potential linearized augmented plane wave [FP-(L)APW] calculations; and (iv) a separate, exact treatment of all non-overlapping core states, which then necessitates only small further (scaled) ZORA-like approximations to the extended semicore and valence states. [1] V. Blum *et al.*, *Comp. Phys. Comm.*, accepted (2008). [2] E. van Lenthe *et al.*, *J. Chem. Phys.* **101**, 9783 (1994).

O 21.3 Tue 11:00 SCH A316

RPA atomization energies of extended systems — ●JUDITH HARL and GEORG KRESSE — Universität Wien, Austria

Density functional theory (DFT) applying the local density or generalized gradient approximation (LDA, GGA) leads to bond length errors of the order of 1 % and atomization energies are not reproduced with chemical accuracy. Furthermore, the (semi-)local LDA and GGA do not properly account for long-range van der Waals interactions.

Therefore, the adiabatic connection fluctuation dissipation theorem (ACFDT), which provides in principle an exact expression for the correlation energy, has received increased interest in the recent years (e.g., [1]-[3]). In the present work, we evaluate lattice constants and atomization energies of molecules and extended systems within the ACFDT framework applying the random phase approximation (RPA).

The RPA energy includes the exact exchange energy and a compatible correlation energy that also accounts for long-range van der Waals interactions. We found that the rare-gas solids Ne, Ar, and Kr are well described within the RPA and the correct long-range $1/V^2$ volume de-

pendence is reproduced. For insulators and metals, covalent and ionic bonded systems, RPA lattice constants are found to deviate on average by only 0.25 % from experiment. Atomization energies, however, are throughout too small (≈ 0.2 eV/atom) and the mean error remains close to the results obtained with DFT-PBE.

[1] F. Furche, *Phys. Rev. B* **64**, 195120 (2001). [2] A. Marini, P. García-González, A. Rubio, *Phys. Rev. Lett.* **96**, 136404 (2006) [3] J. Harl, G. Kresse, *Phys. Rev. B* **77**, 045136 (2008)

O 21.4 Tue 11:15 SCH A316

Real-Space DFT for Molecules on Surfaces — ●PAUL BAUMEISTER¹, TOMOYA ONO², DANIEL WORTMANN¹, and STEFAN BLÜGEL¹ — ¹Institut für Festkörperforschung, Forschungszentrum Jülich, 52425 Jülich, Germany — ²Department of Precision Science and Technology, Osaka University, Suita, Osaka 565-0871, Japan

We present a real-space code for density functional calculations that has been newly developed in Osaka and Jülich. Exploiting that most operations needed for electronic structure calculation are intrinsically local, the tasks on a real-space grid can be very efficiently parallelized by a domain decomposition scheme. In combination with iterative solvers, this favours the performance and scaling behaviour for large system sizes on massively parallel machines. So far our calculations have been based on the pseudopotential approach. Recently, the norm-conserving pseudopotentials have been replaced by the projector augmented wave (PAW) method proposed by Blöchl[1]. In order to save computational costs and to increase the accuracy, the double grid technique[2] is implemented to enable reasonably coarse grids.

The grid-based method is unrestricted with respect to boundary conditions, i.e. any combination of periodic and isolated boundary conditions can be selected which is well suited for calculating single molecules, wire structures and surface (film) setups without the usage of a super cell. We present the equilibrium structure of terephthalic acid adsorbed to a Cu(111) surface.

[1] P. E. Blöchl, *PRB* **50**, 17953 (1994)

[2] T. Ono, and K. Hirose, *PRL* **82**, 5016 (1999)

O 21.5 Tue 11:30 SCH A316

Adsorption of Super-heavy Elements on a Au(111) — ●JOSEF ANTON and TIMO JACOB — Institut für Elektrochemie, Uni Ulm, 89081 Ulm

Understanding the physical properties and the catalytic behavior of super-heavy elements is certainly an interesting but also challenging topic [1]. The major problems are connected with the rather short life time of these elements (≤ 1 s), resulting in very limited experiment times (before the nucleus decays) on only one atom at a time. While most standard techniques of 'traditional' chemistry are not applicable, gas-phase thermo-chromatography is one of the few remaining [1]. In this method the adsorption site of the super-heavy element is measured on a gold-covered detector whose endings are kept at different temperatures.

In order to achieve an accurate theoretical description of the super-heavy elements and their adsorption on the detector surface, the electrons of the systems have to be treated fully-relativistically. We used our four-component DFT code [2,3] to determine the adsorption energies of the elements 112 and 114 and their homologues, mercury and lead respectively, on Au(111) at various surface sites. Besides discussing these results, more general trends as well as a comparison to available experimental data will be given.

[1] *The Chemistry of Superheavy Elements*, Ed. M. Schädel, Kluwer Academic Publishers, Dordrecht (2003).

[2] J. Anton, B. Fricke, E. Engel, *Phys. Rev. A*, **69**, 012505 (2004).

[3] V. Pershina, J. Anton, T. Jacob, *Phys. Rev. A*, **78**, 032518 (2008).

O 21.6 Tue 11:45 SCH A316

Massive parallelization of the KKR Green-function method for large scale ab-initio calculations — ●ALEXANDER THIESS, RUDOLF ZELLER, and STEFAN BLÜGEL — Institut für Festkörperforschung and Institute for Advanced Simulation, Forschungszentrum Jülich, 52545 Jülich, Germany

Recent experimental progress in the analysis of oxide interfaces reveals the importance of complex defects arising from local disorder or oxy-

gen vacancies on transport and magnetic properties of the system [1]. For a theoretical treatment by density-functional calculations large unit cells containing thousands of atoms are unavoidable. As existing density-functional methods are usually restricted to a few hundreds of atoms in the unit cell, we have developed an algorithm based on the KKR Green-function method capable of treating geometries consisting of several thousands of inequivalent atoms. We combine the advantages of the screened KKR formalism and an iterative solution of the Dyson equation [2] with the new generation of massively parallelized

computers to obtain a parallelization over the number of atoms in the unit cell. Calculations of test systems with up to 4096 atoms in the unit cell show that our algorithm scales efficiently on the Jülich BlueGene computer JUGENE [3] for several thousands of processors.

- [1] A. Ohtomo, H. Y. Hwang, *Nature* **427**, 423 (2004)
- [2] R. Zeller, *J. Phys.: Condens. Matter* **20**, 294215 (2008)
- [3] <http://www.fz-juelich.de/jsc/jugene>