O 73: Focussed session: Frontiers of electronic structure theory: Strong correlations from first principles V (jointly with TT)

Time: Thursday 16:00–19:00   Location: HE 101

Topical Talk   O 73.1   Thu 16:00   HE 101
Symmetry Breaking and Restoration in Electronic Structure Theory — •Gustavo Scuseria — Rice University, Houston, Texas, USA

We have derived and implemented symmetry-projected Hartree-Fock-Bogoliubov (HFB) equations and apply them to the molecular electronic structure problem. All symmetries (particle number, spin, spatial, and complex conjugation) are deliberately broken and restored in a self-consistent variation-after-projection approach. We show that the resulting method yields a comprehensive black-box treatment of static correlation with effective one-electron (mean-field) computational cost. The ensuing wave function is of multireference character and permeates the entire Hilbert space of the problem. The energy expression is an independent quasiparticle density matrix functional. All reduced density matrices are expressible as an integration of transition one-particle density matrices over a gauge grid. I will present several proof-of-principle examples demonstrating the compelling power of this new method for electronic structure theory [1].

[1] Projected Quasiparticle Theory for Molecular Electronic Structure, G. E. Scuseria, C. A. Jimenez-Hoyos, T. M. Henderson, J. K. Ellis, and K. Samanta, J. Chem. Phys. 135, 124108 (2011).

O 73.2   Thu 16:30   HE 101
generator coordinate method to recover residual correlation energy — •Daniel Rohr and Gustavo Scuseria — Rice University, Houston, Texas, USA

Building on previous work in our group [1] we explore the possibilities to recover the residual correlation energy with the generator coordinate method (GCM). GCM allows for the expansion of the wave function in a set of non-orthogonal functions. In addition, the expansion functions may not be single slater determinants but rather multi-determinantal wave functions.

As input for the GCM we use wave functions generated by Projected Quasiparticle Theory (PQT) presented in the talk "Symmetry Breaking and Restoration in Electronic Structure Theory". The wave functions generated by PQT are multi-determinantal and non-orthogonal.

We explore the prospects of GCM to recover the residual correlation energy, not captured by PQT. Different generator coordinates are investigated for model hamiltonians and small test systems.

[1] Projected Quasiparticle Theory for Molecular Electronic Structure, G. E. Scuseria, C. A. Jimenez-Hoyos, T. M. Henderson, J.K. Ellis, and K. Samanta, *J. Chem. Phys.* **135, 124108 (2011).**

O 73.3   Thu 16:45   HE 101
reatment of strongly correlated systems within the framework of reduced density matrix functional theory — Sangeeta Sharma, •J. K. Dewhurst, and E. K. U. Gross — Max-Planck-Institut fur Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

One of the most dramatic failures of the usual local density approximation or generalized gradient type approximations to the exchange-correlation functional of density functional theory is the incorrect prediction of a metallic ground state for the strongly correlated Mott insulators, of which transition metal oxides (TMOs) may be considered as prototypical. In the present work we extend reduced density matrix functional theory (RDMFT) to the case of solid-state systems and introduce a new functional for their accurate treatment [1]. Furthermore, a method for calculating the spectrum of extended solids within RDMFT is presented and an application of this method to the strongly correlated TMOs demonstrates: (i) an insulating state is found in the absence of magnetic order and, in addition, (ii) the interplay between the charge transfer and Mott-Hubbard correlation is correctly described. In this respect we find 1. S. Sharma, J. K. Dewhurst, N. N. Lathiotakis and E. K. U. Gross Phys. Rev. B 78, 201103 Rapid Comm. (2008) 2. S. Sharma, S. Shallcross, J. K. Dewhurst and E. K. U. Gross cond-mat/0912.1118

O 73.4   Thu 17:00   HE 101
Treatment of spin in reduced density-matrix functional theory — •Nicole Helbig[1,2], Nektarios Lathiotakis[3], Stefan Kurth[2,4], and Angel Rubio[2] — [1]Peter Grünberg Institut and Insti-

tute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany — [2]NanoBio Spectroscopy group and ETSF Scientific Development Centre, Dpto. Física de Materiales, Universidad del País Vasco, Centro de Física de Materiales CSIC-UPV/EHU-MPC and DIPC, 20018 San Sebastián, Spain — [3]Theoretical and Physical Chemistry Institute, NHRF, Vass. Constantinou 48, 11635 Athens, Greece — [4]IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain

Many currently used approximations to the exchange-correlation energy in reduced density-matrix functional theory do not satisfy the spin constancy condition. Hence, these approximations yield different energies for ensembles of degenerate spin configurations both in an ensemble approach and a direct minimization of the energy. These findings are hardly surprising since the approximations were originally derived and optimized for closed shell systems. We discuss possible extensions to open-shell and spinor functionals.

O 73.5   Thu 17:15   HE 101
Kinetic-energy Functional of Interacting Electrons: Numerical Procedure and Its Statistical Interpretation — •Luca M. Ghiringhelli[1] and Luigi Delle Site[2] — [1]Fritz Haber Institute, D-14195, Berlin — [2]Institute for Mathematics, Freie Universität, Berlin

We introduce a numerical procedure, based on the extension of the Levy-Lieb constrained search principle and Monte Carlo sampling of electron configurations in space, for the derivation of the kinetic-energy functional for interacting particles. We consider a quasi-uniform gas of interacting electrons, for which spin statistics play a crucial role, and we show that when the fermionic character of the electrons is included via a statistical–spin approach, our procedure leads to correlation terms, that have the form of the Shannon entropy and the resulting kinetic-energy functional does satisfy the Lieb-Thirring inequality. By mapping the quantum problem onto a classical one, a statistical interpretation of the results is obtained that clarifies the possible physical meaning of the calculated quantity.

O 73.6   Thu 17:30   HE 101
Electron Tunneling - The Influence of Interaction — •Jessica Walkenhorst[1], Nicole Helbig[2,1], Heiko Appel[3], Angel Rubio[1], and E.K.U. Gross[4] — [1]NanoBio Spectroscopy group and ETSF, Universidad del País Vasco, San Sebastián, Spain — [2]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, Jülich, Germany — [3]NanoBio Spectroscopy group and ETSF, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [4]MPI of Microstructure Physics, Halle, Germany

Electron tunneling plays a fundamental role in many chemical and physical processes and provides evidence of quantum mechanics at the macroscopic level. Whilst much effort has been devoted to modelling electron tunneling, doing so in a non-perturbative way remains a major challenge. Even though time-dependent density functional theory (TDDFT) has become a popular tool to describe and predict many electronic properties, it still fails to describe the tunneling process correctly. This failure of TDDFT is due to the electron-electron interaction being described only approximately through the exchange-correlation potential. To elucidate this further, we investigate the fingerprints of electron-electron interaction in few electron systems during the tunneling process. This is done by exact time propagation of electronic wave functions in different external potentials. For computational feasibility, we consider one-dimensional model Hamiltonians with variable electron-electron interaction.

O 73.7   Thu 17:45   HE 101
Dynamical Vertex Approximation for Nanoscopic Systems — •Giorgio Sangiovanni, Angelo Valli, Georg Rohringer, Alessandro Toschi, and Karsten Held — TU Wien

With the aim of calculating strongly correlated materials more and more reliably, different novel approaches have been proposed in recent years. Quantum Cluster Theories turned out to be very successful for 2- and 3- dimensional Hubbard and related models. Complementary approaches, like Dual Fermion and Dynamical Vertex Approximation, classifiable as diagrammatic extensions of single-site Dynamical Mean Field Theory, become superior when long-range spatial correla-

tions need to be accurately described. An interesting application of Dynamical Vertex Approximation is the one for nanoscopic systems [Phys. Rev. Lett. 104, 246402 (2010)]. The increasing complexity of nanoscopic systems and the possibility of tuning their physical properties through nano-engineering constitute a new challenge for theories, in particular when strong electronic correlations are involved. I will discuss how Dynamical Vertex Approximation can be applied to such problems. I will show how to model a quantum point contact with 110 atoms and present our finding that the contact becomes insulating already before entering the tunneling regime due to a local Mott-Hubbard transition occurring on the atoms which form the point contact. I will also present results for the realistic case of a manganite nano-cluster where we combined our algorithm with a one- electron Hamiltonian calculated within Density Functional Theory [Phys. Rev. Lett. 107, 197202 (2011)].

O 73.8   Thu 18:00   HE 101
Full ab initio theory of the Kondo effect in molecular devices — •DAVID JACOB — MPI für Mikrostrukturphysik Halle

When a magnetic molecule is deposited on a metallic substrate or attached to metallic contacts its magnetic moment can actually be screened by the conduction electrons due to the Kondo effect. In view of possible applications of magnetic molecules for nanoscale spintronics and magnetic storage devices, it is important to reliably predict whether the Kondo effect will take place or not in a given system, and how it can be controlled by various parameters. From a more fundamental point of view, magnetic molecules in contrast to bulk systems offer the possibility to study exotic variants of the Kondo effect such as the orbital Kondo effect [1] or the underscreened Kondo effect [2]. Here we present a *full ab initio* method for molecular devices that combines the COHSEX approximation with more sophisticated many-body techniques like the One-Crossing-Approximation for treating the strong correlations of localized electrons that give rise to the Kondo effect. This method is a further development of our previous method [3,4] that was not yet fully ab initio due its dependence on the interaction between the localized electrons as a parameter. The application of our method to various magnetic molecules attached to metallic leads or deposited on surfaces sheds light on the complex nature of the Kondo effect in molecular-scale devices. [1] M. Karolak *et al.*, PRL **107**, 146604 (2011); [2] J. J. Parks *et al.*, Science **328**, 1370 (2010); [3] D. Jacob *et al.*, PRL **103**, 016803 (2009); [4] D. Jacob *et al.*, PRB **82**, 195115 (2010)

O 73.9   Thu 18:15   HE 101
Charge Kondo effect in PbTe doped with Tl impurities — •THEO COSTI[1] and VELJKO ZLATIC[2] — [1]Peter Grünberg Institut (PGI-2) and Institut for Advanced Simulation (IAS-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Institute of Physics, 10001 Zagreb, Croatia

Semiconducting PbTe is one of the most interesting materials for thermoelectric applications. When doped with a small concentration of Tl impurities, acting as acceptors, a number of anomalous properties are found: e.g.,beyond a critical concentration of about 0.3% Tl, the system exhibits superconductivity with remarkably high critical temperatures for such a low carrier system. This and other anomalous phenomena prompted the idea that Tl impurities act as negative U centres leading to a charge Kondo effect and to superconductivity. Here, we combine ab-initio information and numerical renormalization group methods to explore the consequences of this model for the normal state properties, showing that it can explain a number of features in the temperature and doping dependence of the resistivity, and carrier density. Predictions for the photoemission spectrum are also discussed [1].

[1] T. A. Costi and V. Zlatić, Phys. Rev. Lett. (arXiv:1109.1824).

O 73.10   Thu 18:30   HE 101
Critical properties of the half-filled Hubbard model in three dimensions — •GEORG ROHRINGER[1], ALESSANDRO TOSCHI[1], ANDREY KATANIN[2], and KARSTEN HELD[1] — [1]Institute for Solid State Physics, Vienna University of Technology 1040 Vienna, Austria — [2]Institute of Metal Physics, Ekaterinburg, Russia

By means of the dynamical vertex approximation(DΓA) [1] we include spatial correlations on all length scales beyond the dynamical mean field theory (DMFT) for the half-filled Hubbard model in three dimensions. The most relevant changes due to non-local fluctuations are [2]: (i) a deviation from the mean-field critical behavior with the same critical exponents as for the three dimensional Heisenberg (anti)-ferromagnet and (ii) a sizable reduction of the Néel temperature (T_N) by $\sim 30\%$ for the onset of antiferromagnetic order. Finally, we give a quantitative estimate of the deviation of the spectra between DΓA and DMFT in different regions of the phase-diagram.
[1] A. Toschi, A. Katanin, and K. Held, PRB 75, 045118 (2007).
[2] G. Rohringer, A. Toschi, A. Katanin, and K. Held, PRL in press [arXiv:1104.1919].

O 73.11   Thu 18:45   HE 101
Study of strontium vanadate using a dynamical cluster approximation in the linear augmented plane-wave basis — •HUNPYO LEE[1], KATERYNA FOYEVTSOVA[1], JOHANNES FERBER[1], MARKUS AICHHORN[2], HARALD O. JESCHKE[1], and ROSER VALENTÍ[1] — [1]Institute of Theoretical Physics, University of Frankfurt, Frankfurt, Germany — [2]Institute of Theoretical and Computational Physics, TU Graz, Graz, Austria

Even though the local density approximation in combination with a single-site dynamical mean field theory (LDA+DMFT) explains the Mott transition and the enhancement of the effective mass in real compounds, a numerical approach taking spatial correlations into account would be desirable. In this talk, we introduce a combination of LDA with dynamical cluster approximation (LDA+DCA) in the framework of the full-potential linear augmented plane-wave basis in WIEN2k and apply it to $SrVO_3$. We compare our LDA+DCA results with LDA+DMFT as well as with angle-resolved photoemission spectra (ARPES). We find that LDA+DCA results compare better with ARPES than the LDA+DMFT results due to the inclusion of short-range spatial correlations.