O 62: Focus Session: Frontiers of Electronic-Structure Theory: Correlated Electron Materials IV (joint session O/MM/DS/TT/CPP)

Organizers: Silke Biermann, Ecole Polytechnique, Palaiseau cedex, France; Paul R. Kent, Oak Ridge National Laboratory, USA; Matthias Scheffler, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin

(Synopsis provided with part I of this session)

Time: Wednesday 10:30–13:00 Location: HL 001

Invited Talk O 62.1 Wed 10:30 HL 001

Correlating electrons via adiabatic connection approach: a general formalism, approximations, and applications — •Katarzyna Pernal — Institute of Physics, Lodz University of Technology, Poland

Electronic systems are usually described by assuming a model Hamiltonian, which only partially recovers electron correlation effects. To assure a quantitative description one faces a problem of recovering the missing part of the correlation. Over years different methods have been developed, most of them originating from the perturbation theory.

In my talk I will present another, fairly general, approach based on the adiabatic connection formalism. The idea itself is not novel although it has not been considered as a way of adding electron correlation for multireference models. Until recently it has not been realized that by combining the adiabatic connection (AC) with the extended random phase approximation one obtains a general tool capable of accounting for dynamical electron correlation for a broad class of multireference wavefunctions, applicable even to systems including strongly correlated electrons. It will be shown that the AC-based approximation yields excellent results when applied to multireference models, exceeding in accuracy second-order perturbation-theory-based methods.

O 62.2 Wed 11:00 HL 001

Density functional theory of electron transfer beyond the Born-Oppenheimer approximation: case study of LiF — •Chen Li[1], Ryan Requist[1], and Eberhard. K. U. Gross[1,2] — [1]Max Planck Institute of Microstructure Physics, Halle, Germany — [2]Fritz Haber Center for Molecular Dynamics, Institute of Chemistry, The Hebrew University of Jerusalem, Jerusalem, Israel

We demonstrate that beyond Born-Oppenheimer (BO) effects can be accurately and seamlessly incorporated within a density functional framework. In alkali halides like LiF, there is an abrupt change in the ground state electronic distribution due to an electron transfer at a critical bond length $R = R_c$. We find that nonadiabatic electron-nuclear coupling produces a sizable elongation of the critical R_c by 0.5 Bohr, an effect which is very accurately captured by a simple and rigorously-derived nuclear mass-dependent correction to the exchange-correlation potential in density functional theory. Since this nonadiabatic term depends on gradients of the nuclear wave function and conditional electronic density, $\nabla_R \chi(R)$ and $\nabla_R n(r, R)$, it couples the Kohn-Sham equations at neighboring R points. Motivated by an observed localization of nonadiabatic effects in nuclear configuration space, we propose an approximation that reduces the search for nonadiabatic density functionals to the search for a single function. This work is a step towards bringing density functional theory beyond the limitations of the BO approximation.

O 62.3 Wed 11:15 HL 001

Ground-State Quantum-Electrodynamical Density-Functional Theory — •Michael Ruggenthaler — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

In this talk I present a density-functional reformulation of correlated matter-photon problems subject to general external electromagnetic fields and charge currents [1]. I first show that for static minimally-coupled matter-photon systems an external electromagnetic field is equivalent to an external charge current. I employ this to show that scalar external potentials and transversal external charge currents are in a one-to-one correspondence to the expectation values of the charge density and the vector-potential of the correlated matter-photon ground state. This allows to establish a Maxwell-Kohn-Sham approach, where in conjunction with the usual single-particle Kohn-Sham equations a classical Maxwell equation has to be solved in order to capture the correlation induced by the transversal photon field. In the magnetic mean-field limit this reduces to a current-density-functional theory that does not suffer from non-uniqueness problems and if furthermore the magnetic field is zero recovers standard density-functional theory.

[1] "Ground-State Quantum-Electrodynamical Density-Functional Theory", M. Ruggenthaler, arXiv:1509.01417 (2017).

O 62.4 Wed 11:30 HL 001

Design of auxiliary systems for observables: the dynamic structure factor and the electron addition and removal spectra — Marco Vanzini, Martin Panholzer, Lucia Reining, and •Matteo Gatti — LSI, CNRS, Ecole Polytechnique, Palaiseau, France

Density functional theory tells us that the external potential, and therefore all observables, are functionals of the ground state density. The exact functionals, however, are not known, and one has to find approximations. To obtain the density, Kohn and Sham have proposed the idea to use an "auxiliary system". Much research effort goes into finding better and better Kohn Sham potentials for the density and the total ground state energy. In order to access also observables other than the density, we have proposed to generalize the Kohn-Sham idea of an auxiliary system [1], and to design a "connector" that allows us to profit from calculations done in a model system [2,3]. We have recently shown that this is a successful strategy for the dynamic structure factor [2] and for the one-body spectral function of simple metals, semiconductors and insulators [3]. [1] M. Gatti, V. Olevano, L. Reining, and I. V. Tokatly, Phys. Rev. Lett. 99, 057401 (2007) [2] M. Panholzer, M. Gatti, and L. Reining, arXiv:1708.02992 [3] M. Vanzini, L. Reining, and M. Gatti, arXiv:1708.02450

O 62.5 Wed 11:45 HL 001

Exact exchange energy of the ferromagnetic electron gas with dipolar interactions — •Camilla Pellegrini, Tristan Mueller, Kay Dewhurst, Sangeeta Sharma, and Eberhard K. U. Gross — Max-Planck-Institut fur Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

We propose a density functional treatment of the magnetic dipole-dipole interaction as a spin-spin correction to the Coulomb force in the Breit-Pauli Hamiltonian. Within this microscopic approach, the Hartree-like term for the dipolar coupling corresponds to the classical magnetostatic energy currently implemented in micromagnetic calculations. In addition, we have derived quantum corrections by evaluating analytically the exact exchange energy (Fock term) for the homogeneous electron gas, within the linear response to a noncollinear magnetic field. We expect our functional to open the path towards a full ab initio description of inhomogeneous magnetic structures at the nanoscale, with applications to domain-wall operated spintronic devices.

O 62.6 Wed 12:00 HL 001

Precise total-energy calculations at a significantly reduced cost — •Rudolf Zeller — Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

In density-functional calculations, the total-energy functional is stationary with respect to the density, the Kohn-Sham orbitals and the Kohn-Sham effective potential. This means that approximations for these quantities only lead to total-energy errors of second order provided that the total-energy functional is evaluated accurately without further uncontrolled approximations.

Unfortunately, usually the Kohn-Sham orbitals and thus the kinetic part of the total energy are evaluated by using a projection of the potential into a finite subspace of basis functions. This approximation damages the stationarity of the total energy as a functional of the potential.

A technique will be discussed which can relieve this deficiency so that a considerably smaller subspace of basis functions can be used for a precise evaluation of the kinetic part of the total energy. The advantage will be illustrated for the particular example of angular projection potentials as they are used in the full-potential Korringa-Kohn-Rostoker Green function method.

O 62.7 Wed 12:15 HL 001

Approach to Orbital-free DFT with Englert-Schwinger model — •Jouko Lehtomäki and Olga Lopez-Acevedo — COMP Centre of Excellence, Department of Applied Physics, Aalto University, Finland

We briefly present the Englert and Schwinger (ES) model in comparison with other approaches to orbital-free DFT. Essential failure of many kinetic energy density functionals is that they can not describe the most tightly bound core electrons in a satisfactory manner. Englert-Schwinger model allows treating these problematic electrons with more accurate single-particle wavefunctions while still obtaining the self-consistent orbital-free solution to the electronic problem.

Specifically, we detail how the ES model compares to the more known Thomas-Fermi-Dirac-Weizsäcker model self-consistently in atoms. We look at the total energy and few geometric properties. We show qualitative improvement in Pauli potential, which shows unphysical singularities near nucleus when the most tightly bound electrons are not treated correctly. We present how augmentation of the model with Kohn-Sham orbitals allows us to explore all-electron solution to the OFDFT problem and how this paves way for an orbital-free DFT method which does not need pseudopotentials.

O 62.8 Wed 12:30 HL 001

The Kerker Preconditioner for FLAPW Methods with Charge Density Mixing — •Miriam Hinzen, Edoardo Di Napoli, Daniel Wortmann, and Stefan Blügel — Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

In metallic systems of larger size the self-consistent field convergence of electronic structure calculations is often slowed down substantially due to charge sloshing: close to the Fermi level, little change in energy can cause large fluctuations in charge density. Mathematically speaking, the problem is ill-conditioned. For plane-wave methods the Kerker preconditioner effectively solved this problem, but for many other electronic structure methods, in particular all-electron methods as the FLAPW or KKR methods, a real-space formulation would be needed. We developed a formulation of the Kerker preconditioner for FLAPW methods with charge density mixing, implemented in `FLEUR` [1]. Numerical experiments show an enormous reduction of the number of iterations needed for convergence; even more importantly, the SCF convergence has become independent of the system size.
[1] www.flapw.de

O 62.9 Wed 12:45 HL 001

Effect of spin on the generalized Pauli constraints in Reduced Density Matrix Functional Theory — •Nicole Helbig[1], Iris Theophilou[2], and Nektarios N. Lathiotakis[3] — [1]Peter-Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich, D-52425 Jülich, Germany — [2]Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany — [3]Theoretical and Physical Chemistry Institute, National Hellenic Research Foundation, Vass. Constantinou 48, GR-11635 Athens, Greece

Reduced Density Matrix Functional Theory is a method that relies on the 1-1 correspondence between the many-body ground-state wave function and the first order reduced density matrix (1RDM) and uses the latter as its fundamental variable. Enforcing the generalized Pauli constraints during the energy minimization ensures that the 1RDM corresponds to a fermionic pure state. We demonstrate that these constraints are modified for open-shell systems if the spin degrees of freedom are taken into account. From the generalized Pauli constraints we also derive properties of the exact occupation numbers and natural orbitals which ensure that the 1RDM corresponds to an eigenstate of the total spin.