Symposium Density Functional Theory and Beyond for Real Materials (SYDF)

jointly organized by
the Surface Science Division (O),
the Thin Films Division (DS),
the Semiconductor Physics Division (HL),
the Magnetism Division (MA), and
the Dielectric Solids Division (DF)

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Overview of Invited Talks and Sessions

(lecture room H1)

Invited Talks

SYDF 1.1	Thu	14:45–15:15	H1	**Downfolded Self-Energy of Many-Electron Systems and the Hubbard U** — •Ferdi Aryasetiawan
SYDF 1.2	Thu	15:15–15:45	H1	**LDA+Gutzwiller method for correlated electron systems** — •Zhong Fang
SYDF 1.3	Thu	15:45–16:15	H1	**Localized and itinerant states in *d*/*f*-electron systems unified by *GW*@LDA+*U*** — •Hong Jiang
SYDF 1.4	Thu	16:30–17:00	H1	**Giant polaronic effects in solids and nanstructures** — •Andrea Marini
SYDF 1.5	Thu	17:00–17:30	H1	**Excitation energies with time-dependent density *matrix* functional theory** — •Evert Jan Baerends, Klaas J. H. Giesbertz, Oleg Gritsenko, Katarzyna Pernal
SYDF 1.6	Thu	17:30–18:00	H1	**Calculations of multipoles in magnetic metals and insulators** — •Lars Nordström

Sessions

SYDF 1.1–1.6	Thu	14:45–18:00	H1	**Density Functional Theory and Beyond for Real Materials**

SYDF 1: Density Functional Theory and Beyond for Real Materials

Time: Thursday 14:45–18:00 Location: H1

Invited Talk SYDF 1.1 Thu 14:45 H1

Downfolded Self-Energy of Many-Electron Systems and the Hubbard U — •Ferdi Aryasetiawan — Graduate School of Advanced Integration Science, Chiba University, 1-33 Yayoi-cho, Inageku, Chiba-shi, Chiba, 263-8522 Japan

Correlated materials are usually characterised by a partially filled narrow 3d or 4f bands crossing the Fermi level. These materials, such as the 3d perovskites, are hosts to many intriguing properties which cannot be satisfactorily explained within the local density approximation (LDA) or its extensions. The usual approach for treating these systems is to adopt a model Hamiltonian that contains input parameters, notably the Hubbard U, which is often treated as an adjustable parameter. It is therefore highly desirable to construct reliable model Hamiltonians, where the input parameters are computed from first-principles, thus avoiding adjustable parameters which may lead to misleading results.

Here we propose a constrained random-phase approximation (cRPA) scheme [1,2] that allows for a systematic downfolding of high energy screening channels resulting in a low-energy model with a screened Coulomb interaction or the Hubbard U [3]. The model Hamiltonian can then be solved by a number of approaches such as the LDA+U method, the dynamical mean-field theory (DMFT) scheme or the recently introduced LDA+Gutzwiller approach.

[1] Phys. Rev. B 70, 195104 (2004) [2] Phys. Rev. B 80, 155134 (2009) [3] Phys. Rev. Lett. 102, 176402 (2009)

Invited Talk SYDF 1.2 Thu 15:15 H1

LDA+Gutzwiller method for correlated electron systems — •Zhong Fang — Institute of Physics, Chinese Academy of Science

I will introduce our newly developed LDA+Gutzwiller method, in which the Gutzwiller variational approach is incorporated with the density functional theory (DFT) through the generalized Kohn-Sham formalism, such that orbital fluctuations can be treated from ab initio. We will show that its quality for ground state determination is as accurate as dynamic mean field theory (DMFT), and yet its computational cost is as cheap as LDA+U. Then we will concentrate on FeAs-based compounds, which are typical multi-orbital correlated system. Various corrected properties beyond LDA (or GGA), such as Fe-As distance, soft phonon, and Fermi surface shape, will be discussed based on the calculations by LDA+Gutzwiller. The inter-orbital Hund's coupling J rather than U plays crucial roles for the physical properties of FeAs-compounds.

References:

[1] X. Y. Deng, X. Dai, Z. Fang, EPL 83, 37008 (2008). [2] G. T. Wang, X. Dai, Z. Fang, PRL 101, 066403 (2008). [3] X. Y. Deng, L. Wang, X. Dai, Z. Fang, PRB 79, 075114 (2009). [4] G. T. Wang, X. Dai, Z. Fang, Cond-mat:/0903.1385 (2009).

Invited Talk SYDF 1.3 Thu 15:45 H1

Localized and itinerant states in d/f-electron systems unified by GW@LDA+U — •Hong Jiang — College of Chemistry, Peking University, Beijing 100871, China

Density-functional theory in the local-density or generalized gradient approximation (LDA/GGA) has proven to be inadequate for d and f-electron systems that are characterized by the simultaneous presence of itinerant and localized states and interactions between them. The simplest extension that can overcome the major failure of LDA is the introduction of a local Hubbard-like correction (LDA+U), however itinerant states are still treated at the LDA level. As a first step towards a unified treatment of localized and itinerant states, we combine many-body perturbation theory in the GW approximation with LDA+U (GW@LDA+U) to investigate the quasiparticle band structure of prototypical d/f-electron systems. We observe good agreement between the GW density of states and experimental photoemission spectra using Us determined by constrained DFT. All main features in the experimental band gaps of the lanthanide sesquioxide series (Ln_2O_3) are well reproduced by GW@LDA+U and can be attributed to the evolution of the occupied and unoccupied f-states as the number of f-electrons increases. Consistent with other GW approaches, the satellite structure in late transition metal oxides is still absent, and the binding energy of occupied d/f-states exhibits a tendency to be underestimated. The implications of our studies for strongly correlated electrons are discussed. *In collaboration with R. I. Gomez-Abal, P. Rinke and M. Scheffler at Fritz-Haber-Institut der MPG.

15 min. break

Invited Talk SYDF 1.4 Thu 16:30 H1

Giant polaronic effects in solids and nanstructures — •Andrea Marini — CNISM and department of Physics, University of Rome *Tor Vergata*

The conjugation of Ab-Initio methods with Many-Body techniques, constitutes a well-established approach to interpret the photoexcited properties of bulk materials, surfaces, nanostructures and organic/biomolecules. In the standard approach, however, atoms are assumed to be frozen in their crystallographic positions, thus neglecting the effect of lattice vibrations. As a consequence quasiparticles and excitons turn out to be insensitive to the temperature T and with an infinite lifetime, in stark contrast with the experimental evidence. In the frozen-atom approaches this temperature dependence is not described at all and, even in the $T \to 0$ limit, the calculated absorption spectra is commonly convoluted with some artificial, *ad-hoc* numerical broadening function chose to yield the best agreement with the experiment. In this talk I will show how to solve, in a fully Ab-Initio manner, the equation of motion of quasiparticles and excitons including the coupling with the lattice vibrations. The picture obtained within a frozen-atom approximation will turn out to be deeply modified by the electron-phonon coupling, either at zero and finite temperature. I will discuss several important consequences as the bright to dark (and vice versa) transitions of excitons in layered materials, or the beakdown of the quasiparticle picture in conjugated polymers.

Invited Talk SYDF 1.5 Thu 17:00 H1

Excitation energies with time-dependent density *matrix* functional theory — •Evert Jan Baerends, Klaas J. H. Giesbertz, Oleg Gritsenko, and Katarzyna Pernal — Vrije Universiteit, Amsterdam, The Netherlands

Time-dependent density functional theory in its current adiabatic implementations exhibits three striking failures: a) totally wrong behavior of the excited state surface along a bond-breaking coordinate [1]; b) lack of doubly excited configurations, affecting again excited state surfaces; c) much too low charge transfer excitation energies. We address these problems with time-dependent density matrix theory (TD-DMFT) [2-4].

For two-electron systems the exact exchange-correlation functional is known in DMFT, hence exact response equations can be formulated. This affords a study of the performance of TDDMFT in the TDDFT failure cases mentioned (which are all strikingly exhibited by prototype two-electron systems such as dissociating H_2 and HeH^+). At the same time, adiabatic approximations, which will eventually be necessary, can be tested without being obscured by approximations in the functional.

[1] K. J. H. Giesbertz, E. J. Baerends, Chem. Phys. Lett. 461 (2008) 338 [2] K. Pernal, O. V. Gritsenko, E. J. Baerends, Phys. Rev. A 75, 012506 (2007) [5] K. J. H. Giesbertz, E. J. Baerends, O. V. Gritsenko, Phys. Rev. Lett. 101 (2008) 033004 [6] K. J. H. Giesbertz, K. Pernal, O. V. Gritsenko, E. J. Baerends, J. Chem. Phys. 130 (2009) 114104

Invited Talk SYDF 1.6 Thu 17:30 H1

Calculations of multipoles in magnetic metals and insulators — •Lars Nordström — Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden

The concept of spherical tensors or multipoles of an open atomic shell is reviewed and discussed. Some of these multipoles play an important rule in e.g. x-ray circular dichroism measurements, where with the use of the famous sum rules by Carra et al. the spin and orbital magnetic moments can be deduced.

Here we will describe how such multipoles can be calculated in general in both the ground state as well as excited states in terms of density functional methods including a local correlation term, as in e.g. the so-called LDA+U or LDA+DMFT methods. It will be demonstrated how these multipoles can contribute significantly to the exchange and correlation energies of transition metal systems. Especially, we will discuss in some depth materials where these multipoles act as the

main order parameter, sometimes referred to as an "hidden order". Results for two cases will be presented the magnetic/superconducting iron-pnictide LaOFeAs and the heavy fermion compound URu_2Si_2.