What about U? - Corrective approaches to Density Functional Theory for strongly-correlated systems.

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Sponsors: CECAM and Psi-K

Organizers:
Matteo Cococcioni (University of Minnesota, USA) and
Silke Biermann (Ecole Polytechnique, Paris, France)

http://www.cecam.org/workshop-1-637.html

Workshop report

Correlated materials pose a challenge to solid state theory, due to the necessity of accurately taking into account the effects of electronic Coulomb interactions. While density functional theory in principle addresses this problem exactly by establishing the existence of an equivalent non-interacting system - at least for the purpose of calculating ground state properties - , two large classes of problems are left open:

1. The Hohenberg-Kohn theorem establishes the existence of a functional of the density whose minimization, in principle, should be sufficient to calculate the ground state properties of any system. In practice, however, the impossibility to construct this functional exactly calls for tractable and accurate approximations (beyond the local density approximation (LDA)) or alternative approaches that introduce explicit corrections beyond the density functional framework in the strict sense.

2. Addressing excited states properties requires, in general, to go beyond the one-particle picture and to include intrinsic many-body effects such as quasi-particle renormalisations or Mott localisation, satellite structures, or - for two particle-properties - excitonic effects or vertex corrections. This question is intimately related to the existence of ordering phenomena (magnetic, charge- or orbital-order), and to the temperature dependence of electronic properties beyond simple Fermi distribution effects.

Many corrective approaches to address the first and/or the second problem are implicitly or explicitly based on a correction to a pure LDA description, that involves an interaction term of Hubbard form. The local Coulomb interaction - the so-called "Hubbard U", which played the "title role" in our workshop - is viewed differently in different communities, from a mere fit parameter until a calculable quantity in random-phase approximation - based or linear-response theories. The way it is then treated is as diverse as its definition: explicitly within a static or dynamic mean field approximation within LDA+U and LDA+DMFT respectively or implicitly in density matrix, SIC or GW based schemes, which try to avoid explicit reference to U.

The workshop "What about U – Corrective approaches to density functional theory for correlated systems" aimed at bringing together researchers who work on the definition, development and implementation of various corrective schemes and algorithms designed to improve

the accuracy and the predictive power of (approximate) Density Functional Theory in describing the physical properties of strongly-correlated materials. The objective was to present the latest advancements for each of them, to discuss the approximations behind their formulation and their specific prerogatives, to compare their capabilities and performances and to define a common theoretical background in which these corrective schemes can be formulated. The development of a common theoretical language is a prerogative for discussions between the different communities, necessary to stimulate new efforts to find solutions to common problems, to use the advancements in one method to improve the others, to integrate different numerical approaches into new, more flexible and efficient computational tools that can be used to perform predictive calculations on the widest possible range of different materials.

The workshop gathered 61 participants from 20 different countries for three and a half days at CECAM headquarters in Lausanne. It particularly encouraged the participation of students and junior researchers, providing them with first-hand experience of the theoretical foundations of DFT, and of a broad spectrum of corrective approaches to improve the description of electronic correlation. The intent was to stimulate young researchers to contribute actively to the field and to become involved in the theoretical definition and practical implementation of more general, versatile and efficient computational tools for correlated systems.

The objectives of the workshop were greatly achieved. The many discussions after the talks and in round table sessions allowed to explore the theory of various numerical approaches in depth and also to discuss various fundamental topics and questions. Presenters and participants made a very serious effort to bridge their methods with others, to find a common theoretical background to frame them in, to adopt similar solutions to common problems.

We are particularly glad about the way this event worked out, with the speakers making every effort to adapt to a quite specific but diverse audience, highly motivated chairpersons and a lively participation from the audience, a sparkling atmosphere for the discussions. Many of the participants said it was one of the most inspiring, formative and interesting workshops they have ever attended.

Programme of the workshop

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Day 1 June 18th 2012

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SESSION I: About density matrices, density functionals and Gutzwiller

- 8:45 to 9.00 - Registration

- 9:00 to 9:15 - Welcome

- 9:15 to 9:40 - **E. K. U. Gross** "Spectral information from density-matrix functional theory: Towards an ab-initio theory of strongly correlated systems"

- 09:40 to 10:00 - Discussion

- 10:00 to 10:25 - **Klaus W. Capelle** "What can LDA do for U? What can U do for LDA?"

- 10:25 to 10:45 - Discussion

- 10:45 to 11:15 - Coffee Break

- 11:15 to 11:40 - **Giovanni Borghi** "Adding local correlations to the Kohn-Sham wavefunction: LDA+U improved by means of Gutzwiller variational method"

- 11:40 to 12:00 - Discussion

- 12:00 to 12:15 - **Bo Hellsing** "A Gutzwiller study of correlated multi-orbital systems"

- 12:15 to 12:25 - Discussion

- 12:25 to 14:00 - Lunch Break

SESSION II: Dynamical Mean Field Theory – Part I

- 14:00 to 14:25 - **Antoine Georges** "Not only U, also J: Hund's correlated materials"

- 14:25 to 14:45 - Discussion

- 14:45 to 15:10 - **Frank Lechermann** "Two Scenarios of First-Order Phase Transitions in Strongly Correlated Materials: V_2O_3 and $Sr3Ru_2O_7$"

- 15:10 to 15:30 - Discussion

- 15:30 to 16:00 - Coffee Break Poster flash: 2 slides per poster

- 16:00 to 18:30 - Poster Session

- 19:00 to 21:00 - Dinner

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Day 2 June 19th 2012

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SESSION III: Dynamical Mean Field Theory – Part II

- 09:00 to 09:25 - **Olle Eriksson** "Title: Dynamical mean-field theory and full-potential linear muffin-tin orbitals - methodology and examples"

- 09:25 to 09:45 - Discussion

- 09:45 to 10:10 - **Karsten Held** "Dynamical vertex approximation - electronic correlations beyond DMFT"

- 10:10 to 10:30 - Discussion

- 10:30 to 11:00 - Coffee Break

- 11:00 to 11:25 - **Ferdi Aryasetiawan** "Effective Coulomb Interaction of Many-Electron Systems"

- 11:25 to 11:45 - Discussion

- 11:45 to 12:10 - **Mark van Schilfgaarde** "Systematic Errors in U from the Quasiparticle Self-consistent GW Approximation"

- 12:10 to 12:30 - Discussion

- 12:30 to 14:00 - Lunch Break

SESSION IV: What about U? – Correlation effects in various systems

- 14:00 to 14:25 - **Alexander Lichtenstein** "What about U in Graphene"

- 14:25 to 14:45 - Discussion

- 14:45 to 15:00 - **Philipp Hansmann** "Correlation effects on group IV adatom systems on the Si(111)-surface: Mott or not?"

- 15:00 to 15:10 - Discussion

- 15:10 to 15:40 - Coffee Break

- 15:40 to 16:05 - **Takashi Miyake** "Dynamic screening and nonlocal self-energy effects in 3d compounds"

- 16:05 to 16:25 - Discussion

- 16:25 to 18:00 - **Round table discussion: What about U?**

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Day 3 June 20th 2012

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SESSION V: Pushing functionals to their limits: from DMRG to SIC

- 09:00 to 09:25 - **Kieron Burke** "Kondo plateau given exactly by DFT and how to use DMRG to tackle strong correlation in DFT"

- 09:25 to 09:45 - Discussion

- 09:45 to 10:10 - **Stefano de Gironcoli** "Correlation energy from Adiabatic Coupling Fluctuation-Dissipation"

- 10:10 to 10:30 - Discussion

- 10:30 to 11:00 - Coffee Break

- 11:00 to 11:25 - **Martin Lueders** "Self-Interaction Correction in DFT: where do we stand?"

- 11:25 to 11:45 - Discussion

- 11:45 to 12:10 - **Ismaila Dabo** "Bridging self-interaction corrections with Hubbard corrections"

- 12:10 to 12:30 - Discussion

- 12:30 to 14:00 - Lunch Break

SESSION VI: Orbital-dependent functionals

- 14:00 to 14:25 - **Sohrab Ismail-Beigi** "Luttinger-Ward approaches for going beyond DFT: progress and challenges within the GW-RPA approximation exact rewriting, systematic approximations, and fundamental challenges"

- 14:25 to 14:45 - Discussion

- 14:45 to 15:10 - **Andrea Ferretti** "Bridging density-functional and many-body perturbation theory: orbital-density dependence in electronic-structure functionals"

- 15:10 to 15:30 - Discussion

- 15:30 to 16:00 - Coffee Break

- 16:00 to 16:25 - **Leeor Kronik** "What about γ? Spectroscopy with Optimally Tuned Range-Separated Hybrid Functionals"

- 16:25 to 16:45 - Discussion

- 16:45 to 17:10 - **Christoph Friedrich** "All-electron treatment of orbital-dependent functionals and the GW and GT self-energy"

- 17:10 to 17:30 - Discussion

Day 4 June 21th 2012

SESSION VII: Moving the atoms ...

- 09:00 to 09:25 - **Andrea Floris** "DFT+U-based Density Functional Perturbation Theory: Applications to MnO, NiO"

- 09:25 to 09:45 - Discussion

- 09:45 to 10:10 - **Ivan Leonov** "Electronic correlations and lattice dynamical properties of elemental iron near the *alpha-gamma* phase transition"

- 10:10 to 10:30 - Discussion

- 10:30 to 11:00 - Coffee Break

- 11:00 to 12:30 - **Round table discussion: Open Challenges in the Field**

Spectral information from density-matrix functional theory: Towards an ab-initio theory of strongly correlated systems

E.K.U. Gross
Max Planck Institute of Microstructure Physics, Halle, Germany

Abstract

We present a novel method for calculating the photo-electron spectrum of periodic solids within reduced-density-matrix-functional theory. Results are compared with DMFT and GW calculations, finding excellent agreement in all cases studied. Furthermore, the physics behind the pressure-induced insulator-metal transition in MnO will be described in detail. The driving mechanism of this transition is identified as increased crystal-field splitting with pressure, resulting in a charge redistribution between the Mn eg and t2g symmetry projected states.

What can LDA do for U? What can U do for LDA?

Klaus W. Capelle
Federal University of ABC, Santo André, Brazil

Abstract

The density-functional approach and the model-Hamiltonian approach each constitute one of the pillars of modern many-body theory. In the past, crosslinks between both pillars have been forged in various different ways. In this talk I briefly review some of these connections, highlighting the many possible benefits that arise from using insights from DFT to study model Hamiltonians, and ideas arising from model Hamiltonians to improve functionals of DFT. Next, I address one particular connection in more detail: the use of the Hubbard model as a theoretical laboratory for investigating concepts and computational tools of DFT for a well-controlled model system.

Adding local correlations to the Kohn-Sham wavefunction: LDA+U improved by means of Gutzwiller variational method.

Giovanni Borghi
Swiss Federal Institute of Technology Lausanne, Switzerland

Abstract

By means of the constrained-search formulation of Density Functional Theory [1],the Kohn-Sham auxiliary system of non-interacting electrons can be generalized to a system of particles that are coupled through local Hubbard-type interactions, and whose ground-state wavefunction is computed within Gutzwiller Variational Method. We show how the resulting Gutzwiller Density Functional is a natural extension of DFT+U functionals, with an additional set of many body parameters that are optimized together with the density. Among these, a band-mass renormalization parameter à la Landau accounts for

the reduced mobility of correlated particles and their suppressed kinetic energy. Within Gutzwiller formalism, the Hubbard-U term can be generalized with no extra effort to more complex local Hamiltonians, including couplings with many-body local operator such as total spin and total angular momentum. The increased flexibility of the Gutzwiller Density Functional can lead to a better understanding of the origin of magnetism in transition metals. A comparison of total energies of the paramagnetic and ferromagnetic phases of iron shows the important role of the double-exchange mechanism as an explanation of magnetic ordering in this system [2].

References

[1] M. Levy, Proc. Natl. Acad. Sci. USA 76, 6062 (1979); E. Lieb, Int. J. Quantum Chem. 24, 243 (1983).
[2] G. Borghi, M. Fabrizio, E. Tosatti, (in preparation).

A Gutzwiller study of correlated multi-orbital systems
Bo Hellsing
University of Gothenburg, Sweden

Coauthor(s) : Nicola Lanata[1], Hugo U. R. Strand[1], Xi Dai[2]
[2] Chinese Academy of Sciences, Beijing, China

Abstract

We present a numerical method to minimize the general multi-orbital Gutzwiller variational wave functions [1]. The method can treat any type of local interaction, in particular spin-flip and pair-hopping interactions. To construct the variational wave function, we apply the φ-matrix formalism [2] and a general approach to reduce the variational space by imposing lattice point group symmetries. Results for the multi-orbital Hubbard model of d-electron systems with rotational invariant Kanamori interaction will be demonstrated. The variational wave function is restricted to cubic symmetry. Properties of the model is studied as a function of N (total number of electrons), ΔE (e.g. t2g crystal field splitting), U (interaction strength) and J (spin-exchange coupling). Our numerical results for the quasi-particle weight, Z vs. U and the phase diagram, J/U vs. Uc, indicate dramatic changes when going off half-filling (N=5).

References

[1] N. Lanata, H.U.R. Strand, X. Dai, B. Hellsing, Phys Rev B 85, 035133 (2012).
[2] N. Lanata, P. Barone, M. Fabrizio, Phys. Rev. B 78, 155127 (2008).

Not only U, also J: Hund's correlated materials
Antoine Georges

Abstract

N/A

Two Scenarios of First-Order Phase Transitions in Strongly Correlated Materials: V2O3 and Sr3Ru2O7

Frank Lechermann
University of Hamburg, Germany

Abstract

The combination of first-principles band-structure techniques with explicit many-body approaches has grown beyond its infancy and is nowadays a powerful tool for the investigation of strongly correlated materials. Novel developments like the inclusion of rotational invariant interactions, the introduction of efficient interfaces between the effective single-particle and the many-body scheme or the establishment of charge-self consistency within the combined approach have shifted the methodology to a level of sophistication that renders the claim for predictive power possible.
In the light of these achievements, we here discuss the phenomenology and theoretical description of first-order phase transitions in two prominent strongly correlated materials systems. The well-known V2O3 compound exhibits metal-insulator transitions with negative pressure and elevated temperature [1]. Employing state-of-the-art LDA+DMFT calculations within the charge-self consistent framework allows for a description of the associated phase diagram in line with experiment and reveals a deeper insight in the physics close to the transition regime [2]. On the contrary, the low-temperature metamagnetic transitions in Sr3Ru2O7 are driven by applied magnetic field H with significant dependence on the angle between H and the crystallographic c-axis [3]. An intriguing interplay between spin-orbit coupling and electronic correlations gives rise to complex multi-orbital physics in this Fermi liquid and leads to subtle modifications of the low-energy quasiparticle states with applied field [4].

References

[1] D.B. McWhan, T.M. Rice, J.B. Remeika, Phys. Rev. Lett. 23, 1384 (1969).
[2] D. Grieger, C. Piefke, O.E. Peil, F. Lechermann, arXiv:1203.5354 (submitted in 2012).
[3] A.P. Mackenzie, J.A.N. Bruin, R.A. Borzi, A.W. Rost, S.A. Grigera, arXiv:1201.6639 (submitted in 2012).
[4] M. Behrmann, C. Piefke, F. Lechermann, arXiv:1203.3148 (submitted in 2012).

Dynamical mean-field theory and full-potential linear muffin-tin orbitals - methodology and examples

Olle Eriksson
University of Uppsala, Sweden

Abstract

In this talk I will give some of the details of the implementation of dynamical mean-field theory [1] in a full-potential linear muffin-tin orbital method [2-5]. Various choices of correlated orbitals and impurity solvers, like Hubbard-I, exact diagonalization, and SPTF, will be discussed when making comparisons to the electronic structure of a wide selection of materials. This selection involves bulk and surfaces of transition metals, transition metal oxides, hard permanent magnets like SmCo5 as well as magnetic semiconductors.

References

[1] A. Georges, G. Kotliar, W. Krauth,M. Rozenberg, J.,Review of Mod. Phys. 68, 13 (1996).
[2] A. Grechnev, I.D. Marco, M.I. Katsnelson, A.I. Lichten- stein, J. Wills, O. Eriksson, Phys. Rev. B 76,
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Dynamical vertex approximation - electronic correlations beyond DMFT

Karsten Held
Vienna University of Technology, Austria

Abstract

Dynamical mean field theory includes a major part of the electronic correlations: the local ones for two f- or d-electrons on the same lattice site. Often however interesting correlation physics emerges from non local correlations such as e.g. (quantum) criticality and d-wave superconductivity. I will discuss our recent efforts to include these non-local correlations diagrammatically in the dynamical vertex approximation [1]. This allows among others to calculate critical exponents [2] and to open a pseudogap in the spectrum.

The dynamical vertex approximation is also appropriate for going beyond GW or GW+DMFT [3], on the basis of Hedin's equations.

References

[1] A. Toschi, A.A. Katanin, K. Held, Phys. Rev. B 75, 045118 (2007).
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[3] A. Toschi, G. Rohringer, A.A. Katanin, K. Held, Annalen der Physik 523, 698 (2011).

Effective Coulomb Interaction of Many-Electron Systems
Ferdi Aryasetiawan
Lund University, Sweden

Abstract

One effective approach of studying the electronic structure of correlated materials is to focus on the partially filled narrow 3d or 4f band where most of the electron correlations responsible for interesting physical phenomena are supposed to originate. By mapping the full Hamiltonian to an effective Hamiltonian corresponding to the correlated subspace, the electron-electron interaction experiences a renormalisation. Knowledge on how to calculate this renormalised interaction, or widely known as the Hubbard U, is of utmost importance since nearly all physical phenomena are determined by this interaction. There are several techniques to calculate the Hubbard U from first principle and here we will present a method based on linear response theory within the random-phase approximation. A formal derivation of the method was proposed recently. This method has a crucial merit in that it gives a complete description of the Hubbard U in one calculation. For example, the full U matrix including off-site elements and its energy dependence of U can be obtained. The possibility of having an energy-dependent U is particularly interesting with recent important progress in solving the impurity problem with a frequency-dependent interaction in the so-called LDA+DMFT+U(ω). As an illustration we discuss a recent application to the pnictide BaFe2As2.
Although the LDA+DMFT+U(ω) method represents significant progress in electronic structure calculations, the method neglects the nonlocality or k-dependence of the self-energy, which may significantly alter the quasiparticle band structure. To take full account of the k-dependent self-energy we discuss a method which employs the GW approximation for the nonlocal part of the self-energy while keeping the local self-energy given by the DMFT. Apart from incorporating a k-dependent self-energy, this method, GW+DMFT, also cures the double-counting problem associated with methods based on the LDA such as the LDA+U and the LDA+DMFT scheme.

Systematic Errors in U from the Quasiparticle Self-consistent GW Approximation

Mark van Schilfgaarde
King's College London, United Kingdom

Abstract

As is well known, parameters U and J entering into the LDA+U and LDA+DMFT Hamiltonians should be the screened coulomb interaction W, subject to a constraint to avoid double-counting of self-screening. In any case U and J are constructed in practice in the time-dependent Hartree approximation. Moreover, there are manifold ambiguities inherent in the manner in which U and J are made. First, they are usually taken to be scalars, which indicates that they amount to some (average) matrix elements of W(r; r0; ω) in a basis. Usually U is taken in the static limit, U≈U (ω=0). Finally, the framework, LDA or DFT, which +U or +DMFT improve upon, cannot be regarded as merely a lower order approximation, resulting in further ambiguities. The auxiliary one-electron hamiltonian that is a byproduct of DFT,generates fictitious eigenvalues that are nevertheless assumed to be quasiparticle spectra that LDA+U or LDA+DMFT corrects. In this talk we present the quasiparticle self-consistent GW approximation (QSGW), and show how it sheds light on many of the approximations and assumptions inherent in LDA+DMFT. GW is a low-order perturbation theory, which makes the time-dependent Hartree approximation for W. Like LDA+U and LDA+DMFT, GW is usually built around the LDA (GLDAWLDA) and has its own ambiguities. We first show QSGW is an optical choice for GW, and circumvents many of the ambiguities of GLDAWLDA.QSGW describes optical properties in a wide range of materials rather well.Self-consistency dramatically improves agreement with experiment, and is sometimes essential. It handles both itinerant and correlated electrons on an equal footing, without any ambiguity about how a localized state is defined, or how double-counting terms should be subtracted.
Discrepancies with experiment in the quasiparticle spectra are small and systematic, and can be explained in terms of the approximations made. Thus QSGW provides the optimal framework for determining the limits to the RPA. We argue that W is systematically overestimated in the RPA, by a universal factor of about 5/4. Also we can estimate the error in the static approximation for W. Finally, we use a few examples to argue that because contributions missing from a typical LDA+U or LDA+DMFT calculation make significant contributions to the QP spectra. Thus LDA+DMFT calculations should not necessarily compare to experimental spectra when assessing the validity of their results.

What about U in Graphene

Alexander Lichtenstein
University of Hamburg, Hamburg, Germany

Abstract

Graphene has been attracting an increasing interest due to its remarkable physical properties ranging from the Dirac electron spectrum to ballistic transport under ambient

conditions. The latter makes graphene a promising material for future electronics and the recently demonstrated possibility of chemical doping without significant change in mobility has improved graphene's prospects further. We address the question effective Coulomb interaction in graphene. Results of C-RPA calculation [1] show that the U-physics is very non-local in graphene. Furthermore, recent experimental results on Co impurity on graphene will be discussed from a theoretical point of view

References

[1] T.O. Wehling, E. Sasioglu, C. Friedrich, A.I. Lichtenstein, M.I. Katsnelson, S. Blügel, Phys. Rev. Lett. 106, 236805 (2011).

Correlation effects on group IV adatom systems on the Si(111)-surface: Mott or not?
Philipp Hansmann
Ecole Polytechnique, Paris, France

Abstract

For quite some time now the so called alpha-phases of group IV adatom systems on silicon and germanium 111 surfaces have been under experimental and theorectial investigation. Specifically the role played by electron-electron correlations and their complex interplay with lattice degrees of freedom has been under controversial debate. It will be discussed how we can tackle the problem by means of state-of-the-art ab initio + many body approaches like e.g. density functional + dynamical mean field theory (LDA+DMFT). It will be shown how we can reach a truely ab initio treatment by calculating also the interaction parameters for the effective low energy models with the recently developed constrained random phase approximation (cRPA). Knowing the correct interaction parameters for the respective models will give extremely valuable information even bofre solving the complex many body problem and we can estimate for example if interaction effects can be approximated by a purely local term or if intersite interactions will contribute significantly. Recent results for said silicon adatom systems will be presented and possible consequenses for the subsequent theoretical treatment will be discussed.

Dynamic screening and nonlocal self-energy effects in 3d compounds
Takashi Miyake
National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan

Abstract

The constrained RPA (cRPA) is getting to be one of the standard methods to construct low-energy effective models from first principles. Although the effective electron interaction in cRPA is dynamic, the frequency dependence is normally neglected when the model is solved. In addition, the self-energy is approximated to be local if we take DMFT as the low-energy solver. In the present work, we study many-body effects in SrVO3 in the GW approximation. The self-energy, quasiparticle bandstructure and spectral function are presented and discussed in detail. The GW self-energy reduces the t2g bandwidth by 20 % compared to LDA. When we neglect the non-locality of the self-energy (in the t2g model), the band becomes substantially narrower than that in GW by 40 %. The static approximation to the effective interaction, on the other hand, widens the band, and partly cancels too strong band renormalization in the local approximation. The work is done in collaboration with Cyril Martins and Ferdi Aryasetiawan.

Kondo plateau given exactly by DFT and how to use DMRG to tackle strong correlation in DFT
Kieron Burke
University of California at Irvine, USA

Abstract

I will discuss recent work in transport through molecular junctions, which shows that the Kondo plateau in the transmission through an Anderson junction at weak bias and zero temperature is recovered exactly within ground-state Kohn-Sham DFT [1]. I will also discuss which aspects of approximate functionals are needed to recover this paradigm of strong correlation [2].Then I will discuss a major collaboration with Steve White, inventor of density-matrix renormalization group (DMRG), designed to make progress on all the issues raised in this workshop [3,4]. In the second half of the talk, I will discuss how partition density functional theory might play a vital role [5].

References

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Correlation energy from Adiabatic Coupling Fluctuation-Dissipation.

Stefano de Gironcoli
International School for Advanced Studies (SISSA) and CNR-DEMOCRITOS IOM,
Trieste, Italy

Abstract

Twenty years of exhaustive research and testing on dozens of GGA functionals have left
two physical issues qualitatively not addressed: i) non local correlations associated with
weak but pervasive dispersion interactions responsible for surface physisorption and
binding inorganic systems ii) local on-site correlations/self-interaction effects responsible
for Mott-Hubbard physics but also important for molecular dissociation into integer-
charged fragments. The formally exact Adiabatic Connection Fluctuation Dissipation
(ACFD) formalism can in principle address both issues. I'll report our progress in the
calculation of correlation energy and potential in ACFD as well as the development of
simplified fully non-local functionals for van der Waals systems.

Self-Interaction Correction in DFT: where do we stand?

Martin Lueders
Daresbury Laboratory, United Kingdom

Abstract

The Self-Interaction Correction (SIC) was introduced to DFT about 30 years ago by
Perdew and Zunger (PZ). In this talk, I will discuss where PZ-SIC stands with respect to
other methods, but also in its own development.
I will address the questions how the PZ-SIC fits into the DFT schemes in terms of the
Hohenberg-Kohn theorem and the Kohn-Sham system, but also how it relates to exact
exchange, Hartree-Fock and also LDA+U. Some aspects of these methods, such as the
"correlated orbitals" or the existence of multiple minima and their physical meaning, will
be compared and discussed in more detail. Furthermore, the combination of SIC and the
coherent potential approximation will be discussed, and put in relation to the
LDA+DMFT approach. Here, in particular, the question of quantum fluctuations versus
thermal fluctuations will be addressed. Examples for the discussion will be mainly based
on transition metal oxides.

Bridging self-interaction corrections with Hubbard corrections

Ismaila Dabo
University of Paris-Est, France

Abstract

Density-functional theory (DFT) has been successfully used in diverse areas of materials science. In spite of notable achievements, the lack of piecewise linearity of conventional DFT approximations (i.e., the unphysical dependence of the ground-state energy as a function of the number of electrons) reverberates negatively on the description of systems involving localized, strongly correlated electronic states [1]. This deficiency is particularly marked in predicting the energetic and structural properties of materials involving heavy-metal atoms and represents a severe limitation in studying strongly correlated systems. Among proposed corrections, DFT augmented with Hubbard U contributions (DFT+U) represents a very beneficial compromise between cost and accuracy, and has thus been used extensively. Following a route parallel to DFT+U, the lack of piecewise linearity can be regarded as the consequence of electron self-interaction (i.e., the unphysical interaction of an electron with itself through its contribution to the effective DFT potential), thereby motivating the use of self-interaction corrections (SIC-DFT) [2]. Although it is clear that DFT+U and SIC-DFT methods share a common objective, the correspondence between the two approaches remains largely unexplored, representing a gap in the current understanding of post-DFT corrections. In this presentation, I will identify a direct route to connect linear-response DFT+U [3] and Koopmans-compliant SIC-DFT [4], and explain the implications of this correspondence in restoring the piecewise linearity of approximate DFT functionals with improved accuracy in an effort to extend their predictive scope.

References

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Luttinger-Ward approaches for going beyond DFT: progress and challenges within the GW-RPA approximation exact rewriting, systematic approximations, and fundamental challenges

Sohrab Ismail-Beigi
Yale University, USA

Abstract

As is well known, Density Functional Theory (DFT) is a variational theory for the ground-state with no guarantees to deliver on excited state properties such as quasiparticle band energies or wave function; all that is guaranteed is that the effective electronic states (Kohn-Sham states) generate the right electron density at the variational minimum. Luttinger-Ward approaches to electronic structure are attractive as beyond-DFT schemes since, in principle, they offer a variational functional of the one-particle Green's function that yield both the correct total energy and all one-particle properties at

the variational extremum. However, these methods face a number of additional challenges beyond the familiar problem of choosing an exchange-correlation energy functional: (a) their much greater computational cost compared to DFT, (b) the theoretical problem of choosing an appropriate set of trial Green's functions, and (c) locating the variational extremum. We will be discussing our recent work in this general area focusing primarily on the standard random phase approximation (RPA) correlation functional which yields the GW self-energy for the Green's function. We will present results on computationally more effective approaches for the RPA correlation energy; how various types of approximations to the correlation energy create a ladder of approximate self-energy operators and what type of physics is included in these types of approximations (e.g. exchange, screening, Hubbard U, etc.); and unavoidable difficulties in extremizing the functional. Time permitting, we will discuss some ideas for making practical progress with the extremization and their relation to the successful Quasiparticle Self-consistent GW (QSGW) scheme of Ref. [1].

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Bridging density-functional and many-body perturbation theory: orbital-density dependence in electronic-structure functionals
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Abstract

Energy functionals which depend explicitly on the orbital densities (ODD), instead of the total charge density, appear when applying self-interaction corrections to density-functional theory. In these cases (e.g. the Perdew-Zunger [1] and the non-Koopmans [2] approaches) the total energy loses invariance under unitary rotations of the orbitals, and the minimization of the functionals leads to orbital-dependent Hamiltonians. We show that it is possible to identify the orbital-dependency of densities and potentials with an effective and discretized frequency-dependency, in close analogy to the quasi-particle approximation of frequency-dependent self-energies and naturally oriented to interpret electronic spectroscopies [3].Some of the existing ODD functionals are analyzed from this new perspective. Numerical results for the electronic structure of gas-phase molecules (within the Koopmans-corrected class of functionals) are computed and found in excellent agreement with photoemission (UPS) data.

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What about γ? Spectroscopy with Optimally Tuned Range-Separated Hybrid Functionals

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Abstract

Excitation gaps are of considerable significance in electronic structure theory. Two different gaps are of particular interest. The fundamental gap is defined by charged excitations, as the difference between the first ionization potential and the first electron affinity. The optical gap is defined by a neutral excitation, as the difference between the energies of the lowest dipole-allowed excited state and the ground state. Within many-body perturbation theory, the fundamental gap is the difference between the corresponding lowest quasi-hole and quasi-electron excitation energies and the optical gap is addressed by including the interaction between a quasi-electron and a quasi-hole. A long-standing challenge has been the attainment of a similar description within density functional theory (DFT), with much debate on whether this is an achievable goal even in principle. Recently, we have constructed and applied a new approach to this problem [1]. Anchored in the rigorous theoretical framework of the generalized Kohn-Sham equation, our method is based on a range-split hybrid functional that uses exact long-range exchange. Its main novel feature is that the range-splitting parameter, γ, is not a universal constant, but rather is determined from first principles, per-system, based on satisfaction of the ionization potential theorem. For finite-sized objects, this DFT approach mimics successfully, to the best of our knowledge for the first time, the quasi-particle picture of many-body theory. Specifically, it allows for the extraction of both the fundamental and the optical gap from one underlying functional, based on the HOMO-LUMO gap of a ground-state DFT calculation and the lowest excitation energy of linear-response time-dependent DFT calculation, respectively. In particular, it produces the correct optical gap for the difficult case of charge-transfer and charge-transfer-like scenarios, where conventional functionals are known to fail. Here, I overview the formal and practical challenges associated with gap calculations, explain our new approach and how it overcomes previous difficulties, and survey its application to a variety of systems. Research performed with R. Baer, T. Stein, H. Eisenberg (Hebrew U, Jerusalem); S. Refaely-Abramson, N. Kuritz (Weizmann Inst.)

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All-electron treatment of orbital-dependent functionals and the GW and GT self-energy

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Abstract

We present recent advances in the numerical implementation of the optimized effective potential (OEP) method as well as the GW and GT approximation for the electronic self-energy within the all-electron full-potential augmented-plane-wave (FLAPW) approach. The OEP method is used to construct local potentials from non-local, orbital-dependent exchange-correlation functionals, e.g., exact exchange (EXX). It involves two response functions, which have to be converged to very high precision to obtain smooth and stable local potentials, leading to very costly calculations [1]. We demonstrate that a combination of the usual summation over states with a radial Sternheimer approach improves the quality of the response functions such that accurate results are obtained with a considerably reduced computational cost [2]. The method can be understood as an incomplete-basis-set correction (IBC) for response quantities. We show results for a variety of materials including transition-metal monoxides. The IBC is particularly interesting for the GW approximation where the slow convergence with respect to unoccupied states is a long-standing problem; zinc oxide is a prominent example [3]. While the GW approximation is known to work very well for weakly to moderately correlated materials, it lacks electron-magnon scattering processes that may occur in magnetic materials with localized d and f orbitals. Recent ARPES experiments [4,5] indicate that these processes lead to a pronounced renormalization of the electron band dispersion, creating a kink near the Fermi energy. In order to describe this renormalization from first principles, we use the T-matrix formalism, which describes the correlated motion of an electron-hole pair of opposite spins in terms of ladder diagrams. The multiple scattering gives rise to collective spin excitations [6].Through emitting and absorption of these magnons, the energy of an electron propagating through a spin-polarized system gets renormalized. We describe this renormalization by a GT approximation for the self-energy [7]. We present first results for elementary ferromagnets. All implementations are realized in the FLEUR and SPEX codes, which are part of the Jülich code family [8].

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Title: DFT+U-based Density Functional Perturbation Theory: Applications to MnO, NiO

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Abstract

In this contribution, I will introduce a formalism that extends the density functional perturbation theory (DFPT) to allow self-consistent linear-response calculations from a DFT + U ground state. Using this scheme, the full phonon dispersion of strongly correlated materials, whose ground state can be captured with DFT+U, can be accessed with accuracy and numerical efficiency. The tool is applied to MnO and NiO in their antiferromagnetic (AFII) ground state. The results significantly improve the agreement with the experimental dispersions if compared with standard GGA calculations. Moreover, they confirm the highly non-cubic behavior of these systems and show a strong interplay between phonon spectrum features and occupation of specific d states.

Electronic correlations and lattice dynamical properties of elemental iron near the $alpha$-$gamma$ phase transition
Ivan Leonov
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Abstract

We present results of a theoretical investigation of the electronic and lattice dynamical properties of elemental iron at finite temperatures obtained within dynamical mean-field theory implemented with the frozen-phonon method [1]. This approach allows us to compute correlation induced lattice transformations and their temperature evolution. We find that electronic correlations are important to explain the lattice stability of iron at the bcc-fcc phase transition. We notice a weak anomaly in the transverse T1 acoustic mode in the Γ-N direction of the bcc phase. This behavior can be ascribed to a dynamical precursor effect of the bcc-to-fcc phase transition and is found to occur above the Curie temperature. Upon further heating, the bcc phase becomes dynamically unstable due to the T1 mode near the N point. By contrast, the fcc lattice is found to be dynamically stable in a broad temperature range, including temperatures above and below the bcc-fcc phase transition temperature. Our results for the structural phase stability and lattice dynamical properties of iron are in good agreement with experiment.

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PSI-K WORKSHOP FINANCE REPORT

Title of the Workshop	What about U? - Corrective approaches to DFT for strongly-correlated systems.+ **Scientific**
Workshop Organisers	Matteo Cococcioni (University of Minnesota, USA) and Silke Biermann
Location of the Workshop	EPFL, Lausanne (CECAM HQ)
Dates of the Workshop	June 18-21 2012
Total Grant from Psi-k	8000

EXPENDITURE
(Please complete the tables using numbers ONLY in the amounts fields - the spreadsheet will calculate the totals for you)

WORKSHOP DELEGATES / SPEAKERS

Name and Country of Claimant	Travel		Accommodation		Total (€'s)	
Silke Biermann (France)			€	441.67	€	441.67
Matteo Cococcioni (USA)	€	500.00	€	441.67	€	941.67
Ferdi Aryasetiawan (Sweden)			€	441.67	€	441.67
Kieron Burke (USA)	€	500.00	€	220.83	€	720.83
Klaus Capelle (Brazil)	€	500.00	€	441.67	€	941.67
Ismaila Dabo (France)			€	445.00	€	445.00
Stefano de Gironcoli (Italy)			€	441.67	€	441.67
Olle Eriksson (Sweden)			€	441.67	€	441.67
Andrea Ferretti (Italy)			€	445.00	€	445.00
Andrea Floris (UK)			€	445.00	€	445.00
Cristoph Friedrich (Germany)			€	445.00	€	445.00
E. K. U. Gross (Germany)			€	441.67	€	441.67
Philipp Hansmann (France)	€	149.00	€		€	149.00
Karsten Held (Austria)			€	331.25	€	331.25
Bo Hellsing (Sweden)	€	291.67			€	291.67
Sohrab Ismail-Beigi (USA)	€	500.00	€	441.67	€	941.67
Leeor Kronik (Irael)	€	500.00	€	441.67	€	941.67
Frank Lechermann (Germany)			€	445.00	€	445.00
Ivan Leonov (Germany)			€	445.00	€	445.00
Alexander Liechtenstein (Germany)			€	331.25	€	331.25
Martin Lueders (UK)			€	445.00	€	445.00
Takashi Miyake (Japan)	€	333.33	€	441.67	€	775.00
Mark Van Schilfgaarde (UK)			€	441.67	€	441.67
					€	0.00
			Total Travel & Accommodation		€	**12,130.69**

WORKSHOP REFRESHMENTS / CONFERENCE DINNERS

Date	Tea / Coffee		Lunch / Dinner		Total (€'s)	
18-21/6/2012	€	2,846.67			€	2,846.67
18/6/2012			€	3,666.67	€	3,666.67
					€	0.00
					€	0.00
					€	0.00
			Total Refreshments		€	**6,513.33**

MISCELLANEOUS EXPENDITURE

Details	Total (€'s)	
Silke Biermann (France): per diem	€	133.33
Matteo Cococcioni (USA): per diem	€	133.33
Ferdi ARYASETIAWAN (Sweden): per diem	€	133.33

Kieron Burke (USA): per diem		€	133.33
Klaus Capelle (Brazil): per diem		€	133.33
Philipp Hansmann (France): per diem		€	133.33
Bo Hellsing (Sweden): per diem		€	133.33
Sohrab Ismail-Beigi (USA): per diem		€	133.33
Leeor Kronik (Irael): per diem		€	133.33
Takashi Miyake (Japan): per diem		€	133.33
Total Miscellaneous	€	1,333.33
Total Travel & Accommodation	€	12,130.69
Total Refreshments	€	6,513.33
Total Miscellaneous	€	1,333.33
TOTAL EXPENDITURE	€	19,977.36