Ψ_k Newsletter

AB INITIO (FROM ELECTRONIC STRUCTURE) CALCULATION OF COMPLEX PROCESSES IN MATERIALS

Number 79 February 2007

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Contents

1 Editorial

First of all a very Happy and Successful New Year to all our readers!

In this first issue of the 2007 Psi-k Newsletter we have a number of workshop/meeting/conference announcements, both in the ESF Psi-k Programme section and in the general section, dedicated to meetings that are not directly funded by the Psi-k programme.

In particular, we would like to turn readers attention to the Psi-k Training section and the announcement of the Psi-k Training Graduate School to take place on 25-31 March, 2007, in Bristol, UK. In this section we also have an announcement of a Summer School on "Ab-initio Many-Body Theory" to take place on 22-29 July, 2007 in San Sebastian.

The ESF Psi-k Programme section, in addition to the workshop announcements, contains also two long reports on recent workshops, both including also abstracts of presented papers. Position announcements as well as abstracts of the newly submitted and recently published papers appear just before the scientific highlight of the present issue which is on "Exciting prospects for solids: Exact-exchange based functionals meet quasiparticle energy calculations" by Patrick Rinke (Berlin), Abdallah Qteish (Berlin and Irbid), Jörg Neugebauer (Düsseldorf) and Matthias Scheffler (Berlin and Santa Barbara).

Please check the table of content for details.

The *Networks* have a home page on World Wide Web (WWW). Its *Uniform Resource Locator* (URL) is:

http://psi-k.dl.ac.uk/

The above contains information on the forthcoming Psi-k 2007 workshops and hands-on courses (subject to funding).

Please submit all material for the next newsletters to the email address below.

The following email addresses, which remain in operation, are repeated for your convenience, and are the easiest way to contact us.

	function
psik-coord@dl.ac.uk	**messages to the coordinators, editor & newsletter**
psik-network@dl.ac.uk	**messages to the whole Ψ_k community**

Z.(Dzidka) Szotek, Martin Lüders and Walter Temmerman
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2 News from the ESF Programme

"Towards Atomistic Materials Design"

2.1 ESF Psi-k Programme Workshop Announcements

2.1.1 Psi-k/CECAM Workshop on "Multiscale Approaches to Nanomechanics"

5-7 February 2007, CECAM Lyon, France

`http://www.cecam.fr/index.php?content=activities/workshop&action=details&wid=70`

Objectives: This meeting is focused on the application of first-principles computer simulations to the study of microscopic aspects that determine the mechanical behavior of materials. This includes the structure and interaction of atomic-scale defects involved in macroscopic failure (crack tips, dislocation cores, grain boundaries, etc), mechanical and electronic transport properties of nanocontacts, and the interaction between nanoasperities (tips) and surfaces which is behind both the operation of scanning probes and the tribological contact between two materials. All these problems share an essentially multi-scale nature that is not amenable to a hierarchical or sequential treatment of the different length (or time) scales: the breaking and formation of atomic bonds are dynamically coupled by long-range interactions (elasticity) to the macroscopic boundary conditions (deformation, load) imposed.

Confirmed invited speakers and session chairs:

N. Agrait (UA Madrid, E)
G. Ciccotti (U Rome, I)
G. Csanyi (U Cambridge, UK)
O. Custance (U Osaka, J)
A. de Vita (Kings College London, UK)
M.W. Finnis (Imperial College London, UK)
J. Frenken (U Leiden, NL)
P. Gumbsch (U Karlsruhe, D)
K.W. Jacobsen (DTU Lingby, DK)
H. Jonsson (U Reykjavik, IS)
L. Kantorovich (King's College London, UK)
J. Krim (NC State Univ., Raleigh, USA)
A. Minor (LBL, Berkeley, USA)
C. Molteni (Kings College London, UK)
M.C. Payne (U Cambridge, UK)
B.N.J. Persson (FZ Juelich, D)
A. Shluger (UCL, London, UK)
B. Smit (CECAM Lyon, F)
M. Urbakh (U Tel Aviv, IL)
U. Valbusa (U Genoa, I)
E. Vanden Eijnden (Courant, NY, USA)
E. van der Giessen (U Groningen, NL)
S. Yip (MIT, USA)

Format: The workshop will run for two and a half days, starting on February 5th (Monday) at 2pm and ending on February 7th (Wednesday) around 5:30pm. We expect a meeting with about 45-50 participants with an approximate ratio 2:1 between theory and experiment. Apart from the formal sessions of invited contributions, we encourage all workshop participants to present their work in a Poster Session on Monday evening.

Ruben Perez (Universidad Autonoma de Madrid)
Karsten Reuter (Fritz-Haber-Institut, Berlin)

WORKSHOP ANNOUNCEMENT

2.1.2 Workshop on "Ab-initio approaches to electron-phonon coupling and superconductivity"

May 28-30, 2007 San Sebastian-Donostia, Basque Country, Spain

Organisers: Ole K.Andersen, Evgueni V.Chulkov, Igor I.Mazin, and Warren E.Pickett

See website for applications:
http://dipc.ehu.es/eph2007

The aim of this workshop is to give an overview of recent ab- initio calculations of electron-phonon coupling in superconducting and nonsuperconducting materials, in bulk as well as at the surface. The workshop will cover, for example, linear response calculations of the e-ph coupling resolved both in energy and momentum, the newly developed superconducting density functional theory, and numerical methods of the Eliashberg theory. Important topics like anharmonicity, nonadiabacity, and unconventional order parameters will also be discussed, as well as spectroscopic effects of the e-ph coupling, e.g., in photoemission.

Although the workshop is mainly theoretical, we anticipate having some experimentalists as well, and hope that this will lead to productive cross-fertilization between the theory and the experiment.

We plan to have about 30 oral invited talks and unlimited number of contributed posters. The workshop will take place in the picturesque city of San Sebastian, one of the main cities of the legendary Basque Country.

We do not foresee a necessity to levy any conference fee, and particularly encourage young researchers to join the workshop. The deadline for applications is March 15, 2007.

Important dates:

January 17: First announcement. Registration OPEN.
March 15: Deadline for application and abstract submission.

2.1.3 First Announcement for Workshop on Multiscale approach to alloys: advances and challenges

Place: Stockholm/Sigtuna, Sweden

Date: June 17 - June 19, 2007

Organizers
Igor A. Abrikosov (Linköpings University, Sweden);
H. Dreysse (Universite Louis Pasteur, France);
G. Ceder (Massachusetts Institute of Tecnology, USA);
M. Asta (UC Davis, USA);
L. Dubrovinsky (Universität Bayreuth, Germany)

WebPage under construction

Topic: The goal of the conference is to bring together leading experts working in the field of multiscale modeling for alloys, and to exchange the expertise on the recent advantages in the field, as well as to discuss challenges on the way to the development of a consistent set of tools for ab initio simulations of disordered materials at all levels, from the electronic structure towards the microstructure. We also invite leading experimentalists in the fields of spectroscopy, nanoscience, structural chemistry, and geophysics, to establish closer links between theory and experiment.

Preliminary list of invited speakers:
G. M. Stocks (Oak Ridge National Laboratory, USA) - confirmed
C. Wolverton (Northwestern University, USA)
B. Johansson (KTH, Sweden, and Sandvik Steel) - confirmed
H. Skriver (DTU, Denmark) - confirmed
L. Vitos (Budapest, Hungary and Stockholm, Sweden) - confirmed
B. Gyorffy (University of Bristol, UK) - confirmed
J. Neugebauer (Max-Planck-Institute, Dusseldorf, Germany) - confirmed
Derwyn Rowlands (University of Bristol, UK) - confirmed
Dario Alfe (University College London, UK)
H. Dreysse (Universite Louis Pasteur, France) - confirmed
M. Katsnelson (Nijmegen University, The Netherlands)
A. V. Ruban (KTH, Sweden) - confirmed
P. A. Korzhavyi (KTH, Sweden) - confirmed
S. I. Simak (Linköping University, Sweden) - confirmed
M. Asta (UC Davis, USA)
A. Zunger (NREL, USA)
S. Lidin (Stockholm University, Sweden)
L. Dubrovinskii (Bayreuth, Germany) - confirmed

Axel van de Walle (Cal Tech, USA) - confirmed
M. Sluiter, (Delft, The Netherlands) - confirmed
H. Reichert (MPI Stuttgart, Germany)
BerndSchönfeld (ETH, Switzerland)
Anton Van der Ven (University of Michigan, USA)
D. Johnson (UIUC, USA) - confirmed
P. Turchi (LLNL, USA)
D. Morgan (University of Wisconsin, USA)
Adam Kijena (Uniwersity of Wroclaw, Poland) - confirmed
M. Alatalo (Lappeenranta University of Technology, Finland) - confirmed

In addition, we encourage participation of young researchers and PhD students. Web page for the workshop is under construction.

Deadline for abstract submission is April 16, 2007

For more information and registration please, contact
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2.2 Reports on ESF Psi-k Programme Workshops/Meetings

2.2.1 Report on the 11th Nanoquanta Workshop on Electronic Excitations: A decade of applications of the Bethe-Salpeter Equation

Houffalize (Belgium)

19-22 September 2006

SPONSORS

NANOQUANTA Network of Excellence
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ORGANIZERS

Valerio OLEVANO
(LEPES-CNRS, Grenoble, France)

Jean-Yves RATY
(Université de Liège, Liège, Belgium)

John J. REHR
(University of Washington, Seattle, USA)

Gian-Marco RIGNANESE
(Université Catholique de Louvain, Louvain-la-Neuve, Belgium)

Patrick RINKE
(Fritz-Haber-Institut, Berlin, Germany)

Francesco SOTTILE
(Universidad del Pais Vasco, San Sebastián, Spain)

Ludger WIRTZ
(IEMN/ISEN, Villeneuve d'Ascq, France)

WEBSITE

`http://www.pcpm.ucl.ac.be/etsf/nq06`

The workshop gathered 114 participants from leading international groups. There were 39 oral presentations (17 invited speakers and 22 contributed talks) and 30 posters. The workshop allowed the participants to discuss the advances in the theoretical and computational treatment of optical and dielectric spectroscopy in the framework of many-body-perturbation-theory (MBPT).

Invited speakers from leading international groups gave an in-depth overview of current research activities in MBPT (and related fields) and placed recent results into context. Young researchers (Ph.D. students and post-docs) also had the opportunity to present their work (more than half of the oral presentations were given by non-permanent researchers).

A special emphasis was placed on the assessment of ten years of the Bethe-Salpter Equation (BSE) method presenting its achievements and developments [Session I], applications [Session II], advantages and drawbacks in comparison to the more recent developments of time-dependent density-functional theory (TDDFT) [Session IV & V]. Very interesting discussions took place on the complementarity of the two methods and on the future of BSE with respect to TDDFT.

The state-of-the-art implementations of these ab initio methods into existing numerical codes was presented in this meeting. In the same session, an invited speaker described the most recent experimental advances in the field [Session III].

A session was also devoted to the calculation of excited states using Quantum Monte Carlo simulations [Session VI]. Despite the recent advances which have been presented, the method still remains very demanding in terms of computer time.

A series of presentations focused on quantum transport [Session VII] which is an emerging field of computer simulations and whose importance is increasing in the MBPT community. Indeed, the Green's functions formalism at the heart of MBPT is also particularly well suited to describe electronic quantum transport. Currently, it is used both in the Landauer-Büttiker approach and in the non-equilibrium Green's functions (NEGF) theory, also known as Keldysh formalism. Approaches based on TDDFT have also been presented in this session.

A session was devoted to the latest results obtained in the framework of "Optimized Effective Potentiel"(OEP) within density-functional theory [Session VIII]. It was followed by a series of presentations of applications to surfaces and nanostructures [Session IX]. The last session [Session X] focussed on applications of MBPT.

The poster session gave rise to very interesting discussions. Indeed, wide variety of subjects were being presented and the discussions went well beyond these subjects. The meeting provided an informal atmosphere for stimulating discussions between researchers working in this exciting field. New collaborations were initiated following these discussions.

The presentations were collected (a few are still missing) and are available on the conference website. They will be very useful for both the participants and those people in the field that did not have the opportunity to attend the meeting. In particular, the review talks may also be used as a starting point to enter the field.

Program

TUESDAY, 19 September 2006	
SESSION I : Bethe-Salpeter Equation (Chair : Patrick Rinke)	
9:00	Welcome
9:10	Giovanni Onida (30'+10') The Bethe-Salpeter equation and ab-initio calculations: from the first adventure to a well established tool
9:50	Eric Shirley (30'+10') Progress in GW/Bethe-Salpeter Calculations: what do we know, what do we not know?
SESSION II: BSE applications (Chair : Friedhlem Bechstedt)	
11:00	Claudia Ambrosch-Draxl (25'+5') The Bethe Salpeter Equation applied to organic semiconductors: a critical review
11:30	Catalin Spataru (25'+5') Excitons and their radiative lifetime in semiconducting nanotubes
12:00	Olivia Pulci (25'+5') Electronic and Optical properties of surfaces: ab-initio calculations within MBPT and TDDFT approaches
SESSION III : From experiment to codes via theory (Chair : Gian-Marco Rignanese)	
14:30	Simo Huotari (30'+10') Inelastic x-ray scattering in studies of electronic excitations
15:10	Jelena Sjakste (15'+5') Ab initio calculation of the excitonic linewidth due to electron-phonon interaction in semiconductors
15:30	Yann Pouillon (15'+5') Software integration and development within Nanoquanta
POSTER SESSION	

WEDNESDAY, 20 September 2006	
SESSION IV: Time-dependent density-functional theory (Chair : John Rehr)	
9:00	Mark Casida (25’+5’) Polarization Propagator Corrections to Linear Response Time-Dependent Density-Functional Theory for Molecules with Closed- and Open-Shell Ground States
9:30	Serdar Ogut (25’+5’) Is Nano that Different from the Bulk?
10:00	Olena Butriy (15’+5’) Electron-nuclear interaction from Time-Dependent Multi-component Density Functional Theory
10:20	Hans-Christian Weissker (15’+5’) Dielectric Function of Silicon for Finite Momentum Transfer Signatures of Short-Range Many-Body Effects
SESSION V: TDDFT vs BSE (Chair : Rodolfo Del Sole)	
11:10	Ilya Tokatly (25’+5’) Bethe-Salpeter equation from TDDFT perspective: Diagrammatic representation of the exchange-correlation kernel
11:40	Lucia Reining (25’+5’) Many-Body Perturbation Theory and Density-Functionalapproaches: successful combinations
12:10	Esa Räsänen (15’+5’) Control of electronic currents by laser in quantum rings
SESSION VI: Quantum Monte Carlo (Chair : Francesco Sottile)	
14:30	Claudia Filippi (25’+5’) Excitations in (bio)molecules from quantum Monte Carlo
15:00	Paolo Umari (25’+5’) Linear and non-linear dielectric response of periodic systems from quantum Monte Carlo

THURSDAY, 21 September 2006

	SESSION VII : Quantum Transport (Part 1) (Chair : Carl-Olof Almbladh)
9:00	Ralf Gebauer (25’+5’) Quantum kinetic simulations of electron transport in nanosystems
9:30	Stefan Kurth (25’+5’) An Approach to Quantum Transport Using Time-Dependent Density-Functional Theory
10:00	Kristian Thygesen (25’+5’) Non-equilibrium GW approach to electron transport in nanoscale contacts
	SESSION VII : Quantum Transport (Part 2) (Chair : Rex Godby)
11:00	Hector Mera (15’+5’) Non-local self-energies in quantum transport calculations
11:20	Pierre Darancet (15’+5’) GW description of Correlation effects in Quantum transport on real systems
11:40	Peter Bokes (20’+5’) Dynamics of electrons and ab-initio modeling of quantum transport
12:05	Carl-Olof Almbladh (20’+5’) Classical Nuclear Motion in Quantum Transport
	SESSION VIII: DFT OEP band gap (Chair : Xavier Gonze)
14:30	Fabio Della Sala (15’+5’) Kohn Sham gaps, potentials and excitation energies of finite systems using effective exact exchange-correlation potentials
14:50	Myrta Grning (15’+5’) The effect of nonlocality on the band gap within DFT
15:10	Sangeeta Sharma (15’+5’) Non-collinear magnetism within OEP with a view to spin dynamics

SESSION IX: Optical properties applications: surfaces and nanostructures
(Chair : Ludger Wirtz)

16:00 Luis Ramos (20'+5')
Additional many-body effects in the absorption spectra of Si nanocrystallites
16:25 Federico Iori (15'+5')
Optical Spectra of doped Silicon Nanocrystals
16:45 Daniele Varsano (15'+5')
Exciton localisation of quasi 1D systems and optical properties
17:15 Katalin Gaal-Nagy (15'+5')
Electronical and optical properties of the Si(113)-3x2 ADI surface:
An ab initio study
17:35 Andrei Incze (15'+5')
Ab initio study of optical spectra for oxidized Si(100) surfaces
17:55 Arno Schindlmayr (15'+5')
Electronic Properties of Point Defects at the GaAs(110) and InP(110) Surfaces

FRIDAY, 22 September 2006

SESSION X : MBPT and GW applications (Part 1)
(Chair : Ulf von Barth)

9:00 Robert van Leuween (30'+10')
Propagation of the the Kadanoff-Baym equations for many-electron systems
9:40 Pablo Garcia-Gonzales (25'+5')
Many-body and TDDFT methods for total energy calculations
10:10 Ricardo Gomez-Abal (15'+5')
An all-electron GW code based on FP-(L)APW+lo: the role of core electrons

SESSION X : MBPT and GW applications (Part 2)
(Chair : Valerio Olevano)

11:00 Martin Stankovski (15'+5')
Local and non-local vertex corrections in GW for extended and localised systems
11:20 Riad Shaltaf (15'+5')
First Principle Calculations of Band Offsets of SiO_2 and $ZrSiO_4$ with Silicon
11:40 Vladen Zhukov (15'+5')
The first-principle LMTO+U+SO+GW+T approach for the calculations of
dynamic spin susceptibilities and life-times of excited electron in metals

POSTER SESSION

P01. Hakim Amara
How to identify defects in carbon nanotubes : STM study and vibrational properties

P02. Claudio Attaccalite
Effect of impurities on the optical properties of BN nanotubes

P03. Silvana Botti
Identification of CdSe fullerene-based nanoparticles
from optical spectroscopy calculations

P04. Damien Caliste
A visualisation tool : V_Sim

P05. Giancarlo Cappellini
Electronic excitations of oligoacenes in four different charge states: -1, 0, +1 and +2

P06. Lucia Caramella
Optical properties of oxidized Si(100)(2x2) surface and local
fields contributions on electron energy loss spectrum

P07. Alberto Castro
Time-dependent electron localisation function:
A tool to visualise and analyse ultrafast processes

P08. Marco Cazzaniga
Study of the small-q contribution to the polarizability and the intraband term:
from the jellium to the periodic solid

P09. Letizia Chiodo
Thiol adsorption effects on Au(111) work function

P10. Christoph Friedrich
All-Electron GW Approximation in the Augmented-Plane-Wave Basis-Set Limit

P11. Juergen Furthmueller
Single-particle excitation energies starting from generalized Kohn-Sham schemes

P12. Viviana Garbuio
Excited states of formamide in water

P13. Matteo Gatti
Nonlocality and frequency dependence of effective potentials

P14. Matteo Gatti
Electronic excitations in Vanadium Oxide (VO_2)

P15. Christine Giorgetti
Ab Initio calculations of graphene like systems

P16. Ralf Hambach
Anisotropy in EEL-spectra for large momentum transfer
P17. Paula Havu
Finite Element Implementation of Green’s Function Method
for Transport Problems in Nanostructures
P18. Nicole Helbig
Towards a reduced density matrix functional theory for solids
P19. Conor Hogan
Optical properties of the Sb-stabilized GaSb(001) surface
P20. Andrew Morris
Vertex corrections in localised and extended systems
P21. Tapio Rantala
Photoexcitation of Disperse Red 1: Comparison of Approaches
P22. Xinguo Ren
LDA+DMFT computation of the electronic spectrum of NiO
P23. Claudia Roedl
Modifications of BSE Due to Spin Polarization: Antiferromagnetic MnO
P24. Ersoy Sasioglu
Magnetic phase diagram of the Mn-based Heusler alloys from first-principles
P25. Valerie Véniard
Second order harmonic generation in crystalline semiconductors
P26. Claudio Verdozzi
Entanglement in Anderson Nanoclusters
P27. Claudio Verdozzi
Magnetic field effects on optical and transport properties
in InAs/GaAs quantum dots
P28. Matthieu Verstraete
The GW space-time formalism at finite temperatures
P29. Ludger Wirtz
Electronic excitations in hexagonal layered systems (C and BN)
P30. Zeila Zanolli
Ab initio calculation of structural and electronic properties of
InAs and GaAs having wurtzite crystal structure

ABSTRACTS OF PRESENTED PAPERS

Oral presentations

The Bethe-Salpeter equation and ab-initio calculations: from the first adventure to a well established tool

G. Onida$^{(1,2)}$, L. Reining$^{(1,3)}$, R. Del Sole$^{(1,4)}$, S. Albrecht$^{(5)}$,
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We briefly review the formalism and the physics of the Bethe-Salpeter equation approach to compute electron-hole interaction effects in excitation spectra. We then describe how the main formulas developed in the seventies by Hanke and Sham started to be integrated with first-principle methods in the last years of the past century, making the link with Hedin's equations and the GW method. We will point out how a few months were sufficient to trigger contributions from several independent groups, and how this was boosting the progresses of the field through competition and collaboration.

Progress in GW/Bethe-Salpeter Calculations: what do we know, what do we not know?

E.L. Shirley

NIST, 100 Bureau Dr., MS 8441 Gaithersburg, MD 20899-8441 USA

The past several years, especially the last ten or so, have witnessed considerable progress in Bethe-Salpeter calculations. This progress has been in the two areas, methods and theory. We would like to highlight the progress made at key steps along the way. This survey will start with the milestone work by Hanke, Sham and Rice. Trying to cite much of the representative work (though it may be too hard to identify and cite all of the work), we will point out lessons learned and open questions that remain. Especially of interest should, and will be, what should we do next? Some possible answers to this question will be proposed. Finally, I would like to suggest orbital-less approaches to many aspects of (local) dielectric response in condensed matter systems. A motivation for this type of calculation is its possibly improved scaling with system size to describe local screening effects. One can compare this to traditional methods, which can involve summations over large numbers of occupied and virtual orbitals that stretch long distances across a physical system.

The Bethe Salpeter Equation applied to organic semiconductors: a critical review

C. Ambrosch-Draxl (1), P. Puschnig (1), S. Sagmeister (1), and K. Hummer (2)

(1) Chair of Atomistic Modelling and Design of Materials, University of Leoben
(2) Institut für Materialphysik, University Vienna

The exciton binding energy is a central quantity in the photophysics of semiconductors, intimately related to the probability of radiative mission/absorption and electric-field induced generation of free charge carriers. Hence, for organic semiconductors this quantity is of major interest concerning their technological application in opto-electronic devices. And it had been a long-standing problem, whether the lowest absorption features in polymers are strongly bound excitons or free electron-hole pairs.

To answer this question, the optical properties including excitonic effects have been investigated for a variety of organic molecular crystals and polymers during the last years, where the electron-hole interaction has been included by solving the Bethe-Salpeter equation.

In this talk, it will be shown, how the electron-hole interaction can be tuned, either by dense packing or by the choice of the molecule, since the exciton binding energies are extremely sensitive to the crystalline environment and to the molecular size. Thereby the influence of the three-dimensional crystal versus a one-dimensional chain is addressed for the polymers. Moreover, the dependence of the exciton binding energy on the molecular size is studied for several

series of oligomers and compared to those found for the polymers. As last aspect, the influence of the intermolecular interaction on the excitonic effects is investigated by applying hydrostatic pressure, which provides another way of supressing the Coulomb interaction of the electron-hole pair.

Despite the success of the Bethe Salpeter Equation for the puzzling problem concerning the role of excitons in organic semiconductors, a couple of questions remain. Therefore, our results are not only compared to experimental data and surveyed together with other theoretical findings, but we particularly analyze the sensitivity of the results with respect to different approximations applied in the theoretical approaches.

Excitons and their radiative lifetime in semiconducting nanotubes

C. Spataru

Columbia University, 100 Morningside Drive, Ap. 4M, New York, NY 10027, USA

We present ab initio calculation of self-energy and electron-hole interaction (excitonic) effects on the optical spectra of semiconducting single-walled carbon and BN nanotubes. We employed a many-electron Green's function approach that determines both the quasiparticle and optical excitations from first principles. We found important many-electron effects that explain many of the puzzling experimental findings in the optical spectrum of these quasi-one dimensional systems. The calculated optical spectra are in excellent quantitative agreement with measurements, and show prominent features excitonic in nature.

We have also calculated the radiative lifetime of excitons in semiconducting carbon nanotubes. An intrinsic lifetime of the order of 10 ps is computed for the lowest optically active bright excitons. The intrinsic lifetime is however a rapid increasing function of the exciton momentum. Moreover, the electronic structure of the nanotubes dictates the existence of dark excitons nearby in energy to each bright exciton. Both effects strongly influence measured lifetime. Assuming a thermal occupation of bright and dark exciton bands, we find an effective lifetime of the order of 10 ns at room temperature, in good accord with recent experiments.

Electronic and Optical properties of surfaces: ab-initio calculations within MBPT and TDDFT approaches

O. Pulci[(1,2)]

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(2) Dipartimento di Fisica, Università di Roma "Tor Vergata",
Via della Ricerca Scientifica 1, I-00133 Roma, Italy

The microscopic study of complex systems has nowadays reached a high level of accuracy that allows for a deep understanding of the electronic excitations. Ab-initio descriptions of experiments such as direct or inverse photoemission, optical absorption, electron energy loss, have become possible thanks to the huge progress in the theory and the increased computational power. Charged excitations, as well as neutral excitations, can now be studied within the Many-Body Perturbation Theory (MBPT) based on the Green's function formalism. The so called GW approximation allows to calculate the band structure of solids and surfaces with a very good accuracy. Optical spectra of surfaces can be calculated at different levels of approximation: within single particle schemes (DFT or GW), or including the electron hole interaction within the Bethe Salpeter equation (BSE) or the TDDFT approach. Here we present in details results for band structures, electron affinity, and optical spectra of diamond and silicon surfaces. Several approximations used for computing the electronic band structures and optical spectra will be analized and discussed.

Inelastic x-ray scattering in studies of electronic excitations

S. Huotari

European Synchrotron Radiation Facility (ESRF)

Inelastic x-ray scattering (IXS) is a rapidly developing tool in the studies of excitations in condensed matter. The spectral range covers a multitude of phenomena, from phonons (a few meV) via valence-electron plasmons (a few eV) to core-electron excitations (10 eV to several keV). Basically two different types of experimental setups can be utilized, namely resonant and non-resonant IXS. In resonant IXS, photon energy is tuned to a core level threshold of the target material and several otherwise weak or forbidden excitations can be resonantly enhanced. In non-resonant IXS, the information obtainable is the dynamic structure factor, which is in turn directly related to the macroscopic dielectric function. This technique is closely related to electron energy-loss spectroscopy with certain advantages and disadvantages. These issues will be reviewed and recent developments in synchrotron radiation instrumentation will be discussed in terms of which experiments are feasible today with the IXS method.

Ab initio **calculation of the excitonic linewidth due to electron-phonon interaction in semiconductors**

J. Sjakste$^{(1)}$, N. Vast$^{(2)}$, and V. Tyuterev$^{(1,2)}$

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In indirect band gap semiconductors at low impurity and carrier concentrations, the linewidth of the direct band gap exciton E_0 is controlled by the interaction of the excited electron with short-wavelength phonons [1-3]. This linewidth is temperature- and pressure-dependent, and is due to phonon-assisted scattering of the excited electrons from the Γ point to the local minima of the conduction band, which are lower in energy.

We report on *ab initio* calculation of the excitonic linewidth in GaP and in GaAs under pressure. LDA approximation was used to describe the topology of the conduction bands and the electronic wavefunctions in semiconductors. The phonon frequencies and the corresponding perturbations of the self-consistent potential were calculated in the framework of the Density-Functional Perturbation Theory. We have extended the calculation of the electron-phonon coupling for metals to the case of semiconductors: integration of the electron-phonon coupling over the final states has been performed with our TAU-ISDP extention of the *espresso* package [4].

Our results are in good agreement with experimental results of spectroellipsometry for both GaP and GaAs under pressure [1,2], thus giving confidence in the LDA description of the conduction band, which can be used for the prediction of properties related only to the conduction bands of semiconductors, such as electron-phonon interaction.

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Software integration and development within Nanoquanta

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(2) Nanoquanta / ETSF File Formats Team

IT9 is labelled as “the team for integration of theory and code development”. Its task constitutes an important component of ETSF. Presently each node has complementary theoretical capabilities and software tools, that should be integrated (beyond some existing bilateral or multilateral exchange of data), after the period covered by the FP6 action. Regarding data files, the work is focused on the wavefunction files in a planewave basis, for which there is now one flexible and documented format [1], based on NetCDF [2]. For other file types (crystallographic data, pseudopotentials, etc.), the discussion and documentation of file formats and the development of interconversion utilities is ongoing.

The organisation of a mini-workshop in November 2005 has allowed further progress along these lines. In addition to the Nanoquanta / ETSF file format specifications, it is envisioned that, as a result of this mini-workshop, IT9 might make a request of funding of one or more computer scientists, paid by the network, whose task would be to write conversion utilities, or implement I/O routines in existing software, and later, create a package with all available Nanoquanta Free Software [3].

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Polarization Propagator Corrections to Linear Response Time-Dependent Density-Functional Theory for Molecules with Closed- and Open-Shell Ground States

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A decade ago time-dependent density-functional theory (TDDFT) was introduced to the quantum chemistry community in its linear response (LR) for calculating excitation spectra [1]. Since that time LR-TDDFT has become a method of choice for calculating molecular vertical excitation energies because of its demonstrated accuracy within the domain of applicability [2] determined by present-day approximate exchange-correlation functionals. One limitation of the method comes from the wide-spread use of the TDDFT adiabatic approximation which limits LR-TDDFT to quasiparticle spectra (i.e., one-electron excitations). Two-electron excitations are important when calculating the excitation energies of closed-shell molecules both (i) because of the importance in some molecules of excited states with substantial two-electron character and (ii) because the inclusion of two-electron excitations allows a better description of biradical ground states. Spin-flip excitations are a particular type of two-electron excitation which is important for a correct description of the excitation spectra of molecules with open-shell ground states. Some of these points are nicely illustrated in Ref. [3]. In this talk I plan to outline how the equation-of-motion superoperator approach may be used to derive nonadiabatic polarization propagator corrections [4] to adiabatic LR-TDDFT for application to the above mentionned problems.

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Is “Nano” that Different from the Bulk?

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Large-scale first principles computations for the optical absorption spectra of medium size Si clusters (with 20 to 28 atoms) are presented. The computations are performed using time-dependent density functional theory within the adiabatic local density approximation (TDLDA). We find that prolate and compact clusters have distinct shape signatures, but no clear size-dependence for a given shape in this size range. The shape dependence and size-independence of the spectra, and most of the peak positions and intensities can be explained remarkably well within the *classical* Mie theory, developed for light absorption by *metallic* particles, using the *bulk* dielectric function of Si. Moreover, the experimental spectrum of Si_{21} is in very good agreement with the theoretical spectrum of the prolate cluster, which is lower in energy than the compact one at this size. TDLDA results presented here for the dynamical response of Si clusters to time-varying electric fields provide further support for (i) their metallic behavior, as suggested in previous studies of their response to static electric fields and (ii) recent studies of static, but wavevector-dependent, screening in hydrogenated Si quantum dots, which have indicated that the bulk dielectric function is able to capture the essential screening in nano-objects as long as the appropriate boundary conditions are taken into account through classical electrostatics.

Electron-nuclear interaction from Time-Dependent Multi-component Density Functional Theory

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We use Time-Dependent Multi-component Density Functional Theory (TDMCDFT) to describe the electron-nuclear system fully quantum mechanically. In this theory the electron density and nuclear density matrix of the system are the fundamental variables. The approach allows to describe both the electron and the nuclear motion in the system.

We use TDMCDFT to treat electron-nuclear many-body effects, which are not described within the framework of pure electron Time-Dependent Density Functional Theory (TDDFT).

The success of the approach depends on the approximation for the exchange-correlation functionals used in TDMCDFT. The necessary exchange-correlation functionals can be derived on the basis of many-body perturbation theory.

Dielectric Function of Silicon for Finite Momentum Transfer – Signatures of Short-Range Many-Body Effects

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We present an investigation of the dynamic structure factor and of the dielectric function of the prototypical semiconductor silicon for finite momentum transfer, combining Inelastic X-Ray Scattering and ab initio calculations. We show that, in contrast to optical spectra, for finite momentum transfer Time-Dependent Density-Functional Theory in Adiabatic Local-Density Approximation together with lifetime broadening describes the physics of valence excitations correctly. Short-range crystal and exchange-correlation local-field effects become increasingly important with increasing momentum transfer. Major structures in the spectra are governed by a mixing of transitions of positive and negative energies, in striking difference to spectra for vanishing momentum transfer. This mixing gives rise to a pronounced Fano asymmetry in perfect agreement with experiment. Our results demonstrate the quantitative predictive power of the first-principles description.

Bethe-Salpeter equation from TDDFT perspective: Diagrammatic representation of the exchange-correlation kernel

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Many-body perturbation theory (MBPT) provides a regular method for the calculation of excitation spectra in interacting many-electron systems. Formal application of this approach reduces to the solution of Bethe-Salpeter equation (BSE) for a 4-point (or at least a 3-point) function, which is very demanding from the computational point of view. A promising alternative that has been intensively developed over last years relies on time-dependent density functional theory (TDDFT). The main advantage of this theory is that it requires the solution of a much simpler RPA-like equation with the Coulomb potential being replaced by an effective (and in general unknown) interaction, the exchange-correlation (XC) kernel. In this talk I establish an exact correspondence between MBPT and TDDFT. Starting from BSE, I show that the

corresponding ladder diagrams can be resumed to reduce the final theory to the TDDFT form. I derive an exact diagrammatic representation (and an integral equation) for the XC kernel, and discuss possible applications both for a construction of new approximations in TDDFT and for a development of new approximate schemes within MBPT.

Many-Body Perturbation Theory and Density-Functional approaches: successful combinations

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Today, in the framework of solid state physics two main ab initio approaches are used to describe ground- and excited state properties of condensed matter: on one side, static ground state density functional theory (DFT) and its time-dependent extension (TDDFT) for the description of excited states; on the other side, Many-Boby Perturbation Theory (MBPT), most often used in Hedin's GW approximation [1] for the electron self-energy, or the Bethe-Salpeter equation for the calculation of response functions.

Both approaches have led to breakthroughs, but suffer from different shortcomings: MBPT has a relative conceptual clarity and therefore allows one to find good approximations, but calculations are in general numerically very demanding. DFT-based approaches are in principle computationally more efficient, but a generally reliable and at the same time efficient description of exchange-correlation effects within TDDFT is difficult to obtain. In recent years several research groups (most of them within NANOQUANTA) have therefore worked on schemes to combine MBPT and TDDFT, searching for a formulation that would keep the advantages of both approaches.

Orbital-dependent linear response exchange-correlation kernels f_{xc} have been derived along different lines, including a mapping of the Bethe-Salpeter equation onto the TDDFT screening equation, a perturbative approach, diagrammatic expansions, a variational approach, or the use of the density-functional concept within Hedin's equations [2-6]. The different approaches have led to very similar results; in particular an approximation for f_{xc} was proposed that has been shown [2,3] to reproduce continuum and bound excitons for a wide range of materials.

This talk will give an overview of the various approaches, stressing their common content and discussing the success of the proposed approximations for f_{xc} as well as possible problems. The question of how to use such a combination in order to obtain vertex corrections to the self-energy will also be addressed [6].

As can be seen from the references, this talk represents contributions of a large number of colleagues.

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Control of electronic currents by laser in quantum rings

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We show that the excited states of N-electron semiconductor quantum rings (QRs) can be controlled by a circularly polarized laser field [1]. For this purpose, we define a realistic 2D model potential for the QR and treat the influence of a tunable laser field on the electrons with the dipole approximation. In the numerical calculations we apply the time-dependent density-functional theory within the OCTOPUS code package [2], which enables the evolution of the electronic wave functions in real time and real space. This allows us to monitor the total density and current in the QR during the propagation. As a generalization to a recent study on one-dimensional QRs [3], we find that (i) an external magnetic field creating persistent currents does not affect the controllability of the excited states, and (ii) the finite width of the QR considerably changes the dipole matrix elements required for the determination of the pulse length. Finally we discuss the prominent role of the electron-electron interactions as well as the possibilities to fully control the many-electron system by using the framework of the optimal control theory.

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Excitations in (bio)molecules from quantum Monte Carlo

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Despite significant progress in electronic structure methods, attaining an accurate description of the excited states of even relatively small organic molecules remains a demanding task. To this end, we have been developing a theoretical framework for the study of excited states based on quantum Monte Carlo methods. These correlated approaches are an alternative to conventional quantum chemistry and density functional methods, and have been successfully employed to accurately compute ground-state properties of large molecules and solid systems. Their application to excited states is however quite new and required a rethinking of some of the underlying concepts as well as further methodological developments. We will illustrate the performance of our approach with several photoactive (bio)molecules, and show a comparison with other existing theoretical first-principle methods considered promising for the computation of excited potential energy surfaces.

Work done in collaboration with F. Schautz, A. Scemama, M. Zaccheddu, and L. Guidoni.

Linear and non-linear dielectric response of periodic systems from quantum Monte Carlo

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We present a novel approach that allows to calculate the dielectric response of periodic systems in the quantum Monte Carlo formalism. We employ a many-body generalization for the electric enthalpy functional, where the coupling with the field is expressed via the Berry-phase formulation for the macroscopic polarization. A self-consistent local Hamiltonian then determines the ground-state wavefunction, allowing for accurate diffusion quantum Monte Carlo calculations where the polarization s fixed point is estimated from the average on an iterative sequence. The polarization is sampled through forward-walking. This approach has been validated for the case of the polarizability of an isolated hydrogen atom, and then applied to a periodic system. We then calculate the linear susceptibility of molecular-hydrogen chains with different bond-length alternations. The results found are in excellent agreement with the best estimates obtained from the extrapolation of quantum-chemistry calculations. By adopting trial wavefunctions obtained from Hartree-Fock, we can then calculate with great accuracy the second hyper-susceptibility. Finally, we assess the importance of electronic correlations for the calculated linear and non-linear susceptibilities.

Quantum kinetic simulations of electron transport in nanosystems

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In electronic devices of mesoscopic size the semiclassical Boltzmann kinetic equation is commonly used to calculate properties related to electron transport.

Boltzmann equation treats the electrons in terms of their classical probability distribution in phase space, and deals with collision events quantum-mechanically using Fermi golden rule. This formalism explains why conduction electrons subject to a uniform electric field do not accelerate indefinitely, but settle in a steady state regime of constant current, as a consequence of inelastic collisions with the lattice that lead to energy dissipation.

To describe transport in nanoscopic devices having spatial dimensions of the order of the electron wavelength, it is possible to generalize this method to the fully quantum mechanical case. Boltzmann equation is replaced by a quantum mechanical Liouville-master equation for a reduced density operator.

In this talk I present such a kinetic scheme based on a dissipative Liouville master equation. This approach can be used in the framework of a generalized time-dependent density functional theory for dissipative systems and can therefore be used for realistic nanoscopic systems.

In the presentation I will explain the formalism in detail and show its applicability in the case of transport through suspended carbon nanotubes.

An Approach to Quantum Transport Using TDDFT

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The Landauer formalism is a popular method to calculate the current through a nanoscopic system connected to two (or more) macroscopic electrodes and, in combination with techniques of density functional theory, is now widely used to compute current-voltage characteristics of single molecules. However, in the Landauer formalism the electrons are treated as non-interacting particles. In principle, the interaction can be included by Keldysh non-equilibrium Green's functions techniques but the resulting schemes are difficult to cast in a practical form. Time-dependent density functional theory offers a promising alternative description.

Here we present a description of transport based on the time evolution of the non-interacting time-dependent Schroedinger equation. We develop a numerical algorithm for the time propagation of extended states. For simple model systems, the scheme is used to compute the time-dependent current in response to an external dc or ac bias. As expected, for a dc bias the system evolves to the steady state predicted by the Landauer formula. The formalism is also used to study electron pumping in model systems. In the framework of time-dependent density functional theory the algorithm allows for the description of transport of interacting electrons beyond the Landauer formalism.

Non-equilibrium GW approach to electron transport in nanoscale contacts

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Correlation effects within the *GW* approximation have been incorporated into the Keldysh non-equilibrium electron transport formalism. We show that *GW* describes the Kondo effect and the zero-temperature transport of the Anderson model reasonably well. Combining the *GW* scheme with density functional theory and a Wannier function basis set, we illustrate the impact of correlations by computing the conductance of small molecules attached between metal leads. Our results indicate that self-consistency in the *GW* self-energy is fundamental for transport calculations.

Non-local self-energies in quantum transport calculations

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We calculate the conductance of a metal-vacuum-metal junction, where exchange-correlation effects are taking into account in terms of a gw-based non-local model self-energy operator. This conductance is then compared with the conductances predicted by both the local density approximated and exact Kohn-Sham theories built from the model self-energy. Significant deviations between these three conductances arise in the strong tunnelling regime. An attempt to connect these deviations with the band-gap problem of density functional theory is made. A brief discussion of the implementation of the self-energy model for atomistic models using a basis of localized numerical orbitals will also be given.

GW description of Correlation effects in Quantum transport on real systems

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In the framework of Non-Equilibrium Green's Function Theory (NEGF), we present an *ab initio* approach to electronic quantum transport in nanoscale systems which includes electron-electron interaction effects through a *GW* approximation on the Self-energy. With respect to DFT approaches to quantum transport, we use a real physical electronic quasiparticle structure into a Landauer-like expression for the conductance on a maximally localized Wannier functions basis. We take into account both the non-hermitean part of the *GW* Self-energy and also its full dynamical dependence, thus going beyond the plasmon-pole model. Preliminary results of a calculation on real systems, cupper and gold mono-atomic chains, will be shown. With respect to DFT, the *GW* conductance profile appears modified both by the hermitean part of the self-energy, which gives rise to a *GW* renormalization of the Kohn-Sham eigenvalues; and also by the non-hermitean part, which is associated to the finite lifetime of electronic states due to electron-electron scattering diffusion mechanisms and to the non coherent part of the electronic transport.

Dynamics of electrons and ab-initio modeling of quantum transport

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Within the many theoretical explorations addressing the steady-state ab-initio description of quantum transport there is relatively little material devoted to the problem of establishment of this steady state. The generally accepted points of view use the switching-on of the localised total electric field or the turning-on of the contacting between two initially biased electrodes. We argue that while both of these are equivalent none is satisfactory already at the time-dependent Hartree approximation. Instead, we show that employing an auxiliary homogeneous electric field turned on for a finite time does act as a source of current I which eventually leads to an induced drop in potential ΔV and thereby to the so called four-point conductance. We show how such a formulation is related to the character of the non-local dielectric function of the quantum junction.

Classical Nuclear Motion in Quantum Transport

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An *ab initio* quantum-classical mixed scheme for the time evolution of electrode-device-electrode systems is introduced to study nuclear dynamics in quantum transport. Two model systems are discussed to illustrate the method. Our results provide the first example of current-induced molecular desorption as obtained from a full time-dependent approach, and suggest the use of AC biases as a way to tailor electromigration. They also show the importance of non-adiabatic effects for ultrafast phenomena in nanodevices.

Kohn Sham gaps, potentials and excitation energies of finite systems using effective exact exchange-correlation potentials

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Conventional approximated exchange-correlation functionals are quite accurate in the prediction of total energy and ground-state conformation of different systems, but they yield quite poor Kohn-Sham(KS) spectra. The KS gap is too low or only works thanks to error compensation [1], and, in particular for finite systems, the highest occupied KS orbital eigenvalue much higher than the ionization potential and only few bound virtual orbitals are present. KS orbitals and eigenvalues are input quantities for time-dependent density functional theory(TD-DFT), thus they must be computed accurately in order to obtain accurate TD-DFT excitation energies. We show that the Localized Hartree Fock [2-4] method, an efficient effective exact exchange KS approach, can be extendend to include an ab-initio-like correlation potential. The non-local energy-dependent second-order self-energy operator [5,6] has been localized to obtain a local KS exchange-correlation potential. We propose a very efficient computational scheme and we present results for atoms, metal and semiconductor clusters, small and medium size molecules. Very accurate KS gap, KS potentials and TD-DFT excitation energies have been obtained.

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The effect of nonlocality on the band gap within density functional theory

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In this talk I compare the eigenvalue band gap found with a nonlocal potential within the generalized KS (GKS)[1] scheme and within the KS scheme using the optimized effective potential(OEP)[2] approach. Since I use the same exchange-correlation approximation (Coulomb-Hole screened-exchange) the difference in the band gap is due solely to the locality/nonlocality of the potential. In fact, I show that the difference between the OEP-KS and the GKS band gaps comes from the derivative discontinuity of the exchange-correlation energy that corresponds to the nonlocal potential [3].

Once the derivative discontinuity is taken into account, the corrected KS and generalized KS gaps are very close to each other. Finally I discuss whether the GKS schemes can be successfully applied to calculations of optical absorption spectra of semiconductors and insulators.

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Non-collinear magnetism within OEP with a view to spin dynamics

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The dynamics of spin degrees of freedom is responsible for phenomena like spin injection, spin wave excitations, spin filtering etc. These mechanisms are crucial for recent developments e.g. spintronics and spin transport. The advent of time dependent extension of SDFT has made the ab-initio treatment of spin dynamics possible. Crucial to the practical calculations using SDFT is the approximations made for the exchange-correlation energy functional. In calculations this is most often approximated with the LSDA, GGA or there time dependent extensions. These have been developed for collinear magnetism, and their use in non-collinear situations relies on the magnetisation ($\mathbf{m}(\mathbf{r})$) and exchange correlation magnetic field ($\mathbf{B}_{\rm xc}(\mathbf{r})$) being made collinear in a local reference frame at each point in space and at each time. Under such an approximation the $\mathbf{m}(\mathbf{r}) \times \mathbf{B}_{\mathrm{xc}}(\mathbf{r})$ vanishes everywhere in space. This renders all such calculations unsuitable for ab-initio spin dynamics in the adiabatic approximation, a serious limitation of the existing functionals.

In this conference I will talk about the generalization of OEP formalism for SDFT to non-collinear magnetic systems. Crucially, I do not rely on a condition of local collinearity. Using the exact exchange (EXX) functional and with the example of an unsupported Cr(111) monolayer, I will show that in general magnetisation and $\mathbf{B}_{\mathrm{xc}}$ are not parallel – in contrast to what is usually assumed in all to date non-collinear calculations. This renders the TD extensions of EXX functional suitable for the study of dynamical spin effects.

Additional many-body effects in the absorption spectra of Si nanocrystallites

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The quantum confinement of carriers in silicon nanocrystallites (NCs) gives rise to interesting properties such as the increase of the radiative recombination rates. The confinement properties of Si NCs have already been applied in electronic devices such as memories and may be useful for opto-electronic devices and detectors as well. Although an intensive experimental investigation on Si NCs has been carried out in the last two decades, the optical absorption and emission phenomena are not yet well understood. Since the results from experiments usually correspond to an ensemble of Si NCs with different sizes, theoretical investigations of a single Si NC become important to understand the mechanisms of light absorption and light emission in these nanoparticles. The traditional theoretical methods to calculate the absorption spectra based on density-functional theory and on the independent-particle Kohn-Sham scheme are very efficient nowadays, but they do not account for important many-body effects. We present ab initio investigation of Si NCs based on many-body perturbation theory for electronic excitation energies. The GW approximation (G stands for Green's function and W for the screened Coulomb potential) is used to calculate the quasiparticle energies and lifetimes. A dynamical screening is considered in the screened Coulomb potential W. The effects of the electron-hole interaction on the absorption spectra are investigated by means of a two-particle Hamiltonian combining the quasi-electron and the quasi-hole single particle states. We find that a redistribution of the states occurs in a wide energy range of the absorption spectra of Si NCs, which is qualitatively in agreement with results obtained within time-dependent density-functional theory (TD-DFT). The importance of each term in the two-particle Hamiltonian is discussed and compared with available results within TD-DFT.

Optical Spectra of doped Silicon Nanocrystals

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The absorption and the emission spectra of doped silicon nanocrystals including geometry optimization have been calculated within a first-principles framework. Starting from hydrogenated silicon nanocrystals, simultaneous n- and p-type doping with boron and phosphorous impurities have been considered. We found that the B-P co-doping results easier than simple B- or P-doping and that the two impurities tend to occupy nearest neighbours sites rather than other positions inside the nanorystal itself. The co-doped nanocrystals present band edge states localized on the impurities that are responsible of the red shifted absorption threshold with respect to that of pure un-doped nanocrystals in fair agreement with the experimental outcomes. The emission spectra show then a Stokes shift with respect to the absorption due to the structural relaxation after the creation of the electron-hole pair. Moreover the absorption and emission spectra have been calculated for a small co-doped nanocrystal by going beyond the single particle approach through the self-energy correction and the Bethe-Salpeter equation scheme showing the important role played by the many-body effects.

Exciton localisation of quasi one dimensional systems and optical properties

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Atomic chains and polymers are paradigmatic cases where it is known that TDDFT when used with local and gradient corrected density functionals does not give an adequate description of optical properties. We have performed calculations in Many Body Perturbation Theory including electron-hole interaction and in TDDFT using a new non local and energy-dependent kernel [1-3] for trans-polyacetylene and a molecular hydrogen chain. We show that this kernel that until now had been tested on bulk systems and surfaces gives very good results also for finite and quasi one dimensional systems when compared to Bethe-Salpeter results and experiments. The great overestimation of the ALDA in the calculation of the axial polarizability per monomer unit is partially corrected when the new kernel is used and we argue that the polarizability of long finite chains approach the infinte limit when the length of the chain reach the characteristic length of the size of the exciton of the infinite chain. Such localisation of the electron-hole wave function is due to the bond alternation of the polymer and dictates the saturation of the static polarizability with polymer length.

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Electronical and optical properties of the Si(113)-3×2 ADI surface: An ab initio study

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The electronical and optical properties of the high-index (113) surface of silicon with the 3x2 ADI reconstruction have been investigated using ab initio methods. The groundstate calculations for a periodic slab cell have been performed within the plane-wave pseudopotential approach to the density-functional theory as implemented in ABINIT. The optical properties have been obtained from the matrix elements of the momentum operator using the TOSCA package.

First of all, we have investigated the electronic density of states and, furthermore, the electronic band structure where we assigned the surface and the bulk states. These results are discussed in comparison with available experimental data.

The reflectance anisotropy spectra (RAS) has been calculated. A layer-by-layer analysis yields the surface contribution of the spectra, which has initially been derived for the full slab. The states which are responsable for the surface peaks have been identified. In the discussion, also available experimental and theoretical (tight-binding) reference data have been taken into account.

For all calculations, careful tests of the convergence parameters, e.g., the number of special points in the irreducible Brillouin zone, the slab thickness, the number of included conduction states, and the number of plane waves, have been performed.

Ab initio study of optical spectra for oxidized Si(100) surfaces

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Understanding the mechanisms of the oxide growth on the Si(100) surface is a key issue in the miniaturisation of the MOS devices. Research is prompted on the initial stages of the oxidation process as the gate insulators should be thinner than 2 nm [1]. Recently, surface-sensitive linear optical techniques like Reflectance Anisotropy (RAS) or Surface Differential Reflectivity (SDR) were applied for monitorizing in situ the Si(100) surface oxidation in its first stages [2]. However, the link between the way the surface is oxidized and its optical response is still not clear.

Using a plane-wave pseudopotential approach [3,4], we computed the RAS and SDR spectra for a set of representative surface models with nominal coverages from 0.5 ML to 1.5 ML (saturation for room temperature oxidation, molecular oxygen). Based on a model considering that at the surface many possible structures coexist, we were able to describe the evolution of both experimental RAS and SDR with exposure. For exposures up to 4.7 L, a model considering a mixture between two different oxidized surface models and a clean one can account for the changes in optical spectra starting from exposures as low as 0.6 L.

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Electronic Properties of Point Defects at the GaAs(110) and InP(110) Surfaces

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The electronic properties of semiconductor surfaces are strongly influenced by point defects that are either intrinsic or result from the accidental or intentional incorporation of impurities. Electrically active defects, in particular, may act as compensation centers and are responsible for Fermi-level pinning. Two important characteristics are the optical defect levels inside the band gap and the related charge-transition levels. The former can be probed by photoemission spectroscopy, for which the Franck-Condon principle is typically well justified, whereas the latter are thermodynamic quantities specifying the values of the Fermi energy where the charge state of the defect changes. As the theoretical results reported so far deviate significantly from experimental data, a quantitative determination of the electronic properties of individual point defects still remains a challenging task. The reason for the discrepancy lies in fundamental limitations of the computational schemes, which employed density-functional theory within the local-density approximation. As a remedy, we propose a new method that separates structural and electronic energy contributions and includes proper quasiparticle corrections. While the structural relaxation is well described by density-functional theory, we use the *GW* approximation for the self-energy in order to determine the electronic transition energies. In this way the discontinuity of the exchange-correlation potential as well as other shortcomings of commonly used functionals are treated appropriately. Our method is general and can be used to study point defects both at surfaces and in the bulk. As an example, we demonstrate its applicability for anion vacancies at the (110) surfaces of III-V semiconductors. For the P vacancy at InP(110) we find the (+/0) charge-transition level to be 0.82 eV above the surface valence-band maximum. Our result thus shows a clear improvement over the local-density approximation and is in very good agreement with the experimental value 0.75±0.1 eV. For the As vacancy at GaAs(110) we obtain 0.49 eV.

Propagation of the Kadanoff-Baym equations for many-electron systems

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We will present a general approach to solving the equations of motion of the nonequilibrium many-body Green's functions, commonly known as the Kadanoff-Baym equations. We implemented the time-propagation starting from a system in the ground state where for the self-energy we use the second order diagrams and the *GW* self-energy which are both conserving approximations.

In this way we can study the nonequilibrium properties of a wide variety of electronic systems such as atoms, molecules and quantum dots. The initial stage of the calculation requires solving the Dyson equation fully self-consistently for the equilibrium Green's functions. From these initial Green's functions we obtain total energies that we compare to results obtained from variational functionals of the Green's function. We also use time-propagation for calculation linear response functions as a means for obtaining the excitation energies of the system. In our scheme these response functions automatically satisfy important frequency sumrules.

Many-body and TDDFT methods for total energy calculations

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Since the pioneering works by Lein et al. [1] and Holm et al. [2,3]there has been an increasing interest in the evaluation of electron total energies using techniques either based on Many-body Perturbation Theory (MBPT) or on Time-dependent Density Functional Theory (TDDFT). These approaches are much more expensive than conventional implementations of the Kohn-Sham (KS) theory. However, they provide a truly first-principles description of electron-electron correlations without resorting to mean-field-like approximations, thus overcoming many well-known limitations of the KS-LDA or KS-GGA. On the other hand, these MBPT and TDDFT methods do not require the demanding computational effort of quantum Monte Carlo simulations and benefit from the continuous development of efficient algorithms to evaluate the relevant operators required to obtain both ground- and excited-state properties.

In the first part of this talk, I shall briefly summarize and compare both MBPT and TDDFT methods, focusing on their conceptual and technical advantages and limitations when applied to calculate electron total energies. In the second part, I shall concentrate on a recent application of TDDFT, where structural properties and vibrational spectra of hexagonal Boron Nitride are obtained [4]. This system is a paradigmatic example of layered material, where weak (dispersion-like) and strong (covalent) bonds coexist. The Kohn-Sham LDA and, specially, GGA badly fail when evaluating the layer-layer interactions. On the contrary, the in-plane and out-of-plane bonding is well described using TDDFT, also when the layers are pulled apart. This work paves the way for further implementations of truly ab-initio applications in systems where standard Kohn-Sham techniques lacks of predictive accuracy.

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An all-electron GW code based on FP-(L)APW+lo: the role of core electrons

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In recent years the *GW* approximation (GWA), typically applied as perturbation to DFT, has proven to be very successful describing quasi-particle excitations in semiconductors and insulators [1]. Most of the existing codes are based on the pseudopotential (PP) method, which is well established for ground state DFT calculations. In this scheme, the self energy is computed for the valence states only. There is, however, no guarantee that “core-valence partitioning” done in this fashion is also justified for the dynamical self energy, which is a highly non-linear functional of the total density. G_0W_0 results obtained with PP implementations are usually in better agreement with experiments than those reported from existing all-electron implementations. Self-consistent schemes allegedly improve the agreement of the all-electron calculations [2], but drastically worsen the PP results [3,4]. The reasons for this discrepancy and the necessity for self consistency are still a matter of debate [2,3].

In order to address these questions, we developed our own all-electron *GW* code. It is based on the FP-(L)APW+lo method, which currently provides the most reliable results within DFT. In this talk we will give a brief overview of the main characteristics of the code and show quasiparticle energy calculations for Si, GaAs and NaCl. By removing the core contribution to the self-energy and to the DFT-exchange correlation potential we analyze the validity of ”core-valence partitioning” in *GW* calculations.

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Local and non-local vertex corrections in *GW* for extended and localised systems

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A non-local operator like the self-energy can be consistently calculated through many-body perturbation theory. This is usually done within the framework of the *GW* approximation which is obtained by ignoring vertex corrections in Hedin's equations. For a one-iteration G_0W_0 calculation it is only consistent to ignore the vertex correction if the starting point is the Hartree approximation. If the initial Green's function is obtained within a local approximation like DFT-LDA, there is in principle a local vertex given by the static exchange-correlation kernel in the first iteration [1]. We have implemented these local vertex corrections in Hedin's equations for jellium and closed-shell atoms. We present total energies and bandwidths for jellium and the equivalent quantities for He, Be and Ne. We show that a vertex implemented consistently in both the screened interaction and the self-energy leads to unphysical results and that an inclusion of the vertex in only the screened interaction provides results on par with or slightly better than standard G_0W_0. Finally, we discuss non-local vertex corrections derived from a non-local starting approximation for the self-energy and present preliminary results for jellium.

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First Principle Calculations of Band Offsets of SiO$_2$ and ZrSiO$_4$ with Silicon

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The size reduction of MOS transistor requires the usage of alternative materials to replace SiO$_2$ as gate oxide layers. The candidate materials so far were mainly high-κ oxides. As the high-κ/Si interface is formed, it is crucial that the band offsets be large enough to prevent electron or hole injection. In this study first-principle DFT calculations within the local density approximation was used to estimate the band offsets of various interfaces, mainely, SiO_2 and $ZrSiO_4$ with Si. The many body corrections was included as calculated by GW approximation. The calculated band offets of SiO_2 were found to be in agreement with the existing experimental results.

The first-principle LMTO+U+SO+GW+T approach for the calculations of dynamic spin succeptibilities and life-times of excited electron in metals

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We develop a method for calculations of dynamic spin succeptibilities and life-times (inverse line-widths) of low-energy excited electrons in metals. The method includes the evaluation of the lowest term of self-energy within GW approach. The higher self-energy terms are also calculated within T-matrix approach. The T-matrix for the evaluations of the higher self-energy terms and spin succeptibility is obtained by solving Bethe-Salpeter equation with first-principle static RPA screened potential.

The method is based on the linear muffin-tin orbital (LMTO) approach for the band-structure calculations, including also U-corrections (LDA+U) and spin-orbital coupling (SO). The many-body calculations are performed in the basis of the products of LMTOs.

We discuss the results of the line-width and spin susceptibility calculations for Al, Au, Fe, Ni, Pd, Ta, Mo, Rh, Yb, comparing them with experimental data on photoemission spectroscopy, two-photon time-resolved photoemission spectroscopy, tunneling spectroscopy and experiments on measuring inelastic mean free path.

We find that the T-matrix terms of self-energy stronly depend on the details of the band-structure. For the metals with weak screening, i.e. Al, Au, Yb, they provide important contributions to self-energy, bringing the results of calculations in better agreement with experimental data.

We also discuss the ways of improving the methods of solving the Bethe-Salpiter equation in the LMTO basis set.

Posters

How to identify defects in carbon nanotubes : STM study and vibrational properties

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Scanning tunnelling microscopy (STM) and scanning tunnelling spectroscopy (STS) are powerful techniques for investigating the electronic topographic properties of nanomaterials. Whereas the already large amount of experimental STM data obtained so far on perfect nanotubes is perfectly understood, the identification of topological and non-topological modifications of the hexagonal lattice of a carbon nanotube remains an experimental challenge. Thanks to a simple, though accurate theoretical approach, it is possible to simulate the topographic and spectroscopic signatures of many types of defects in graphene and single-walled nanotubes, and to contribute therby to their identification from STM and STS observations. Several illustrations will be provided.

Effect of impurities on the optical properties of BN nanotubes

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The effect of impurities and defects on the optical properties of h-BN and BN nanotube were investigated using first-principle calculations. We showed that recent experiments of photoluminescence in BN nanotube can be explained by the presence of acceptor or donor levels in the band gap that occur in the visible and ultraviolet regions.

Identification of CdSe fullerene-based nanoparticles from optical spectroscopy calculations

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Semiconducting nanoparticles are the building blocks of optical nanodevices as their electronic states, and therefore light absorption and emission, can be controlled by modifying their size and shape. Likewise, optical properties can be a powerful tool to probe the atomic arrangement of synthesized nanoparticles. In view of that, we simulate light absorption of CdSe nanostructures with sizes up to 1.5 nm within density functional theory. We study both bulk fragments with wurtzite symmetry and novel fullerene-like core-cage structures. The comparison with experimental optical spectra allows to confirm the synthesis of these fullerene-based CdSe clusters.

A visualisation tool : V_Sim

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V_Sim [1] is a 3D visualisation program for atomic structures and spin representations. Several interactions are possible, such as, changing the view, displacing an atom, measuring distances and angles, or exporting into image files. Some advanced features allow to draw clipping planes, masking some portion of the rendered crystal ; or to colourise the atoms using some external data. Finally, several atomic files can be rendered one after another as if in a film while all interacting tools are still available.

The current development version includes some plug-ins capabilities allowing the creation of a reader for NETCDF files. Moreover the integration of the OpenBabel library, gives to V_Sim the capability to read a huge variety of file formats used both in the chemistry world and by physicists, such as CML, Gaussian output files...

This program is free software, using commonly used libraries such as OpenGL or GTK.

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Electronic excitations of oligoacenes in four different charge states: -1, 0, +1 and +2

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We present a systematic study of the electronic excitations for the five smallest oligoacenes (naphthalene, anthracene, tetracene, pentacene and hexacene) in their anionic, neutral, cationic and dicationic charge states. We used two different implementations of TDDFT. The low-lying electronic transitions computed by the two methods are found to be in good agreement with previous theoretical results and available experimental data. The strength of the excitonic effects has been also evaluated for the neutral systems. General trends in the electronic excitation properties are discussed as a function of the charge state and molecular size.

Optical properties of oxidized Si(100)(2x2) surface and local fields contributions on electron energy loss spectrum

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We use an efficient scheme to evaluate the independent particle dynamic polarization, used as a starting point in TDDFT to calculate the optical response of a system perturbed from a time dependent external potential. The method, based on an efficient use of the Hilbert transforms, has been implemented in a large-scale ab initio computational code, and the improvement of the scaling of the required CPU time with the system size has been carefully tested also checking the memory occupation of the algorithm and optimizing it. Previous results showing the application of this method to study the optical response of the oxidized Si(100)(2x2) surface on Reflectivity Anisotropy and Surface Differential Reflectivity spectra (RAS and SDR) motivated this work. The starting point of is the remark that the most important modification of the spectrum due to the local fields contributions comes from the component of the perturbing light with polarization perpendicular to the surface plane. We study here the optical response of the same system simulating the Electron Energy Loss Spectrum, where the response to the perpendicular component of the incident light, the most sensitive to the local fields effects, is involved.

Time-dependent electron localisation function: A tool to visualise and analyse ultrafast processes

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The time-dependent electron localisation function (TDELF) is a generalisation of the electron localisation function (ELF) proposed by A. D. Becke and K. E. Edgecombe [1], which allows for time-resolved observation of the formation, the modulation, and the breaking of chemical bonds, and can thus provide a visual understanding of complex reactions involving the dynamics of excited states. We discuss the rationale behind the definition of the ELF, and sketch the general derivation that permits its use for the analysis of time-dependent processes. We present some examples: First, ground-state results showing the characteristic topology of the ELF for various prototypical chemical cases. Secondly, the time-dependent ELF of a few processes is monitored, which demonstrates how it permits to enhance our visual intuition of the electronic behaviour during fast phenomena: the chemical reaction leading to the formation of the water molecule following the collision of a proton and a hydroxide ion, the capture of a proton by the lone pair of the electron rich, smallest imine or unprotonated Schiff base (formaldimine), and the bond breaking of the ethyne triple bond due to the interaction with a strong and ultrashort laser pulse.

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Study of the small-q contribution to the polarizability and the intraband term: from the jellium to the periodic solid

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The computation of the *GW* correction for a bulk jellium shows the importance of considering the intraband contribution in the q→0 limit for the polarizability since its absence induce the appearance of a gap. To include this contribution to the polarizability we propose an approach that avoids the Fermi surface integration, which would require a heavy numerical cost. The approach we propose is based on the idea of detrmining the limiting behaviour of the polarizability

by a fit of its Taylor expansion in the finite small-q region, where the standard computational approach has no numerical problems in evaluating the intraband contribution. In this way we expect will to be able to perform computations not only for the jellium, case in which the RPA polarizability is analitically known (the Lindhard function), but also for real solids.

Our work starts from bulk jellium since the system is simpler, the off-diagonal terms of the polarizability matrix are identically zero, and the polarizability depends only by the modulus of the wave-vector. In this case we know the limiting behavior for the different frequencies. The next step has been to traspose what we learned by the study of the jellium model on the general case. In this situation we deduced the limiting behaviour by theoretical consideration and by the comparison with test simulations.

Thiol adsorption effects on Au(111) work function

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The interface between SAMs and gold surfaces, in particular the Au(111), is one of the most investigated hybrid systems [1-4]. A lot of information is available about the conformation of the adsorbate and the energetic properties of methanthiol on Au(111)[2]. Due to the large computational costs required for these systems, usually short-chain thiol molecules have been investigated ab initio. In particular, being the S-metal interaction dominant in the molecular adsorption geometry, the attention has focused on methanthiol on Au(111), treated as a prototype for SAMs-metal interface. Among electronic properties of these systems, it is known that short chain alkanethiols adsorbed on gold induce a work function reduction [5]. Reported values for Au work function range from 5.0 to 5.3 eV [1,6], and the thiol induced reduction is of 1.2 eV, with maximum values of 1.4 eV [7].

Here, we investigate the effect on the gold work function of adsorbed methanthiol and of CH3(CH2)5SH (hexanethiol) which introduces the effect of the long alkyl chain on the workfunction. Ab initio calculations have been performed in the framework of DFT, using the plane-waves code PWSCF [8], the PW91 parameterization [9] for the exchange-correlation potential, and UltraSoft Pseudopotentials [10]. The coverage of 1 ML corresponds to one thiol molecule for three Au surface atoms, in the hexagonal adsorption geometry. Different adsorption sites and coverages have been calculated to understand the work function behaviour, obtaining results in good agreement with experimental findings.

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All-Electron GW Approximation in the Augmented-Plane-Wave Basis-Set Limit

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The GW approximation for the electronic self-energy is known to yield quasiparticle band structures in much better agreement with experimental measurements than the local-density approximation (LDA) for a large class of materials. However, almost all actual implementations so far are based on the plane-wave pseudopotential approach, which effectively restricts applications to simple metals and sp-bonded semiconductors. In order to overcome this limitation, we have developed an alternative implementation within the all-electron full-potential linearised augmented-plane-wave (FLAPW) method, which is also applicable to systems with localised electrons like transition metals or rare-earth elements. Additional simplifications, such as plasmon-pole models, are avoided.

While the FLAPW method treats the core electrons explicitly and does not suffer from the uncontrolled pseudisation of the wave functions, another source of errors resulting from the linearisation is frequently overlooked. It arises because the radial wave functions in the muffin-tin spheres are expanded around fixed reference energies. Irrelevant for the valence bands in density-functional theory, it cannot be ignored in the *GW* approximation, where unoccupied states more than 100 eV above the Fermi level must typically be included in the self-energy to guarantee converged results. To eliminate this error, we have extended the FLAPW basis set by including second and higher energy derivatives of solutions to the radial scalar-relativistic Dirac equation in the form of local orbitals. With this modification, we show that the linearisation error is eliminated and the basis set becomes complete. In the case of spherical muffin-tin potentials the results in the basis-set limit are identical to augmented-plane-wave calculations. This careful approach finally allows us to comment on the controversy surrounding the apparant discrepancy between pseudopotential and all-electron *GW* results addressed in several recent studies [1,2]. For the example of silicon, we demonstrate that the inclusion of a much larger number of unoccupied states and the elimination of the linearisation error both contribute to an increase of the band gap. While a certain deviation from pseudopotential calculations remains, it is smaller than previously claimed, and both results lie in comparable proximity to the experimental value.

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Single-particle excitation energies starting from generalized Kohn-Sham schemes

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The *GW* approximation of Hedin is arguably the most successful approach for the calculation of quasi-particle (QP) energies. Its accuracy has been proven for a variety of systems. Usually, the QP eigenvalues are calculated in a perturbative approach, starting from solutions of the Kohn-Sham equations with an exchange-correlation (XC) potential in local density or generalized gradient approximation (LDA/GGA). However, this standard approach fails for a number of systems such as InN which share shallow semi-core electrons and a negative gap in LDA/GGA. Here we present *GW* calculations for Si, ZnO and InN which start from solutions of the generalized Kohn-Sham (gKS) equations [1] for the screened exchange (sX [1]), HSE03 [2], PBE0 [3] and HF model of exchange and correlation. Such a procedure is found to yield a positive gap for all the investigated gKS functionals. Furthermore, with exception of HF the resulting gaps are almost the same for all the gKS functionals choosen as starting point. The results are critically analysed with respect to the wave-function character and localization. In addition we discuss the influence of different core-valence XC models. The calculations were performed using a model dielectric function for the *GW* self-energy. The electron-ion interaction was modeled via the PAW method which provides direct access to the all-electron wave functions.

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Excited states of formamide in water

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The electronic structure of liquid water is still not fully elucidated, even though it is essential to understand the chemical and physical properties of many biochemical and industrial processes that occur in solution, where it is crucial to include the role of the solvent in the reactions.

Excited state properties have not been investigated yet, neither for pure water, nor for water as a solvent. We present here ab-initio calculations of pure liquid water and of a solution of formamide. We use snapshots taken from molecular dynamics as input geometries for the study of the electronic and optical spectra. The optical absorption spectra are first obtained within the Density Functional Theory and then calculated by solving the Bethe-Salpeter equation. We show that the effect of the solvent on the optical spectra of the solution can not be neglected.

Nonlocality and frequency dependence of effective potentials

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Many-body perturbation theory (MBPT) [1] is a very powerful method. The key variable is the Green's function $G(r, r', \omega)$ which contains a great deal of information, like the one-quasiparticle spectra. However, one is often interested in just a part of the information carried by G, so in these cases it is preferable to work with simpler quantities from the beginning. This is precisely the case of density-functional theory (DFT) [2], where one wants to obtain the ground-state total energy and density $\rho(r)$. In principle, one doesn't need the full G to obtain this information. In the Kohn-Sham approach [3] it is sufficient to solve an effective one-particle Schrödinger equation, with a local and static potential $V_{xc}(r)$. The Green's function, in turn, is determined by a nonlocal and frequency dependent self-energy $\Sigma(r, r', \omega)$. A possible link between DFT and

MBPT is the Sham-Schlüter equation [4]:

$$V_{xc}(r_1) = \int \quad d\omega dr_2 dr_3 dr_4 \times$$

$$e^{i\eta\omega}\, \tilde{\chi}^{-1}(r_1,r_4)G_{KS}(r_4,r_2,\omega)\Sigma(r_2,r_3,\omega)G(r_3,r_4,\omega)$$

$$\tilde{\chi}(r_1,r_2) = \int d\omega\, e^{i\eta\omega}G_{KS}(r_1,r_2,\omega)G(r_2,r_1,\omega)$$

Here we generalize the Sham-Schlüter equation to situations where one is interested in other parts of the Green's function. For example, one can consider the trace of the spectral function, which is needed to interpret photoemission spectra. In this case a local and dynamical potential $V_{SF}(r,\omega)$ can be introduced. We show that in this case the nonlocality of the self-energy can be converted into a frequency dependence of V_{SF}. This is illustrated for model systems. We discuss also the link with dynamical mean-field theory (DMFT) [5] and other approaches.

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Electronic excitations in Vanadium Oxide (VO$_2$)

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Vanadium Oxide has a metal-insulator transition at T$_C$ = 340 K. The role of correlation in this first-order transition has been debated for a long time (see, for instance, Ref. [1,2]): is VO$_2$ a Peierls or a Mott-Hubbard insulator?

Despite a good agreement with experimental lattice parameters, DFT-LDA yields a negative gap in the insulating phase and hence is not able to give a good answer to this question. This seeming failure of LDA is discussed and different possible approaches to solve this problem are reviewed: from standard perturbative G_0W_0 to self-consistent GW [3,4]. Also results concerning other kinds of electronic excitations (in particular: electron energy loss spectra) are presented.

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Ab Initio calculations of graphene like systems

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We study the possibility to simulate with planar systems, optical absorption and EEL spectra of graphite nanotubes. The calculations are performed ab initio using TDDFT method. We calculate the electronic excitations of graphite and graphene like systems. More specifically, we study the influence of the distance between planes, as well as their stacking to characterize interaction between tubes and walls in multi-wall nanotubes. We also develop a simple model to calculate systems with large inter-plane distance using as reference the system with a smaller one, in order to reduced the size of calculations. The band structure calculations are done using Abinit [1]. The spectra are calculated using DP [2].

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Anisotropy in EEL-spectra for large momentum transfer

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Using Time-Dependent Density Functional Theory (TDDFT), we studied Electron Energy Loss Spectra (EELS) for different graphitic systems at large momentum transfers q. In particular, we investigated their anisotropy with small deviations in q, which seems to occur not only for vanishing (q=0), but also for large momentum transfer, corresponding to reciprocal lattice vectors (q=G) and should be attributed to local-field effects (LFE).

Finite Element Implementation of Green's Function Method for Transport Problems in Nanostructures

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A popular method to model electron transport in nanostructures is to use the simple ballistic transport model including interactions within the density functional theory. We have applied the scheme using the Green's function method which avoids the calculation of electron wavefunctions. Thereby a nanostructure can be connected to the leads by open boundary conditions. Within the nonequilibrium formulation it is also possible to add a finite bias voltage between the leads and calculate the current in a non-linear fashion.

We have implemented the scheme using the finite element method with the so-called p-elements for one-, two- and three- dimensional problems in order to model several types of nanostructures. The method allows a systematic error control and it is not restricted to a particular type of a problem. Moreover, the use of higher order elements results in a convergence which is faster than that obtained by the finite difference method. For the electron-ion interactions we use non-local norm-conserving pseudopotential operators.

We have applied our implementation to model the insulating properties of thin HfO2 layers, two dimensional quantum point contacts, and a magnetic resonant tunneling diode.

Towards a reduced density matrix functional theory for solids

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Reduced-density-matrix-functional theory (RDMFT) is one possible way to treat electron correlation beyond density-functional theory. In order to calculate the fundamental gap we generalize RDMFT to fractional particle numbers. For each fixed particle number, M, the total energy is minimized with respect to the natural orbitals and their occupation numbers. This leads to a function, E_{tot}^{M}, whose derivative with respect to the particle number has a discontinuity identical to the gap. Numerical results are presented for alkali atoms, small molecules, and periodic systems.

Within RDMFT the correlation energy is approximated in terms of the one-body reduced density matrix. One drawback of the first generation functionals is their failure to reproduce the correlation energy of the spin-unpolarized homogeneous electron gas (HEG). Recently, new functionals were introduced which considerably improve the results for atoms and molecules [1]. We apply these functionals to the HEG and show that they also improve the results dramatically. Furthermore, we generalize the second-generation functionals in such a way that they reproduce the correlation energy of the HEG exactly over the whole range of electron densities. We discuss ways to apply the resulting functionals to both finite and inhomogeneous periodic systems.

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Optical properties of the Sb-stabilized GaSb(001) surface

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Thin film InAs/GaSb semiconductor heterostructures show great promise as components of micro- and optoelectronic devices. A proper understanding of the interface formation requires,

however, a thorough understanding of the (clean) GaSb surface before adatom deposition. In spite of several experimental studies of the surface [1], few realistic calculations exist. The structure remains a matter of some controversy, with several c(2x6) and (4x3) reconstructions being proposed on the basis of total energy calculations [2]. The surface sensitivity of reflectance anisotropy (RA) spectroscopy makes it possible to identify (by means of a combined experimental and theoretical approach) the correct surface structure and to understand the underlying electronic structure. High quality experimental data is indeed available for this surface [3]. Of particular interest is the role of heterodimers, which have recently been shown to characterize the RA spectra of GaAs [4]. Recent tight-binding calculations [5] have shown that spin-orbit effects can have considerable and unpredictable effects on RA spectra for compounds containing antimony (Sb). In bulk GaSb, the hole spin-orbit splitting at the Brillouin zone centre is almost as large as the direct gap itself (about 0.8 eV). Hence we also investigate the influence of spin-orbit coupling on the optical properties, using a specially developed interface between the quantum-espresso and SELF codes [6].

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Vertex corrections in localised and extended systems

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Many-body perturbation theory (MBPT) is a leading method for excited-state calculations. It has also been shown to yield good results for total energies. Within MBPT, GW is the most widely used approximation to the self-energy. This ignores the vertex function, setting it to unity. However this is only theoretically justified if the non-interacting Green's function used initially is made from Hartree wavefunctions. This is a poor starting point and in practice we use Density-Functional Theory (DFT) wavefunctions: there is no theoretical justification for this though.

We use the methods of Del Sole *et al.* [1] which we call GW-LDA and $GW\Gamma$-LDA.These use the static exchange-correlation kernel, Kxc, in the vertex function to correct for the DFT starting point. We apply these methods to atomic systems and compare to GW-RPA.

We show that GW-LDA gives similar results to GW-RPA for total energies and first-ionisation energies. However $GW\Gamma$-LDA is poor compared to GW-RPA. We also present results for bulk systems showing that GW-LDA gives more accurate bandwidths than GW-RPA and $GW\Gamma$LDA and compare to a simple non-local vertex.

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Photoexcitation of Disperse Red 1: Comparison of Approaches

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Disperse Red 1 (DR1) is a 38 atomic azobenzene molecule with nonlinear optical response due to its delocalized electrons [1]. We test several one-electron methods, including Hartree-Fock and selected Density Functional Theory (DFT) approximations for prediction of the molecular structure and related photoexcitation within the dipole approximation. Also exact exchange (EXX), and Time-Dependent DFT (TDDFT) approaches [2] are considered [1,3-5].

In one-electron picture the HOMO-LUMO transition is forbidden due symmetry, emphasizing the role of transition probabilities. The TDDFT results do not essentially depend on the chosen exchange-correlation functional and turn out compare best with experimental absorption spectra [1,6]. Performance of the other approaches is discussed.

Success of the rapidly evolving real-space methods is also considered.

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LDA+DMFT computation of the electronic spectrum of NiO

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The electronic spectrum, energy gap and local magnetic moment of paramagnetic NiO are computed by LDA+DMFT(QMC). To this end the noninteracting Hamiltonian obtained within the local density approximation (LDA) is expressed in Wannier functions basis, with only the five anti-bonding bands with mainly Ni 3d character taken into account. Complementing it by local Coulomb interactions one arrives at a material-specific many-body Hamiltonian which is solved by dynamical-mean field theory (DMFT) together with quantum Monte-Carlo (QMC) simulations. The large insulating gap in NiO is found to be a result of the strong electronic correlations in the paramagnetic state. Although the energy gap calculated in this way is of Mott-Hubbard rather than charge-transfer type, the shape of the electronic spectrum in the vicinity of the gap region is in good agreement with the experimental x-ray-photoemission and bremsstrahlung-isochromat-spectroscopy results of Sawatzky and Allen. The value of the local magnetic moment computed in the paramagnetic phase (PM) agrees well with that measured in the antiferromagnetic (AFM) phase. Our results for the electronic spectrum and the local magnetic moment in the PM phase are in accord with the experimental finding that AFM long-range order has no significant influence on the electronic structure of NiO.

Modifications of BSE Due to Spin Polarization: Antiferromagnetic MnO

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Extensive studies of optical properties beyond the independent-particle approximation using the Bethe-Salpeter equation (BSE) have been performed for a wide range of materials, such as bulk semiconductors, surfaces, and molecules. As spin-polarized systems gain more and more interest, it is desirable to describe also their excitation properties within this approach.

Therefore, we consider the effect of spin polarization on the optical properties of materials with collinear magnetization. The Bethe-Salpeter equation for such systems starting from a *GW* treatment of the electronic self-energy contribution is derived. An electron-hole exchange term is taken into account in order to describe local-field effects in the macroscopic dielectric function.

We present numerical results for the antiferromagnetic insulator MnO which can be treated as a paradigmatic example for such a spin-polarized system. The one-particle wave functions are calculated in density-functional theory (DFT) within the generalized-gradient approximation (GGA) using the projector-augmented wave (PAW) method. The screening of the Coulomb interaction in the effective two-particle Hamiltonian is described by a model dielectric function. Instead of solving the eigenvalue problem of the Hamiltonian, an equivalent initial-value problem is considered.

Magnetic phase diagram of the Mn-based Heusler alloys from first-principles

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The magnetic phase diagram of the Mn-based semi- and full-Heusler alloys are determined at T=0 using first-principles calculations in conjunction with the frozen-magnon approximation. We show that magnetism in these systems strongly depends on the number of conduction electrons, their spin polarization and the position of the unoccupied Mn 3d states with respect to Fermi energy. Various magnetic phases are obtained depending on these characteristics. Conditions leading to diverse magnetic behavior are identified. We find that in the case of large conduction electron spin polarization and unoccupied Mn 3d states far from the Fermi level an RKKY-like ferromagnetic interaction is dominating while antiferromagnetic superexchange becomes important in the presence of large peaks of unoccupied Mn 3d states close to the Fermi energy. The overall magnetic behavior depends on the competition of these two mechanisms. The obtained results are in very good agreement with the available experimental data.

Second order harmonic generation in crystalline semiconductors

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A comprehensive understanding of the nonlinear optical properties of solids is crucial for the improvement of nonlinear materials and devices and provides an opportunity to search for new materials. However, the theoretical description of nonlinear effects in solids is a formidable task and important difficulties have delayed any accurate calculations for many years.

We formulate a derivation for the calculation of second-order susceptibility tensor for crystals of any symmetry [1]. For cubic symmetries, we show how this approach can be simplified and expressed in terms of the second-order density response function and of the dielectric function. Numerical results will be presented for SiC and GaAs.

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Entanglement in Anderson Nanoclusters

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We investigate the two-particle spin entanglement in magnetic nanoclusters described by the periodic Anderson model. An entanglement phase diagram is obtained, displaying the role of the entanglement in the temperature dependent competition between local Kondo screening and nonlocal Ruderman-Kittel-Kasuya-Yoshida spin ordering. This provides an entanglement perspective on a central property of magnetic nanoclusters. We also show that multiparticle entangled states are present for finite magnetic field as well as in the mixed valence regime and away from half filling.

Magnetic field effects on optical and transport properties in InAs/GaAs quantum dots

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A photoluminescence study of self-assembled InAs/GaAs quantum dots and under influence of a magnetic field perpendicular to the dot layer is presented. At low temperatures, the magnetic field alters the in-plane transport properties due to localization of carriers in WL potential fluctuations. Decreased transport in the WL results in a reduced capture into the quantum dots and consequently a weakened dot related emission. The effect of the magnetic field exhibits a

considerable dot density dependence, which confirms the correlation to the in-plane transport properties. An interesting effect is observed at temperatures above approximately 100K, for which the magnetic field induced an increment of the quantum dot photoluminescence. This effect is ascribed to the magnetic confinement of the exciton wave function, which increases the probability for carrier capture and localization in the dot.

The GW space-time formalism at finite temperatures

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We present the generalization of the space-time formulation of the GW approximation in many-body perturbation theory. The main changes are introduced in the treatment of the imaginary time and frequency dependency of the polarizability, screening, and self-energy. The discrete Matsubara frequencies and finite imaginary time intervals for integration are taken into account. Efficient fitting and interpolation schemes are developed to avoid a large increase in the grid sizes when going to metallic and finite-T systems.

Electronic excitations in hexagonal layered systems (C and BN)

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We present different phenomena involving electronic excitations in layered systems. For hexagonal boron nitride (BN), we show that excitonic effects play an important role for the optical properties. Measured and calculated values for the band-gap still display a strong scattering between 4.3 and 6.8 eV. We show that optical absorption and electron energy loss spectroscopy cannot be explained in terms of independent-electron transitions but can be properly understood only by proper inclusion of excitonic effects. We discuss the origin of the different values for the measured/calculated band-gaps. For the interpretation of Raman spectra of graphite, the double-resonant Raman model which involves electron-hole pair excitations is qualitatively successful. Quantitative evaluation based on DFT band-structures display, however, a mismatch between theory and experiments. We discuss the achievements and short-comings of the double-resonant model for the explanation of the differences in the Raman spectra of single-layer, few-layer, and bulk graphite.

Ab initio calculation of structural and electronic properties of InAs and GaAs having wurtzite crystal structure

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The growth of new materials is a frequently occurring event in current semiconductor physics. The novelty of the fabricated systems demands a theoretical study of their structural and electronic properties which will provide the necessary knowledge to exploit their features, corroborate the experimental analysis and design the materials of the future.

Motivated by the finding that InAs NanoWires (NWs) grow purely wurtzite and GaAs NWs as a mixture of zinc-blende and wurtzite crystal structures, we focused our attention on the study of the latter polymorph for which no previous calculations exist.

We report our results on ab initio calculations of bulk InAs and GaAs having wurtzite structure. All-electron plane wave calculations within the DFT/LDA approximation are used to obtain the structural properties and the electronic band structure. The latter is computed with and without the inclusion of the Spin-Orbit interaction. The role played by the d electrons is investigated with the use of pseudo-potential calculations.

To properly describe the excited-state properties a method based on many-body perturbation theory is needed. Hence, the quasiparticle band structure was calculated whithin the GW approximation for both materials and for both polymorphs, finding good agreement with very recent experimental results.

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2.2.2 Report on Workshop Models and Theory for Molecular Magnetism

CECAM- Lyon

2006 July 18/21

We would like to thank CECAM, Psi-k and the Netwok of excellence MagmaNet for financial support, which made this workshop possible.

Jens Kortus
TU Bergakademie Freiberg Germany

Carlo Massobrio
Institut de Physique et de Chimie des Materiaux, Strasbourg France

Eliseo Ruiz
Univ. de Barcelona Spain

Mark Pederson
NRL United States

`http://www.cecam.org/index.php?content=activities/pastWorkshops&action=details&wid=42`

Highlights and achievements of the workshop

One possible road to increase the capacity for information storage is decreasing the system size of the storage devices. This eventually suggests the need for a transition from bulk matter to nanoscale molecules and clusters. To achieve that goal one needs the ability to assemble these structures with the required electronic and magnetic properties in a controlled way. To a large extent, this explains the enduring interest in molecular magnetism. The progress in research on molecular magnetism is driven by advances in chemical synthesis of the materials and ingenious experiments. The advances in the capability to predict properties from first-principles calculations gives hope for an rational design of magnetic molecular materials. This workshop has been a focus point of the leading theoretical groups in order to define long term prospects and challenges for future research in the field of molecular magnetism.

The list of invited speakers contained a mix of the leading theoretical groups in the field together with world-wide regocnised leading experimentalists. Further the sessions were complemented by contributed talks from young scientists.

In the following we will only mentions a few highlights the workshop was focusing on.

Highlights day 1

The workshop started with a challenging talk by Roberta Sessoli discussing the emerging trends in molecular nanomagnetism form the point of view of an experimentalist, which was an important start for further discussion. The proper understanding of experimental techniques was another topic discussed by an very educational talk given by Hans-ulrich Güdel which explained how to measure magnetic properties of molecular materials. The morning sessions was then concluded by theoretical talks. Trond Saue and Martin Kaupp demonstrated that a proper theoretical description requires the correct treatment of relativistic effects. The accurate calculation of spin-orbit coupling is one of the major challenges and requirements. That this can be achieved by density-functional theory (DFT) calculations has been shown by several applications to estimate the zero field splitting parameters or the magnetic anisotropy.

The afternoon session started with with general discussion of stereochemistry and spin states of tetrahedral building blocks by Santiago Alvarez and a bottom-up approach for theoretical studies presented by Juan J. Novoa. The power of constrained DFT for the application of exchange parameters was shown by Troy Van Vooris and Coen de Graaf discussed the problems of calculation of magnetic interaction.

One of the main results of the lively discussion of the first day has been the importance of Non-Heisenberg Hamiltons. The Heisenberg-Modell may not be sufficient to describe some magnetic systems accurately enough. As a more general question it appeared the question if theory is indeed calculating the same parameters as experiments measure. The ambiguity appears because both experiment and theory normally map there result to some Hamiltonian. However, the approximations involved in both cases may be different.

Highlights day 2

The interesting discussion from day one on the validity of the Heisenberg Hamiltonian continued. Frederico Totti posed this question directly for multi-nuclear paramagnetic clusters and Jürgen Schnack demonstrated for a Ni_4Mo_{12}-System that the Heisenberg Hamiltonian with constant exchange parameters in indeed unable to explain the observed magnetic field dependence. A possible explanation was found in the suggestion of magnetostriction at the molecular level. Natalie Guihery continued the discussion in the afternoon in mixed valence systems.

John McGrady explored the interface between antiferromagnetism and metal-metal bonding, whereas Mark Cassida introduced the rich phenomena of spin crossover in the discussion. Another new class of materials was discussed by Vincent Robert, which focused on the theoretical aspects of non-innocent ligands in organnometallic chemistry.

Highlights day 3

The third day started with an excellent educational talk by Stephen Blundell on muon studies applied to molecular magnets. The application of limitations of DFT has been shown by Furio Cora in case of some examples form solid state systems. Illaria Ciofini modelled photo-magnetic molecular devices. Andrei Postnikov discussed non-collinear states and frustration, a topic very important for molecular magnetism but other fields of magnetism as well. Sergei Borshch was talking on energetics of spin transition complexes.

Kyangwha Park demonstrated the power of DFT to calculate exchange coupling and magnetic anisotropy for manganese based single molecules. This talked made clear that one needs a proper understanding of both these magnetic properties, if one attempts to optimize molecular magnetic materials. Valerio Bellini focused on ab-initio studies of Cr-based single molecule materials, which have been also of interest by a contribution of Daria Tomecka.

Highlights day 4

The last day contained only invited talks. The importance of synthesis was nicely shown by Hiroki Oshio. Carlo Canali discussed Cern-Number spin Hamiltonians which may become important for the proper choice of Hamiltonian. Alexander Lichtenstein as a well known expert on techniques beyond DFT showed the imporatance of these extension for the calculation of exchange parameters. The final talk was given by Mark Pederson by a nice review of application of DFT to molecular magnetism.

Main conclusions

1. The Heisenberg Hamiltonian may not be always adequate for a proper description of magnetic properties of molecular materials.
2. In comparison experiment and theory one has to be very cautious, if one is comparing indeed the same.
3. DFT is can be accurate enough to calculate magnetic anisotropy in molecular magnetic materials. However, there are exceptions which require to go beyond standard DFT.
4. The calculation of magnetic exchange coupling for multi-nuclear systems is possible within DFT, but requires a proper chosen Hamiltonian.
5. Experimentalists are producing continously new exciting systems which poses new theoretical challenges for theory.
6. There is still a long way to go for a rational design of molecular materials with predetermined magnetic properties from first-principles.

Program

Day 1: July 18 2006

Session : 0 Welcome-Opening
08:50 to 09:00 : Welcome

Session : 1 Experiment & Theory I

09:00 to 09:45 : Presentation

Emerging trends in Molecular Nanomagnetism: The point of view of an experimentalist

Roberta Sessoli

09:45 to 10:30 : Presentation

Inelastic neutron scattering on Single Molecule Magnets and antiferromagnetic wheels

Hans-Ulrich Güdel

10:30 to 11:00 : Coffee Break

11:00 to 11:45 : Presentation

Magnetism in a relativistic perspective

Trond Saue

11:45 to 12:30 : Presentation

Computation of magnetic parameters by density functional theory: One-component perturbational vs. two-component variational inclusion of spin-orbit coupling.

Martin Kaupp

12:30 to 12:45 : Presentation

Calculation of Zero Field Splitting Parameters from First Principles

Jordi Cirera

12:45 to 13:00 : Presentation

A DFT study of the magnetic anisotropy of FeII cubes

Jordi Ribas

13:00 to 14:30 : Lunch Break

Session : 2 Session II

14:30 to 15:15 : Presentation

Stereochemistry and Spin Sates of Tetrahedral Building Blocks Revisited

Santiago Alvarez

15:15 to 16:00 : Presentation

Magnetism in low-dimensional systems: theoretical studies using a first-principles bottom-up methodology

Juan J. Novoa

16:00 to 16:30 : Coffee Break

16:30 to 17:15 : Presentation

Computing Exchange Couplings with Constrained Density Functional Theory

Troy Van Voorhis

17:15 to 18:00 : Presentation

Toward a variational treatment of the magnetic coupling between centers with elevated spin moments

Coen de Graaf

18:00 to 18:15 : Presentation

Contrasting direct versus mediated through-space magnetic interactions in pyridyl-verdazyl : hidroquinone molecular co-crystal

Joaquim Jornet

18:15 to 18:30 : Presentation

Magneto-structural correlations in polynuclear complexes: The Fe4 butterflies.

Thomas Cauchy

Day 2: July 19 2006

Session : 3 Session III

09:00 to 09:45 : Presentation

Is still the Heisenberg Spin Hamiltonian reliable for multi-nuclear paramagnetic clusters?

Federico Totti

09:45 to 10:30 : Presentation

Field-dependent magnetic parameters in Ni4Mo12: Magnetostriction at the molecular level?

Juergen Schnack

10:30 to 11:00 : Coffee Break

11:00 to 11:45 : Presentation

Exploring the interface between antiferromagnetism and metal-metal bonding

John McGrady

11:45 to 12:30 : Presentation

Theoretical studies of spin-crossover compounds

Mark Casida

12:30 to 12:45 : Presentation

Modeling of non-Heisenberg exchange interactions in tetrameric Ni(II) clusters of S4 symmetry
Nadeschda Kirchner

12:45 to 13:00 : Presentation
Cu-based metalorganic systems: an ab-initio study of the electronic structure
Andrea Salguero

13:00 to 14:30 : Lunch Break

Session : 4 Session IV
14:30 to 15:15 : Presentation
The magnetic properties of polyoxovanadates
Carmen Calzado

15:15 to 16:00 : Presentation
Modeling of the Magnetic Behavior of Low Dimensional Magnetic Systems
Marc Drillon

16:00 to 16:30 : Coffee Break

16:30 to 17:15 : Presentation
Competition between double exchange and purely magnetic Heisenberg models in mixed valence systems : application to half doped manganites
Nathalie Guihery

17:15 to 18:00 : Presentation
Non-Innocent Ligands in Organometallic Chemistry : Theoretical Aspects
Vincent Robert

18:00 to 18:15 : Presentation
Ab-inition study on a chain model of the Cr8 molecular magnet
Daria Tomecka

18:15 to 18:30 : Presentation
A first principles bottom-up theoretical study of the bis(2,3-dimethylpyridinium) tetrabromocuprate spin-ladder
Merce Deumal

Day 3: July 20 2006

Session : 5 Session V

09:00 to 09:45 : Presentation

Muon studies of molecular magnets

Stephen Blundell

09:45 to 10:30 : Presentation

Hybrid DFT functionals in the solid state

Furio Cora

10:30 to 11:00 : Coffee Break

11:00 to 11:45 : Presentation

DFT for the modeling of Photo-Magnetic Molecular Devices (PMMDs)

Ilaria Ciofini

11:45 to 12:30 : Presentation

Magnetic frustration, resolved by non-collinearity, in a Co-trinuclear triazin-based system

Andrei Postnikov

12:30 to 12:45 : Presentation

Molecular Dynamics of Antiferromagentically Spin-Coupled Systems

Nisanth Narayanan Nair

12:45 to 13:00 : Presentation

Energetics of binuclear spin transition complexes

Sergei Borshch

13:00 to 14:30 : Lunch Break

Session : 6 Session VI

14:30 to 15:15 : Presentation

Magnetic anisotropy and exchange coupling for manganese-based single molecule magnets

Kyungwha Park

15:15 to 16:00 : Presentation

Calculation and madel of the electronic g-matrix of transition metal and actinide ions.

Hélène BOLVIN

16:00 to 16:30 : Coffee Break

16:30 to 17:15 : Presentation

An ab-initio study of Cr-based single molecule magnets
Valerio Bellini

17:15 to 17:30 : Presentation
Strong antiferromagnetic coupling between orthogonal orbitals: an ab initio study of non-innocent ligand-based complex
Maria Carvajal

20:00 to : Dinner

Day 4: July 21 2006

Session : 0 Sesion VII
09:00 to 09:45 : Presentation
Facile syntheses of homo- and hetero-metal single molecule magnets
Hiroki Oshio

09:45 to 10:30 : Presentation
Chern-number spin Hamiltonians for magnetic clusters by ab-initio methods
Carlo M. Canali

10:30 to 11:00 : Coffee Break

11:00 to 11:45 : Presentation
Exchange calculations in molecular magnets beyond standard DFT
Alexander Lichtenstein

11:45 to 12:30 : Presentation
Applications of Density Functional Theory to Molecular Magnets
Mark Pederson

12:30 to 12:35 : Closing word

Abstracts

Facile syntheses of homo- and hetero-metal single molecule magnets

Hiroki Oshio
University of Tsukuba, Japan

Abstract

Single molecule magnets (SMMs) have attached an intense research interest due to their characteristic quantum phenomena and possible applications to quantum devices. There have been many homo-metal SMMs reported since the discovery of the first SMM, but the number of hetero-metal systems were still limited. Syntheses of hetero-metal complexes are not simple, and ligands with different coordination sits suitable for different metal ions are sometimes indispensable to prepare hetero-metal systems. During the course of synthetic effort, we found that simple Schiff bases with alkoxo-group assemble hetero-metal ions and form multi-nuclear complexes. We report here syntheses, structures, and magnetic properties of homo- and mixed-metal clusters, some of which were recognized as single molecule magnets.

Inelastic neutron scattering on Single Molecule Magnets and antiferromagnetic wheels

Hans-Ulrich Güdel
University of Bern, Switzerland

Abstract

Exchange and anisotropy interactions split the ground state of polynuclear molecular complexes of transition metal ions. The magnetic properties of such materials are governed by these splittings. Inelastic neutron scattering (INS) is a powerful tool for the direct spectroscopic determination of such splittings. Besides the energies, INS also provides important information about the wavefunctions. The principles shall be briefly explained and examples from recent research in our group used for illustration. Single Molecule Magnets(SMM), in particular the prototype SMM Mn12-acetate, shall be covered as well as antiferromagnetically coupled wheels.

References

G. Chaboussant, A. Sieber, S. Ochsenbein, H.-U. Güdel, M. Murrie, A. Honecker, N. Fukushima and B. Normand
Exchange interactions and high-energy spin states in Mn12-acetate
Phys. Rev. B 70, 104422/1-16 (2004)

O. Waldmann, C. Dobe, H. Mutka, A. Furrer and H. U. Güdel
Né el-Vector Tunneling in Antiferromagnetic Molecular Clusters
Phys. Rev. Lett. 95, 057202/1-4 (2005)

O. Waldmann, C. Dobe, S. T. Ochsenbein, H. U. Güdel and I. Sheikin
Field-Induced Magnetoelastic Instabilities in Antiferromagnetic Molecular Wheels
Phys. Rev. Lett. 96, 027206/1-4 (2006)

O. Waldmann, G. Carver, C. Dobe, D. Biner, A. Sieber, H. U. Güdel, H. Mutka, J. Ollivier and N. E. Chakov
Magnetic relaxation studies on a single-molecule magnet by time-resolved inelastic neutron scattering
Appl. Phys. Lett. 88, 042507/1-3 (2006)

Roland Bircher, Gré gory Chaboussant, Chris Dobe, Hans U. Güdel, Stefan T. Ochsenbein, Andreas Sieber and Oliver Waldmann
Single-Molecule Magnets Under Pressure
Adv. Funct. Mater. 16, 209-220 (2006)

Field-dependent magnetic parameters in Ni4Mo12: Magnetostriction at the molecular level?

Juergen Schnack
Uni Osnabrueck, Germany

Abstract

We investigate the bulk magnetic, electron paramagnetic resonance, and magneto-optical properties of Ni4Mo12, a magnetic molecule with antiferromagnetically coupled tetrahedral Ni ions of spin s=1 in a diamagnetic molybdenum matrix. The low-temperature magnetization exhibits steps at irregular field intervals, a result that cannot be explained using a Heisenberg model even if it is augmented by magnetic anisotropy and biquadratic terms. Allowing the exchange and anisotropy parameters to depend on the magnetic field provides the best fit to our data, suggesting that the molecular structure (and thus the interactions between spins) may be changing with applied magnetic field. Such a behavior could be denoted as Magnetostriction at the molecular level.

The presentation will focus on existing experimental and theoretical results as well as on forthcoming investigations.

Calculation and madel of the electronic g-matrix of transition metal and actinide ions.

Hé lène BOLVIN
CNRS, France

Abstract

Starting from the formula proposed by Gerloch and McMeeking in 1975, the electronic g-matrix is expressed as a sum of two matrices called Lambda and Sigma describing the orbital and spin contributions respectively. This approach is applied on benchmark diatomic and triatomic molecules and on TiF3 and [Cu(NH3)4]2+ using either CASPT2 or CCSD(T) methods to calculate the spin-free states and SO-RASSI to calculate spin-orbit coupling. Results compare very well to experimental data and to previous theoretical works and for each molecule, the anisotropy of the g-matrix is modelled by the mean of few parameters. This approach is then applied to a mixed valence compound, the Creutz-Taube molecule: numerical results are in good agreement with experiment and are fully interpreted in the frame of a model that shows that the g-matrix is independent of the electronic coupling between the two metallic atoms. Finally, the method is applied to two actinide complexes: [NpO2]2+ with equatorials ligands and NpF6 where again, our results are in accordance with experimental data and permits the full interpretation of the anisotropy.

Calculation of Zero Field Splitting Parameters from First Principles

Jordi Cirera

Universitat de Barcelona, Spain

Abstract

We have performed first-principles density functional calculations on some simple molecules with one metal center and different oxidation states and molecular environments, using the NRLMol code, which has been used succesfully to study several Single Moleculer Magnets, in order to calculate the second order magnetic anisotropy parameters. The theoretical results will be compared with the available experimental information from HF-EPR. The dependence of this method on different basis sets and DFT functionals will be discused. Using simple models we have studied the variation of the magnetic anisotropy with some typical strucutral distortion pathways for four-, five- and six coordinate complexes. The ability to predict computationally the magnetic anisotropy barrier should allow us to understand which factors modify the zero field splitting values, which is crucial for a rational design of single molecule magnets

AB-INITIO STUDY ON A CHAIN MODEL OF THE Cr8 MOLECULAR MAGNET

Daria M. Tomecka

Adam Mickiewicz University, Poznan, Poland

Abstract

In that contribution we present an ab-initio investigation by means of density functional theory (DFT) simulation techniques of the electronic and magnetic properties of the antiferromagnetic Cr8 molecular ring (i.e. [Cr8F8Piv16], where HPiv - pivalic acid, trimethyl acetic acid). Here the linearized augmented plane wave method (LAPW) (in the Wien2k package [1]) is used to calculate the electronic density of states, band structures and exchange couplings J of Cr8 [2] and that of a chain model system, which aim to represent the magnetic interactions in the Cr8 ring. The smaller size of the simulation cell of the model, as compared to the one of the original ring-like molecule, enables us to calculate the above mentioned properties more efficiently and systematically. By the thorough comparison between the model complex and the Cr8 ring, we prove that the chain model is reliable and mimics with good approximation the electronic and magnetic properties of Cr8. Moreover the model can be easily extended to systems like 'Ni doped' Cr rings or broken Cr segments, which have recently attracted much attention for their application in the field of quantum computing.

References

[1] P. Blaha, K. Schwarz, G. Madsen, D. Kvasnicka, and J. Luitz, WIEN2k, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties, (Karl-heinz Schwarz, Techn. Universitaet Wien, Austria) (1999), ISBN 3-9501031-1-2.

[2] V. Bellini, A. Olivieri and F. Manghi; Phys. Rev. B 73, 184431 (2006).

DFT for the modeling of Photo-Magnetic Molecular Devices (PMMDs)

Ilaria Ciofini

ENSCP Paris, France

Abstract

With the aim of designing new Photo-Magnetic Molecular Devices (PMMDs), able to display a target magnetic behavior at the ground and at the excited state, a series of systems containing transition metal ions (Os(II) and Ru(II)) have been investigated. To form target magnetic excited states we rely on photoinduced intramolecular electron transfers (ET) within covalently linked assemblies of active components usually referred to as polyad systems. Basically, the topology adopted for the novel photo-magnetic molecular devices derives from that of acceptor dyads (P− A) devised to produce charge separated states (CS) upon light excitation in the framework of researches devoted to artificial photosynthesis. These supermolecules are comprising the following functional elements: -P: the photosensitizer unit; here a Ru(II) or Os(II) complex. -A: an electron(s)-accepting unit which traps the electron promoted from *P -Spin carriers (Sc) surrounding A. The basic working principles of such devices are the following: upon light excitation, an electron will be promoted from P to A to form the target charge separated excited state [P+-A-]*. As a consequence the coupling between the Sc surrounding A will be modified inducing a different magnetic coupling at the excited state. In this contribution, we propose an analysis, based on DFT and TD-DFT, of the properties of existing dyads (P-A) as well as of new acceptor units specifically designed to obtain efficient PMMDs. Various mechanisms of coupling, as a function of the topology of the acceptor units, are also discussed

Magnetic frustration, resolved by non-collinearity, in a Co-trinuclear triazin-based system

Andrei Postnikov

Paul Verlaine University, Institute de Physique Electronique et Chimie, Laboratoire de Physique des Milieux Denses, France

Abstract

The electronic structure of [Co3(L)3(H2O)(OH)]HNEt3, where L stands for a dianion of the 2,6-bis(5-hydroxy-6-phenyl-1,2,4-triazin-3-yl)pyridine, is analyzed on the basis of first-principles calculations within the density functional theory (DFT), applying the computer code SIESTA [1]. The calculations are done for an isolated molecular fragment (147 atoms), using the generalized gradient approximation after Perdew-Burke-Ernzerhof for the exchange-correlation. Due to a predominantly antiferromagnetic interaction between Co ions, which are all connected to the central hydroxyl ion, the system is magnetically frustrated. The DFT calculation finds the ground state with two high-spin (s=3/2) Co ions set antiparallel and the third Co ion being in the low-spin state (s=1/2), yielding the net spin S=1/2. This finding was further refined by taking into account non-collinear (i.e. smoothly changing its direction in space from one point to another) variations of the magnetic density, which yielded all three Co local magnetic moments

of about 2.6 956;B, with different spatial orientation.

From considering different trial orientations of Co magnetic moments, the values of interatomic interaction parameters are estimated.

Further on, we considered the effect of the spin-orbit interaction (SOI), recently implemented in SIESTA in the on-site approximation [2]. In this approach the SOI and the non-collinearity of magnetic density are treated in a natural way on equal footing. According to our preliminary results, the high-spin configuration on all three Co atoms survives also in the presence of SOI.

AP gratefully acknowledges the cooperations within the priority program SPP 1137 by the Deutsche Forschungsgemeinschaft (DFG), discussions with S. Blü gel and J. Schnack, and the use of computational facilities provided by the DFG.

References

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Computing Exchange Couplings with Constrained Density Functional Theory

Troy Van Voorhis
MIT Boston, United States

Abstract

We demonstrate an accurate method for extracting Heisenberg exchange coupling constants from constrained DFT calculations. We note that the true Ising-like low-spin state of a given molecule can be identified with the ground state of the system subject to a constraint on the spin density of the atoms. Using an efficient optimization strategy for constrained DFT we obtain these states directly, leading to a simple, physically motivated formula for the exchange coupling. Preliminary applications to bi- and poly-nuclear transition metal complexes show that this C-DFT approach is competitive with, if not more accurate than, the best broken symmetry (BS) DFT techniques. In particular, we find that C-DFT gives a robust treatment of frustrated magnetic molecules, which have heretofore been problematic for DFT. Time permitting, we will also discuss how the exchange couplings obtained from either BS- or C-DFT can be incorporated into a simple model Hamiltonian that accounts for magnetic anisotropy.

An ab-initio study of Cr-based single molecule magnets

Valerio Bellini
CNR-INFM-S3, Modena, Italy

Abstract

Molecular nanomagnets, which show intriguing quantum and critical phenomena, like quantum tunneling of magnetization, have recently attracted much interest as next-generation nanoscale, monodisperse magnets for specialized applications. We are interested in molecular rings, a subclass of molecular magnets, composed of heterometallic nanostructures with a cyclic shape and an antiferromagnetic exchange coupling between nearest neighbouring magnetic ions. We have

characterized the electronic and magnetic properties of the ground state of Cr8 molecule, as well as investigated the role of doping, by substituting Cr(III) ions with divalent Ni(II) atoms in changing the ground state of such molecules. The tailoring of the ground and excited states of such substituted Cr rings has been pointed out to carry important applications in the field of quantum computing. The calculations have been performed by density functional theory, by means of the Wien2K package [1]. The exchange interaction constants J's, which have been extracted by mapping the ab-initio total energies onto an effective localized spin classical Hamiltonian, compare well with the values extracted by the experiments, on condition that the electronic correlations in the transition metal d orbitals is taken into account beyond the standard LDA or GGA approximations [2], e.g. by the LDA(GGA) +U method [3].

References

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[2] V. Bellini, A. Olivieri and F. Manghi, to appear in Phys. Rev. B (2006).
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Magnetism in a relativistic perspective

Trond Saue
CNRS UMR7177, France

Abstract

My talk will be divided in two parts:

1. In the first part I will discuss magnetism in a relativistic perspective. I will discuss the strict non-relativistic limit of electrodynamics and what can be learned from my conclusion. I will also discuss the separation of paramagnetic and diamagnetic contributions at the 4-component relativistic level, that is starting from the Dirac equation. It should be noted that when a magnetic field is introduced at this level only a linear and no quadratic term in the field appear in the Hamiltonian.

2. In the second part I will discuss more general aspects of 4-component relativistic molecular calculations and possible savings when going to the 2-component level. I will furthermore discuss the difference between the introduction of spin-orbit coupling in a perturbational and variational manner.

Some real applications will be shown for illustration.

Competition between double exchange and purely magnetic Heisenberg models in mixed valence systems : application to half doped manganites

Nathalie Guihery
Toulouse, France

Abstract

The low-energy spectrum of the Zener polaron in half-doped manganites studied by means of explicitely correlated ab initio calculations has been confronted to the spectrum of different model Hamiltonians. It will be shown that the electronic structure of the low-enery states results from a subtle interplay between double exchange configurations and Oxygen $2p_\sigma$ to Mn 3d charge transfer configurations that obey a Heisenberg logic. The purely magnetic Heisenberg model is analytically solved in the general case of two metals (having N magnetic orbitals) bridged by a magnetic oxygen. Unexpectedly the confrontation of the Heisenberg energies to those of the double exchange model reveals that the two spectra are analytically identical except for one state which does not belong to the model space of the double exchange Hamiltonian. Since the ab initio energy of this state fits perfectly with the predicted energy of the Heisenberg model, the comparison of the spectra does not provide any arguments to discriminate between the two models. In order to settle the question, a model which combines different antiferromagnetic contributions to the Zener Hamiltonian is used. It leads to a significant improvment in the reproduction of the low energy spectrum showing that the Zener polaron is ruled by a refined double exchange model in which the local excited non-Hund states play a non negligible role.

Chern-number spin Hamiltonians for magnetic clusters by ab-initio methods

Carlo M. Canali
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Abstract

Combining field-theory methods and ab-initio calculations, we construct an effective Hamiltonian with a single giant-spin degree of freedom, capable of describing the low-energy spin dynamics of ferromagnetic metal nanoclusters consisting of up to a few tens of atoms. In our procedure, the magnetic moment direction of the Kohn-Sham spin-density functional theory wave-function is constrained by means of a penalty functional, allowing us to explore the entire parameter space of directions, and extract the magnetic anisotropy energy and the Berry curvature functionals. The average of the Berry curvature over all magnetization directions is a so-called Chern number, a topological invariant that can only take on values equal to multiples of half-integers, which represents the dimension of the Hilbert space of the effective spin system[1]. The spin Hamiltonian is obtained by quantizing the classical anisotropy-energy functional, after a change of variables to a constant Berry-curvature space. We illustrate this procedure by explicitly constructing the Hamiltonian for dimers and trimers of transition-metal atoms, whose spin dynamics has been recently investigated experimentally by STM methods[2].

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Hybrid DFT functionals in the solid state

Furio Cora
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Abstract

The focus of this talk is the computational study of crystalline inorganic materials, and the accuracy with which their structural and electronic properties can be predicted. In particular, we shall examine functional materials, which are characterised by the presence of competing structural and electronic states, and/or of long-range ordered magnetic states separated by very small energy differences. Determining the details of the electronic and magnetic properties requires the application of techniques with sufficient accuracy and resolution. The computational characterisation of magnetic oxides requires the application of quantum mechanical (QM) techniques that consider explicitly the unpaired electrons at the origin of magnetism. The method of choice for electronic structure calculations on solids is Density Functional Theory (DFT); however, the standard formulations of DFT based on local and gradient-corrected functionals (LDA, GGA) fail in describing well localised electronic states, such as those of the d (f) electrons responsible for magnetism. This failure is related to the selfinteraction error, so called because the DFT Hamiltonian contains a spurious interaction of each electron with its own electronic density. The computational study of magnetic solids requires techniques that go beyond local DFT. One possible approach is represented by the hybrid exchange functionals, and consists in mixing the DFT exchange with the corresponding definition in the Hartree-Fock (HF) theory. The HF exchange corrects for the self-interaction error of the local DFT functionals; however, the amount of HF exchange to include in the hybrid functional should be treated as parametric, and determined by comparison to known experimental observables. We have recently investigated systematically the accuracy that is obtained in describing the electronic properties of transition metal oxides, including rocksalt-structured materials, ferroelectric and ferromagnetic perovskites, and other ternary and more complex compounds. By analysing results, we shall show that not only the localisation of the electronic states, and hence the magnetic coupling in the solid, is very sensitive to the amount of HF exchange employed in hybrid DFT studies, but also that the equilibrium structures depend on the functional. In ternary and more complex materials, the self-interaction influences to a different extent bonds with a different ionicity, resulting in non-uniform structural errors that may hide important structural and electronic distortions. The availability of reliable solid-state functionals, able to reproduce at the same time the electronic and structural properties of solids is an important result, because it makes the computational work independent from knowledge a priori of experimental geometries. This feature is particularly desirable for cases in which modelling has to be employed predictively, for instance in the design of new compositions, or in the investigation of defect chemistry, and how this modifies the electronic properties. Examples taken from recent works will include magnetic oxides, but also a number of non magnetic compounds, added to illustrate important structural trends. Finally, hybrid functionals will be compared with the results of GGA+U calculations, now commonly used in solid-state physics.

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Emerging trends in Molecular Nanomagnetism: The point of view of an experimentalist

Roberta Sessoli
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Abstract

In the field of molecular magnetism theory has kept pace with experiments, sometime anticipating them as in the case of the phase interference in the tunnel pathways for the reversal of the magnetization. Novel materials comprising lanthanide metal ions or linear arrays of slow relaxing units appear as promising candidate to increase the blocking temperature. Moreover , in the generalized trend towards nanoscale enegineered structures, synthetic chemists and experimentalists in molecular magnetism have started to address the individual molecules by organizing them on surfaces or putting them between electrodes. This new research field, undoubtfully promising for applications like spintronics, requires however a synergic effort between experimentalists and theoreticians in order to understand how the interaction with the substrate affects the magnetic properties and vice versa. Preliminary results and open questions we have encountered in our recent research in this field will be discussed.

Stereochemistry and Spin Sates of Tetrahedral Building Blocks Revisited

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Abstract

Now that a great deal of control can be exerted on the exchange interaction between paramagnetic centers in large polynuclear systems, it is timely to look back at the stereochemical and spin state preferences of the basic building blocks.[1] We have done that for the case of tetrahedral transition metal complexes with a qualitative theoretical perspective supported by extensive shape analysis of experimental structural data and density functional calculations on thoughtfully designed model complexes covering all the dn electron configurations (0 8804; n 8804; 10). The results to be presented show an excellent match between the calculated potential energy surfaces and the frequency of experimental structures, when both are plotted as a function of the generalized minimum distortion interconversion coordinate between the tetrahedral and square planar geometries.[2] The importance of several factors in determining the stereo-

chemistry and ground spin state in each particular case will be evaluated, including the number of valence d electrons, the presence of 960;-donor ligands, the metal oxidation state and the row of the periodic table to which the metal belongs.

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Computation of magnetic parameters by density functional theory: One-component perturbational vs. two-component variational inclusion of spin-orbit coupling.

Martin Kaupp
Univ. Wuerzburg, Germany

Abstract

Computation of magnetic parameters by density functional theory: One-component perturbational vs. two-component variational inclusion of spin-orbit coupling.

Martin Kaupp

Universitä t Wü rzburg

Spin-orbit coupling is frequently of substantial importance for magnetic properties of molecules, sometimes the by far dominant contribution, as for g-tensors or zero-field splittings. When using quantum chemical calculations to compute these or related magnetic resonance parameters, one has in principle two choices when treating spin-orbit coupling: One may introduce it as a perturbation (typically in leading order, for example within the framework of the Breit-Pauli Hamiltonian) on top of a one-component nonrelativistic or scalar relativistic wavefunction. Or one may introduce it variationally into the wavefunction within a relativistic framework (four- or two-component). Within the ReSpect code developed jointly in Wü rzburg and Bratislava, this is done in a non-collinear spin-density two-component Douglas-Kroll-Hess DFT approach. In this talk, the insight provided by comparing these treatments is discussed for a variety of magnetic parameters, including zero-field splittings and g-tensors. Recent developments of computing NMR chemical shifts for paramagnetic systems will also be reported. Here a combination of NMR experiment and theory offers substantial potential to map spin-density distributions of building blocks in magnetic materials, e.g. of prussian blue type.

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Non-Innocent Ligands in Organometallic Chemistry : Theoretical Aspects

Vincent Robert
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Abstract

A variety of intriguing ligands which may display different oxidation states when coordinated to metallic centers has been under intense investigation over the last decade. The possibility to generate radical ligands in coordination compounds has given rise to intense investigation since interesting magnetic materials can be anticipated. Magnetic properties are split between a metallic center and a so-called non-innocent ligand . In particular, excited state coordination chemistry (K. Wieghardt et al.) has become a tremendously important field of research. Correlated ab initio calculations have been performed to look into the low-energy spectrum of one particular system. The electronic structure of different systems will be discussed and compared to available experimental data and previous DFT calculations. Such approaches are very powerful to elucidate the actual charge organization and to detail spectrum characteristics.

Is still the Heisenberg Spin Hamiltonian reliable for multi-nuclear paramagnetic clusters?

Federico Totti

Uni Florence, Italy

Abstract

Alessandro Bencini and Federico Totti

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Attempts to prove the validity or not of the Heisenberg Spin Hamiltonian when two or more paramagnetic spin centers are considered have been recently done. Here we want to report a detailed study on model systems with paramagnetic centers with a local spin ⟩ and with nuclearity ranging from 2 to 4. The role played by spin delocatisation will be also discussed. Moreover, the advantages and the drawbacks of the use of the Broken Symmetry approach in multi-centered paramagnetic clusters will be covered.

Toward a variational treatment of the magnetic coupling between centers with elevated spin moments

Coen de Graaf

Univ. Rovira-Virgili, Spain

Abstract

A multireference configuration interaction scheme is presented to calculate electronic structure parameters for systems with an elevated number of unpaired electrons. The presentation concentrates on the magnetic coupling but the method will also be applied to other parameters such as the on-site repulsion energy and hopping parameters. The reference wave function contains not only the usual configurations contained in the Anderson model but is extended with ligand-to-metal charge transfer configurations. Subsequently a small subset of the complete difference dedicated configuration interaction space is included in the calculation. Different strategies to introduce the charge transfer configurations in the reference wave function are compared. Projected model ligand vectors ensure the optimal inclusion of the charge transfer effects and good agreement with more extensive calculations is obtained at a more reduced computational cost.

Energetics of binuclear spin transition complexes

Sergei Borshch

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Theoretical studies of spin-crossover compounds

Mark Casida

Univ. of Grenoble , France

Abstract

A knowledge of the spin state of a molecule is often of fundamental importance for understanding its physical properties and chemical reactivity. This is certainly the case for 3d transition spin crossover complexes. We are primarily interested in the spin states of Fe(II) complexes where the size of the compounds of experimental interest and the presence of a transition metal makes densityfunctional theory (DFT) the most promising method for the quantitative description of differences between the high spin (HS) and low spin (LS) states. Our comparison of functionals with ab initio calculations on small complexes [1,2] and with experiment [3,4] show that the ability of DFT to correctly describe the spin pairing energy is very much functionaldependent but that the right generalized gradient approximation (GGA) can give results, which still imperfect, compare favorably with what can be expected from ab initio calculations on small Fe(II) clusters.

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Exploring the interface between antiferromagnetism and metal-metal bonding

John McGrady

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Abstract

Metal clusters are traditionally divided into two distinct categories, one where direct covalent bonds link the metal centres, and the other where indirect exchange interactions dominate. In this presentation, I will describe the remarkable electronic structure of the linear tricobalt chain compound, Co3(dpa)4Cl2, which spans these two extreme limits. The cluster exists into two distinct forms, one symmetric, the other with very different Co-Co bond lengths.1 The structural and magnetic properties can only be fully rationalised using 3 distinct electronic states, two doublets and a quartet, all of which are populated under different conditions (temperature, solvent of crystallisation). One of the two doublet states is an 'open-shell' species containing a high-spin (S = 3/2) CoII centre, antiferromagnetically coupled to a Co-Co dimer unit in a locally excited triplet state. The relaxation of this unusual electron distribution occurs along a very flat potential energy surface gives rise to the remarkable temperature dependence of the unsymmetric form of Co3(dpa)4Cl2.2

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Magnetic anisotropy and exchange coupling for manganese-based single molecule magnets

Kyungwha Park
Virginia Tech, United States

Abstract

Molecular magnets are large, well-defined, discrete molecules consisting of several transition metal ions interacting through organic and/or inorganic ligands. Among thousands of synthesized molecular magnets, there is a class of molecular magnets known as single-molecule magnets (SMMs) which have large effective energy barriers between different directions of the magnetic moment and behave as single-domain magnetic nanoparticles in an external magnetic field. They are particularly interesting because of observed quantum tunneling of magnetization and their possible applications in magnetic recording and molecular electronics. In this talk, I will present density-functional calculations of the electronic, vibrational, and magnetic properties of selected manganese-based SMMs, such as the total magnetic moment, electronic energy gaps, Raman scattering spectra, exchange constants, spin excitation energetics, and magnetic anisotropy barriers. I will also discuss what types of molecular environmental changes can significantly influence the exchange interaction, magnetic anisotropy, and observed quantum tunneling in the SMMs. [This work has been done in collaboration with Mark Pederson, Tunna Baruah, Noam Bernstein, Stephen Hellberg, Steven Richardson, N. Aliaga-Alcalde, and George Christou.]

Muon studies of molecular magnets

Stephen Blundell

Oxford, United Kingdom

Abstract

I will describe recent results on molecular magnets studied using the technique of muon-spin rotation. These studies include work on Cu-chain systems, single molecule magnets and other low-dimensional molecular magnets.

Modeling of non-Heisenberg exchange interactions in tetrameric Ni(II) clusters of S4 symmetry

Nadeschda Kirchner
Universitä t Stuttgart, Germany

Abstract

A new developement of the generalized effective spin hamiltonian formalism will be presented: antisymmetric exchange interactions and local crystal fields were introduced into the Hamiltonian for the first time as non-collinear tensors. Application of the model for simulation of the temperature dependence of dc susceptibility, magnetization and INS-measurements on Ni(II)tetrameric cluster of S4 symmetry will be shown.

The magnetic properties of polyoxovanadates

Carmen Calzado
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Abstract

Polyoxovanadates containing quasi spherical $V_{18}O_{42}$ shells present an interesting magnetic behaviour: the antiferromagnetic coupling increases with the oxidation degree of the shell (1). While structural effects induced by the change on the electronic populations have been discarded, it is not clear the role that electronic effects could play on their magnetic properties.

We have studied the dependence of the magnetic properties of these systems on their oxidation state by means of a computational strategy combining ab initio calculations and the diagonalization of a model t-J-V Hamiltonian. From ab initio quantum chemistry calculations we have estimated the amplitudes of the nearest-neighbour (NN) and next-nearest-neighbour (NNN) magnetic coupling constants and hopping integrals. These values have been injected onto a t-J-V model Hamiltonian to evaluate the susceptibility at different temperatures.

The results to be presented show that (i) the NN and NNN coupling constants have similar values, despite larger V-V distances (ii) the hole doping induces a general increase of the AF coupling, which could be related to the observed magnetic behaviour, (iii) the interaction betweeen magnetic coupling and electron delocalization seems to be in the origin of the enhancement of the antiferromagnetic coupling.

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Magnetism in low-dimensional systems: theoretical studies using a first-principles bottom-up methodology

Juan J. Novoa
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Abstract

The nature of the magnetic interactions are evaluated and the macroscopic magnetic properties are computed for a series of 1D and 2D low-dimensional crystals using a recently introduced first-principles bottom-up methodology. This methodology uses first-principles methods to compute the microscopic JAB radical-radical interactions. Then, using these JAB values the macroscopic magnetic properties are computed by diagonalization of the matriz representation of the Heisenberg Hamiltonian (in a finite space that properly reproduces the magnetic topology of the infinite crystal). The procedure allows to connect the computed macroscopic results with their microscopic origins. We have used this procedure in the study of isolated chains, spin-ladders, and 2D-planes, to test the quality of the first-principles bottom-up procedure on these systems. An excellent agreement is found between the computed and experimental results.

Magneto-structural correlations in polynuclear complexes: The Fe4 butterflies.

Thomas Cauchy
Universitat de Barcelona, Spain

A first principles bottom-up theoretical study of the bis(2,3-dimethylpyridinium) tetrabromocuprate spin-ladder

Merce Deumal
Universitat de Barcelona, Spain

Abstract

The bis(2,3-dimethylpyridinium) tetrabromocuprate (2,3dmpyH)2CuBr4 spin-ladder has been studied using a first principles bottom-up theoretical procedure [1], which rigorously computes the macroscopic magnetic properties from the only knowledge of the JAB microscopic magnetic interactions. Spin-ladders have been a special topic of interest due to the presence of an energy gap in their spin excitation spectrum. Moreover, under certain conditions, ladders have been shown to undergo a transition to a superconducting phase. The JAB magnetic interactions responsible for the magnetism of (2,3dmpyH)2CuBr4 have been analyzed and evaluated. The magnetic topology of (2,3dmpyH)2CuBr4 results to be a set of strong-rail exchange spin-ladders (Jrail = − 7.8 cm-1, Jrung = − 3.5 cm-1). The 2,3dmpyH+ (2,3-dimethylpyridinium) counterions are shown to be important when computing the value of the JAB microscopic magnetic interactions. The singlet-triplet spin-gap has been computed, and agrees with the available experimental data. Two crystal structures of (2,3dmpyH)2CuBr4 determined at 88K and 273K have been used to show the effect of the temperature on the magnetic topology and susceptibil-

ity data. It will be shown that at 88K the numerically computed magnetic susceptibility data properly reproduces the experimental magnetic curve for (2,3dmpyH)2CuBr4 magnet.

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Contrasting direct versus mediated through-space magnetic interactions in pyridyl-verdazyl : hidroquinone molecular co-crystal;

Joaquim Jornet
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Abstract

The magnetism in non-purely organic molecule-based magnets is mostly explained in terms of through-bond metalligandmetal superexchange interactions, where two spin-containing metals are connected by means of the orbitals of a diamagnetic ligand. Similarly, the mechanism of the magnetic interactions in the pyridyl-verdazyl:hidroquinone (pyvd:hq) molecular co-crystal was suggested to originate by a unique “mediated through-space” magnetic interaction. This interaction was proposed to magnetically connect two non-adjacent pyvd radicals within a p stack, where adjacent radicals pile up in a head-over-tail orientation, through a third radical sitting between the previous two mediated radicals. Given the relevance of this proposal, the magnetic properties of pyvd:hq co-crystal were reinvestigated using a first-principles bottom-up theoretical procedure [1,2]. The microscopic JAB values for all direct through-space magnetic interactions between nearby pyvd radicals were computed. There are two non-negligible magnetic interactions of similar strength (– 56 and – 54 cm-1) corresponding to the direct through-space interactions between two adjacent radicals of a p stack. The co-crystal also exhibits a radical-mediated through-space interaction of – 0.31 cm-1 between two non-adjacent radicals of a p stack. Direct through-space magnetic interactions are two orders of magnitude larger than the mediated through-space interaction. Thus, first-principles calculations do not support a mediated through-space mechanism to explain the magnetism of the pyvd:hq co-crystal. The magnetic topology generated by the two dominant antiferromagnetic interactions in pyvd:hq consists of one-dimensional alternating chains (interacting very weakly along the b and c axes). By using this topology, the computed macroscopic magnetic susceptibility values properly reproduce the experimental data. ;

Magneto-structural correlations in the [*Mn(porphyrin)*][*TCNE*] *family of molecule-based magnets;*

Jordi Ribas
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Abstract

The dependence of the JAB interaction with the geometry in the meso-tetraphenylporphyrinato-manganese(III) tetracyanoethenide family of molecule-based magnets has been re-evaluated by performing B3LYP calculations. The calibration of the B3LYP results with the experimental

data of seven [Mn(porphyrin)][TCNE] representative systems and with CASSCF and CASPT2 calculations, shows that the B3LYP method is the most cost/quality efficient form of evaluating the magneto-structural correlations in these systems. After this calibration, the JAB was systematically evaluated as a function of the intermolecular coordinates that define the position and orientation of [TCNE].- relative to the [Mn(porphyrin)]+. The JAB have been found to be antiferromagnetic in all regions, with the largest values being associated to the smallest angles between the [Mn(porphyrin)]+ and the TCNE planes. On the other hand, the B3LYP results have been compared to those obtained using the three spin exchange me chanisms applicable to these systems (i.e. orthogonality, spin coupling, and CI mechanisms). None of these three mechanisms gives trends in fully agreement with the B3LYP results. Therefore, one has to resort to results from first principles methods. ;

Applications of Density Functional Theory to Molecular Magnets

Mark Pederson
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Abstract

In addition to summarizing some of the density-functional theory and methods that are now widely used for calculations on molecular magnets, I will summarize the results of DFT-based applications to anisotropic molecular magnets (AMM) and to molecular spin-systems (MSS). The idealization of the former class of molecules, exemplified by Mn12-Acetate, is that the exchange interactions are very large. Within this idealization the lowest-energy excitations and consequently resonant tunneling of magnetization are due to second-order spin-orbit effects which can be calculated using perturbation theory on a single configuration of the system. Even within the ansatz of a density-functional-theory or single-configuration picture, additional effects due to self-consistency exist. Recent work aimed at further justifying a second-order approach and in identifying additional corrections will be presented. At the opposite extreme of AMM are the MSS. For these systems, such as the V15 cluster, the low energy excitations are due to many-spin excitations that can be extracted from density-functional-based determination of Heisenberg Hamiltonians followed by exact diagonalization. DFT-based calculations can be used to determine whether a given molecule containing transition-metal ions fits within one of these idealizations and for determining the magnetic excitations for either idealized case. A discussion of the challenges for understanding intermediate cases within DFT will be presented. I will also discuss how magnetic anisotropy barriers or a spin excitation depend upon environmental factors such as addition of charges, electric fields, or vibrational excitation and show that these factors can be accounted for within a density functional framework.

I thank S.N. Khanna, J. Kortus, T. Baruah, K. Park, N. Bernstein, S. Hellberg, and S. Richardson for interesting discussions and collaborations on many different aspects of molecular magnets during the last decade.

Modeling of the Magnetic Behavior of Low Dimensional Magnetic Systems

Marc Drillon
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Abstract

The use of adapted theoretical tools for modeling the thermodynamical functions of magnetic solids is of prime importance for an accurate analysis of their experimental behaviors allowing the establishment of detailed structure-properties relationships. A model of "hierarchical superparamagnetism" was developed recently which generalizes the idea of scaling by taking advantage of the non-singular solutions that are introduced, together with the singular ones, when the hypotheses of "critical scaling" are formulated. These non-singular solutions, although they have the same legitimacy, have simply been set aside when the goal was to describe the singularities of phase transitions. They happen to be very useful when correlations exist, but which are not sufficient to trigger a long range order at a finite TC, either because frustration is strong, near e.g. an antiferromagnetic (AF) order, or because we sit at, or below, a lower critical dimensionality. Model systems, such as the 1D or 2D-Heisenberg systems of spin S=1/2, 1,. . . 8734; display such behaviors. For this reason, much effort has been devoted in performing exact calculations on such finite systems of increasing size, and in trying to infer which type of limit is reached when the size diverges. On another hand, the progresses of chemistry have made it possible to design organometallic clusters, chains or planes, of axial, planar or isotropic spins, which are close approximants of the above mentioned systems, and are well appropriate to investigate the properties of interest. We will show on a few examples, that the model of hierarchical superparamagnetism provides the right framework to approach these problems and suggests a strategy adapted to each case.

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Molecular Dynamics of Antiferromagentically Spin-Coupled Systems

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Abstract

Proteins with iron-sulfur clusters of the type Fe1-4S08722;4(SR)2-4 are important electron transfer agents in living cells from bacteria to higher animals. These clusters are characterized by complex spin coupling between the iron atoms.

The exact electronic groundstate of such iron-sulfur clusters cannot be accessed from conventional single determinant density functional theory. We propose a generalized approach for determining the electronic ground state structure of binuclear spin-coupled systems. A Car-Parrinello Lagrangian is then formulated in this frame work for studying their dynamics. Further, this method is extended to a mixed QM-MM coupled scheme, which allows the treatment of [2Fe-2S]

proteins fully in our calculations, including the effect of its environment. Initial results from our ab initio molecular dynamics simulations are presented.

Cu-based metalorganic systems: an ab-initio study of the electronic structure

Andrea Salguero
Frankfurt University, Germany

Abstract

Within a first principles framework, we study the electronic structure of the recently synthesized polymeric coordination compound Cu(II)-2,5-bis(pyrazol-1-yl)-1,4-dihydroxybenzene (CuCCP), which has been suggested to be a good realization of an antiferromagnetic Heisenberg spin-1/2 chain. By using a combination of classical with ab-initio quantum mechanical methods, we design on the computer reliable modified structures of CuCCP aimed at studying effects of Cu-Cu coupling strength variations and dimensional crossover on this spin-1/2 system. For this purpose, we performed two types of modifications on CuCCP. In one case, we replaced H in the linker by i) an electron donating group (NH_2) and ii) an electron withdrawing group (CN), while the other modification consisted on adding H_2O and NH_3 molecules in the structure which change the local coordination of the Cu(II) ions. With the downfolding method within the framework of N-th order muffin-tin orbital technique (NMTO-downfolding) we provide a quantitative analysis of the modified electronic structure and the nature of the Cu-Cu interaction paths in these new structures and discuss implications for the underlying microscopic model.

Strong antiferromagnetic coupling between orthogonal orbitals: an ab initio study of non-innocent ligand-based complex

Maria Carvajal
ENS Lyon, France

Abstract

Structural magnetic information upon Fe(bpb)(CN)2, bpb- = 1,2-bis(pyridine-2-carboxamido) benzoate complex is obtained by means of density functional theory and correlated ab initio calculations. It is shown that strong antiferromagnetism is achieved between an iron(III) ion and a flat non-innocent bpb2- ligand, the singlet state lying 600cm-1 lower in energy than the triplet. Even thought magnetic orbitals are orthogonal, antiferromagnetism goes through the dynamical polarization of the ionic valence bond forms between metallic center and organic ligand part. The intrinsically delocalized nature of the magnetic orbital of the bpb ligand and its redox versatile character clearly enhance antiferromagnetism.

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Exchange calculations in molecular magnets beyond standard DFT

Alexander Lichtenstein
Uni Hamburg, Germany

Abstract

I will present calculations of exchange parameters in molecular magnets using methody beyond standard DFT in order to treat correlations more accurate (LDA+U, DMFT).

Contribution of correlated ab initio calculations to the description of magnetic behaviors in Prussian blue analogues;

Boris Le Guennic
Ecole Normale Superieure de Lyon, France

Exchange calculations in molecular magnets beyond standard DFT

Alexander Lichtenstein
Uni Hamburg, Germany

Abstract

Realistic approach to the electronic structure of complex oxides which contains correlated d-electrons will be discussed. The density functional theory within the local spin density approximation have been highly successful for electronic structure calculations and zero temperature magnetic properties of non-correlated systems. We investigate some failures of the LDA-scheme for the charge, spin and orbital ordering in transition metal compounds. General formulation of the LDA+U method which takes into account local Coulomb correlations for the d-shell of transition metals ions in the crystal within the mean-field approximation will be presented. The LDA+U scheme describe well the antiferromagnetic Mott insulators and gives reasonable values of superexchange interactions. Electronic structure, spin and orbital moments and lattice distortions of transition-metal compounds can be analyzed in the framework of rotationally invariant LDA+U method. We discuss the result of LDA+U investigations of electronic structure and exchange interactions of different molecular magnets.

List of participants

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3 Psi-k Training

3.1 Psi-k Training Graduate School

Bristol University, U. K.

25-31 March 2007

Description:

This Graduate School is an activity of the Psi-k Training Programme in the EU-funded 'Series of Events' Marie Curie Conferences and Training Courses. The other two activities are Hands-on Tutorials and Summer Schools.

The combined theory-hands-on Graduate School on electronic structure methods will take place at Burwalls and the Physics Department in Bristol from Sunday March 25 until Saturday March 31, 2007. There will be 20 lectures over 6 days, 10 lectures on theory and 10 lectures on electronic structure methods. The rest is all hands-on experience with relevant codes. The 10 lectures on theory are:

a.. 3 on Density Functional Theory (DFT) by E.K.U. Gross
b.. 3 on many body perturbation theory (MBT) by R.W. Godby
c.. 3 on dynamical mean field theory (DMFT) by M. Katsnelson
d.. 1 on the Bethe-Salpether equation (BSE) by F. Sottile.

The 10 lectures on methods we will give a bird's eye view of 5 methods:

a.. the plane-wave methods (PW) by X. Gonze
b.. the full potential local orbital method (FPLO) by M. Richter

c.. the linear augmented plane waves (LAPW) by P. Blaha
d.. the linearized muffin tin orbital method (LMTO) by O.K. Andersen
e.. the Korringa-Kohn-Rostocker (KKR) method by H. Ebert.

Each method is introduced by two lectures followed by hands on experience with a modern state of the art code.

Scientific Objectives:

The Bristol University Graduate School is aimed towards giving masters and beginning PhD students

a.. an understanding of the theories underlying electronic structure calculations;
b.. hands-on experience in electronic structure calculation codes;
c.. a perspective how these methods are used in state of the art research.

Practical Information:

Registration Deadline: March 01 2007

Location: Bristol University, UK

Fees: The workshop fee of EUR 250 is payable in advance. A limited number of scholarships will be available. Accommodation costs, breakfast and evening meal and travel reimbursements will be paid to qualifying participants.

To apply please register on the web-site:

http://www.mc-psi-k-training.cecam.org/

3.2 Summer School: Ab-initio Many-Body Theory

Organisers
A. Rubio, A. Lichtenstein, C. Filippi, A. Georges, and W. M. Temmerman

22-29 July 2007 (one week)

Palacio Magdalena, San Sebastian, Spain

For registration please go to:
http://www.mc-psi-k-training.cecam.org/index.php?content=activities/tutorial

Summary:

The School is a shared activity of the three working groups, WG1 (GW method), WG2 (quantum Monte Carlo) and WG3 (dynamical mean field theory). It aims at giving a unified overview of the three many-body approaches.

The School is the first Summer School in the Psi-k Marie Curie Series of Events (Psi-k Training in Computational Nanoscience). These Schools are complementary to the Hands-on tutorials offered within the same Programme and aim at providing understanding of the physical phenomena and materials in addition to the computational techinques which are then covered in the hands-on tutorials.

The School will reach out to young researchers already working in the general field of many-body ab-initio theory, as well as researchers who are not yet practitioners but are interested in acquiring the necessary expertise. The School will give a solid introduction to the description of electronic correlation effects on an ab-initio basis in both weakly and strongly correlated materials. It will provide the necessary background and context, highlighting the most challenging problems, and the topics of current interest. Finally, the program will try to achieve a good balance between theoretical and experimental lectures.

Proposed Program:

The program will consist of 6 hours of lectures per day. Ample time will be given to informal discussions and interaction between students and teachers, including round tables addressing questions by the participants as well as highlighting new frontiers in the field.

The list of topics are:

Basic density functional theory (DFT)

Time-dependent DFT New experimental techniques for excitation spectrum of materials

Many-body theory in a nutshell and MBPT and Hedin equation (GW)

Introduction to the physics of correlated materials - theory and experiments: Mott transitions, Hubbard bands, Kondo systems, f-electron physics, Basics of Quantum Monte Carlo and Quantum chemical methods

Introduction to DMFT and LDA+DMFT scheme

Application of DFT and TDDFT to complex systems GW methods and applications

f-electrons materials: SIC

Applications of QMC and Quantum chemical methods for real materials

DMFT impurity solver and practice of LDA+DMFT

Cluster extension of DMFT and LDA+CDMFT scheme

New applications of QMC methods

Combining the best of different worlds: MBPT-GW-DMFT and TDDFT

Invited Lecturers

S. Biermann (Paris)
K. Burke (Rutgers)
M. Foulkes (Imperial College London)
E.K.U. Gross (Berlin)
J. Grossman (Berkely)
F. Himpsel (Wisconsin)
M. Katsnelson (Nijmegen)
G. Kotliar (Rutgers)
F. Lechermann (Paris)
S.G. Louie (Berkeley) and R.W. Godby (York)
L. Mitas (North Carolina)
O. Parcollet (Paris)
F. Sottile/L. Reining (Paris) and R. van Leeuwen (Groningen)
A. Santander (Paris)
G.A. Sawatzky (Vancouver)
S. Sorella (SISSA)
A. Svane (Aarhus)
M. Troyer (Zurich)
D. Vollhardt (Augsburg)

Recognition of the course:

We will use the European Credit Transfer system to express the work load of a given training activity: One European Credit (EC) corresponds to a work load of 25-30 hours including preparation. Therefore, a certificate will be given to all PhD and master students that have successfully participated in one of the training activities. The total ECTs for the present activity is 2.

Participants:

The call for participation will be mainly directed to students and scientists specialized on computational physics and quantum chemistry. We will limit the number of participants to 70, in order to ensure a maximum interaction between all the participants and the lectures.

Applications/Support:

All persons who wish to participate should fill out the application form at including the request for finantial support. The list of students that get support and those accepted will be automatically notified through the web page of the Marie Curie Series of Events program: Psi-k Training in Computational Nanoscience (http://www.mc-psi-k-training.cecam.org/)

4 General Workshop/Conference Announcements

4.1 Quantum Monte Carlo and the CASINO program II

Sun 29th July - Sun 5th August 2007

The Towler Institute, 22 via del Collegio,

Vallico Sotto, Tuscany, Italy

WEB PAGE : `www.tcm.phy.cam.ac.uk/~mdt26/tti/tti.html`

POSTER: `www.tcm.phy.cam.ac.uk/~mdt26/tti/qmcatcp_07/tti.jpg`

The second international summer school in the series *Quantum Monte Carlo and the CASINO program* will take place in July/August 2007 at the Towler Institute monastery in the Tuscan Apuan Alps, organized and hosted by members of Cambridge University physics department's Theory of Condensed Matter (TCM) Group. The aim of the school is to give students a thorough introduction to quantum Monte Carlo as a method for performing highly accurate calculations of the electronic structure of real materials. The course is designed for young scientists who have no previous experience with this technique, though anyone interested is welcome to take part.

Many-body quantum theory provides the key ideas for understanding the behaviour of materials at the level of the electrons which bind the atoms together. Describing the complex behaviour of materials at the atomic level requires a sophisticated description of the correlated motion of the electrons, which can be achieved using *ab initio* computational methods. Quantum Monte Carlo in particular has many attractive features in this regard. It is an explicitly many-body method which takes electron correlation into account from the outset, and is capable of giving highly accurate results while at the same time exhibiting a very favourable scaling of computational cost with system size. Over the last sixteen years, the Cambridge group have been researching QMC methods and we have created a powerful, general computer program - CASINO - to carry out the calculations. The school will focus both on the basic theory of QMC and on more advanced practical techniques, and will include a thorough introduction to the CASINO program. A background in density functional theory or similar is normally thought to be useful.

The monastery is a delightful 16th century building incorporating an ancient church, and is situated in the isolated but spectacular setting of the Tuscan mountain village of Vallico Sotto. It is fully equipped with state of the art presentation and computer technology. All accommodation is on-site. There is a strictly-enforced policy for all events at the Institute that considerable time should be set aside for mountain hikes, caving, canyon walking, swimming in thermal baths, eating in wonderful local restaurants, visits to classical concerts in Lucca and Barga, and similar activities. It is to be hoped that the visitor will not mind this too much.

Instructors will include the main authors of the CASINO program, (Prof. Richard Needs, Dr. Mike Towler, Dr. Neil Drummond, and Dr. Pablo Lopez Rios) and very probably others.

The cost of the school for one week including accommodation will be around 400 GBP. Participants would need to arrange a flight to Pisa airport from where onward transportation will be arranged.

Interested students should contact Mike Towler (mdt26 at cam.ac.uk) for registration and further details.

Nice emails from students attending "QMC and the CASINO program" in 2006

"*Just a brief note to thank you again for hosting the QMC summer school. I had a wonderful time in Tuscany, it was probably the best conference/school I've attended. The science was excellent, and the location, obviously, stunning. The Towler Institute is a very impressive place. Every one of the participants was a pleasure to spend time with, and I've come away with some friends with whom I intend to keep in contact.*"

"*Thank you so much, it was a great pleasure being at the school. The presentations were all very good and the place and people really nice.*"

"*Thanks for all, the school was really great, helpful and very well organized.*"

"*I will advice your school to anyone on the earth*"

DETAILS AND PHOTOS FROM LAST YEARS EVENT ACCESSIBLE ON TTI WEB PAGE.

TTI CURRENTLY TAKING BOOKINGS FOR THE HOSTING OF CONFERENCES, SCHOOLS AND GROUP MEETINGS IN EASTER/SUMMER 2007. Enquiries to mdt26 at cam.ac.uk .

4.2 37th Spring School 2007

Research Center Juelich, Germany

March 12-23, 2007

This series of two-week Spring Schools at our institute is very successful and attracts every year more then 200 students. The topic of the 2007 School is

"PROBING THE NANOWORLD"

with about 50 hours of lectures, discussions and laboratory courses by highly qualified speakers.

Rather cheap accommodation is provided in a youth guest house, amounting to 320 Euro in total for two weeks including breakfast and dinner. All participants receive free of charge a hard cover book with all lectures upon arrival to the course. There is no participation fee.

For more details and an application form see

http://www.fz-juelich.de/iff/fs/

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4.3 International Conference on Fine Particle Magnetism (ICFPM-07)

First announcement

CNR Conference Hall, Rome, Italy

October 9-12, 2007

The 7th International Conference on Fine Particle Magnetism (ICFPM-07) will be held at the CNR Conference Hall in Rome, Italy, from Tuesday, October 9 through Friday, October 12, 2007. ICFPM serves as the forum for scientists and engineers around the world to discuss various issues relating to the recent developments and new trends in nanoparticles magnetism, such as:

Organization of magnetic arrays (self assembled nanoparticles, nanopatterned media)
Multifunctional nanomagnets
Magnetic core/shell nanocomposites
Metamaterials
Surface/interface induced phenomena
Spin dynamics
Magnetotransport
Sensors and devices
Biomedical applications

The Program consists of invited and contributed (oral and poster) papers.

You are kindly invited to submit an abstract by May 15, 2007.

For further information, please visit the ICFPM-07 web site at the address:

http://www.icfpm.mlib.cnr.it

or contact the ICFPM-07 secretary (e-mail: icfpm@ism.cnr.it)

Dino Fiorani

ICFPM-07 Chairman

4.4 International Conference on Materials for Advanced Technologies (ICMAT2007)

http://www.mrs.org.sg/conference/icmat2007/

SYMPOSIUM O: FRONTIERS IN COMPUTATIONAL MATERIALS SCIENCE

1-6 July, 2007, Singapore

CALL FOR ABSTRACTS

The ICMAT2007 Conference will take place in Singapore, 1-7 July 2007. There will be 18 Symposia covering contemporary topics of importance for science, engineering and technology of materials. ICMAT2007 also incorporates the GEM4 Conference on Cancer. I would like to draw your attention to Symposium O: Frontiers in Computational Materials Science. The Symposium aims to provide a forum for scientists and engineers who are involved in computational materials science to present and discuss latest development in computational methods related to materials science and their applications. We will also organizing a Postconference WIEN2k Workshop, 6-9 July 2007, to be conducted by Peter Blaha and his team.

The following is a partial list of invited speakers:

Peter BLAHA, Technische Universitat Wien, Germany
Gerbrand CEDER, Massachusetts Institute of Technology, USA
C. T. CHAN, HKUST, China
G. P. DAS, India Association for the Cultivation of Science, India
Sergei DUDAREV, UKAEA Culham Division, UK
Alphonse FINEL, CNRS-ONERA, France
Thomas FRAUENHEIM, Universitat Bremen, Germany
Xingao GONG, Fudan University, China
Guang Yu GUO, National Taiwan University, Taiwan
Andrew HORSFIELD, University College London, UK
Jisoon IHM, Seoul National University, Korea
P. JENA, Virginia Commonwealth University, USA
Yoshiyuki KAWAZOE, Tohoku University, Japan
Adam KIEJNA, University of Wroclaw, Poland
Sukit LIMPIJUMNONG, Suranaree University of Technology, Thailand
Steven G. LOUIE, University of California, Berkeley, USA
Risto M. NIEMINEN, Helsinki University of Technology, Finland
Nguyen Hong QUANG, Vietnam Academy of Science and Technology, Vietnam
Jae-Hyeok SHIM, Korea Institute of Science and Technology
Mojmir SOB, Academy of Sciences, Czech Republic
Cathy STAMPFL, University of Sydney, Australia

Kiyoyuki TERAKURA, Hokkaido University, Japan
John TSE, University of Saskatchewan, Canada
Vasek VITEK, University of Pennsylvania, USA
Enge WANG, Chinese Academy of Sciences, China
Su-Huai WEI, National Renewable Energy Laboratory, USA
Franois WILLAIME, Service de Recherches de Mtallurgie Physique, France
Christopher WOLVERTON, Ford Research and Advanced Engineering, USA
Shengbai ZHANG, National Renewable Energy Laboratory, USA
Zhenyu ZHANG, Oak Ridge National Laboratory, USA

Deadline for abstract submission: 31 January 2007

For more information, please contact

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Chair, Symposium O, Frontiers in Computational Materials Science, ICMAT2007 Department of Physics National University of Singapore
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4.5 EUROMAT 2007 Conference: Symposium D3 on "Materials Modelling on all Length Scales"

Sept. 10-13, 2007, Nuremberg, Germany

Organized by D. Raabe and P. Turchi

The EUROMAT 2007 Conference will offer 24 topics within 87 Symposia. Here we would like to draw your attention to the Symposium D3 on "Materials Modelling on all Length Scales". There will be four symposia within this topic:

D31 - Atomistics and Ab Initio Materials Modelling, organized by M. Sob
D32 - Modelling Plasticity at small Scales organized by M. Geers
D33 - Process Modelling of Metallic Alloys, organized by Y. Bréchet
D34 - Biomaterials Simulation, organized by T. Frauenheim.

In the D31 Symposium we plan to consider most of the oral contributions as invited talks, although, according to the rules of EUROMAT, we will not be able to waive the conference fee for invited speakers.

A detailed information on the EUROMAT Conference may be found on the web-page

http://www.euromat2007.fems.org/

The most important date just now is

31 Jan 2007: Deadline for the submission of abstracts.

Participating in EUROMAT will give you a possibility to see the latest development in materials science and solid-state physics. We are looking forward to meeting you in Nuremberg! Please, submit your abstracts soon!

Prof. Dr. Mojmir Sob, DSc.
Department Head
Department of Theoretical and Physical Chemistry
Faculty of Science
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Fax : (+420-5) 49491411
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4.6 WIEN2007: Hands on Workshop on the WIEN2k package

Penn State University, University Park, PA, USA

June 11-14, 2007

J.O. Sofo and the WIEN2k developers team

http://www.msc.psu.edu

The third US workshop on the WIEN code is a hands-on activity dedicated to teach the use of the WIEN2k Density Functional Theory Package (http://www.wien2k.at). WIEN2k is one of the most popular electronic structure codes used to perform calculations with the Full Potential LAPW method. The workshop is a unique opportunity to learn the use, power and limitations of the package with the tutoring of the authors and developers of the code. The activities will be aimed at graduate students and researchers from industry and academia. The only pre-requisite is to have a basic knowledge of solid state physics, and chemistry. It is planned as a four-day activity with lectures on the scientific aspects of the method, applications, hand-on activities on selected examples, and the opportunity to receive a head start on a personal project.

The registration is now open. This year the participation is limited to 40 people. For more information or to register go to http://www.msc.psu.edu

The tentative list of invited speakers includes:

-Prof. C. Ambrosch-Draxl (Univ. Leoben, Austria)
-Prof. P. Blaha (TU Vienna, Austria)
-Dr. J. Luitz (TU Vienna, Austria)
-Dr. G. Madsen (Univ. Aarhus, Denmark)
-Prof. L. Marks (Northwestern Univ, USA)
-Dr. K. Jorissen (Univ. Washington, USA)
-Prof. K. Schwarz (TU Vienna, Austria)

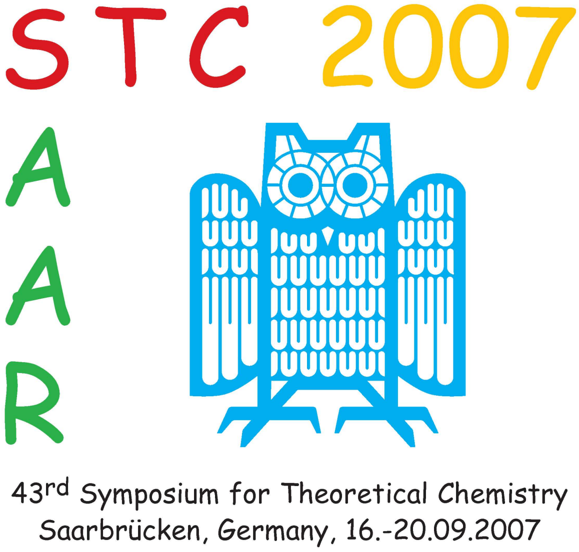
Chair: Prof. J. O. Sofo (Penn State Univ, USA)

4.7 43rd Symposium for Theoretical Chemistry: Large, Non-Biological Systems

Saarbrücken, Germany

16.–20.09.07

http://stc2007.uni-saarland.de



SUBJECT

A 4-days conference on theoretical methods for the calculation of electronic and structural properties of materials shall be held in Saarbrücken, September 2007. It shall cover various aspects of theoretical chemistry, including the boarder region between chemistry and physics. The conference shall be held as a low-cost conference (conference fee: 90 Euro, including meals, excursion, and conference dinner).

TOPICS

Invited lectures will be given on the following topics:

1. Clusters (David Wales, Cambridge)
2. Polymers and oligomers (Benoit Champagne, Namur)
3. Surfaces and catalysis (Joachim Sauer, Berlin)

4. Crystals (Karlheinz Schwarz, Wien)
5. Density functional theory (John Perdew, New Orleans)
6. Relativistic effects (Richard L. Martin, Los Alamos)
7. Local-increment methods (Hermann Stoll, Stuttgart)
8. Coupled-cluster methods (Poul Jørgensen, rhus)
9. Excitations, Responses, Spectroscopy (Arne Lüchkow, Aachen)
10. Dynamics (Dominik Marx, Bochum)
11. Solvation (Benedetta Mennucci, Pisa)
12. Spin effects (Martin Kaupp, Würzburg)

In addition, of the contributed papers, some will be selected for oral presentations. Any contribution within the field of theoretical chemistry / computational materials science is most welcome.

PROGRAMME

	Sunday 16.09.	Monday 17.09.	Tuesday 18.09.	Wednesday 19.09	Thursday 20.09.
Morning		Oral presentations	Oral presentations	Oral presentations	Oral presentations
Afternoon	Arrival + registration	Oral presentations	Oral presentations	Excursion	Departure
Evening	Inofficial gathering	Posters + buffet	Posters + buffet	Conference Conference	

CONFERENCE SITE

The conference will be held at the Saarbrücken campus of the University of Saarland. Saarbrücken is the capital of Saarland, one of the smallest states of Germany, lying on the boarder to France and Luxembourg. It can easily be reached by air, train, and car.

EXCURSION

The excursion will bring us to Völklinger Htte (a Unesco World Cultural Heritage) and the *Saarschleife*.

REGISTRATION AND FURTHER INFORMATION

Bus transportation to / from the hotels will be included in the conference fee.

Registration and abstract submission will be accepted until July 1st 2007.

The conference language is English.

Please consult the homepage of the conference:

http://stc2007.uni-saarland.de

for any further information.

CORRESPONDENCE

STC07
c/o Michael Springborg
Physical and Theoretical Chemistry
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Germany

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5 General Job Announcements

Postdoctoral Position

”Energies and charges of point defects and clusters in uranium oxide”

CEA Saclay, Paris, France

The position is opened for July 2007 in the Physical Metallurgy Laboratory of French Atomic Energy Commission, CEA/SRMP, in Saclay, 25 km south of Paris, France.

The Post-doc will be part of a group of 15 theoretical researchers working on material science at the atomic scale with a focus on diffusion properties and behaviour under irradiation

The goal of this post-doc is to study by numerical simulation the charge and the structure of point defects in uranium dioxide. Previous studies have shown that the modelling of these defects by ab initio Density Functional Theory gives results which are only partly satisfactory. The obtained formation energies are indeed correct. However, two problems arise that we wish to correct during this post-doc.

First, it was not possible to study the charge of the defects which certainly plays a role in their relative stability. Indeed the usual Density Functional Calculation (DFT) calculations we did predicted UO_2 to be metallic whereas it is experimentally an insulator. This discrepancy comes from the fact that this material is not a band insulator (as most semi-conductors or insulators are), but a Mott insulator, i.e. a solid where local electronic correlations play a major role. Modelling such insulator requires to use an advanced method to describe the electronic structure. Such a method (probably the one called “LDA+U”) shall be used to obtain an insulator and therefore to be able to study the charge of the defects.

Second, preliminary results indicate that the oxygen interstitials are not isolated but are forming clusters of complex structure. The structure and energy of these clusters shall be studied.

From the defect properties macroscopic transport properties (electrical conductivity, thermopower, diffusion coefficients) will be calculated which will enable the comparison with experimental measurements.

To sum up, we propose in this post-doc to do ab initio electronic structure calculations. The simulation codes (Siesta and/or Abinit) that will be used are of the highest standard. However it is quite possible that some numerical developments will be needed to adapt them to our calculations. The candidate should therefore have a PhD in Solid State Physics or Material Science and knowledge of electronic structure calculations.

The initial contract, starting in July 2007, will be for one year with a possible extension for one more year. The salary is approximatly 2200 euros per months.

The administrative conditions are :

-PhD thesis prepared outside of a CEA laboratory ;

-Thesis defence less than two years old ;

-Maximum one previous post-doc (outside CEA) or less than 6 months unemployment since thesis defense ;

-Aged under thirty.

Please send CV, motivation letter and letters of recommendation or contact information for references to

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Visiting Assistant Professor Position
Case Western Reserve University, Cleveland, OH, U.S.A.

The Department of Physics at Case Western Reserve University invites applications for up to two Visiting Assistant Professor Positions to begin in academic year 2007-2008. These positions will be for a period of 1-3 years. Candidates should have a Ph.D. and be committed to excellence in teaching and research. The teaching load will allow ample time for research and the candidates will have the opportunity to collaborate with any of our active research programs. Successful candidates may be in any field of physics in which the department is currently active, including theoretical, computational and experimental condensed matter physics, particle/astrophysics and cosmology, and medical physics. More information on the departments research and teaching programs can be found at http://www.phys.cwru.edu. Interested candidates should apply to Visiting Professor Search, Department of Physics, Case Western Reserve University, 10900 Euclid Avenue, LC 7079, Cleveland, OH 44106-7079 or by email to pab6@cwru.edu. Electronic applications in pdf format are encouraged. Applications should include curriculum vitae and a statement of teaching and research interests. The candidates should arrange for three references to send letters of recommendation. Applications received by February 28, 2007 will receive full consideration. Case Western Reserve University is committed to diversity and is an affirmative action, equal opportunity employer. Applications from women and minorities are encouraged. Case Western Reserve University has received an NSF ADVANCE grant to increase the participation of women in science and engineering.

Post-Doctoral Position

Fraunhofer Institute for Mechanics of Materials (IWM), Freiburg, Germany

A postdoc position is immediately available jointly at the Fraunhofer Institute for Mechanics of Materials (IWM), Freiburg and at the Institute for Reliability of Systems and Devices (izbs), University of Karlsruhe, Germany, within the newly established independent junior research group "Biomaterials Interfaces".

The postdoc researcher will use simulations techniques of her/his choice to investigate the processes of protein adsorption on oxidised and hydrated metal and semiconductor surfaces. The aim of his/her research work will be to understand how different surface chemistries influence structural changes in typical proteins of the extracellular matrix (or of the blood plasma) upon adsorption.

The initial appointment will be for two years, but the position is renewable up to four years. Remuneration will be offered according to the German TVöD (EG 13) salary scheme.

Applicants are expected to possess a solid background in (bio)physics, (bio)chemistry, polymer science or life sciences, computer programming skills and the ability of working independently. Experience in the theoretical modelling of macromolecule dynamics or protein folding represents a strong advantage. Knowledge of the German language is not required.

Interested candidates should send their applications including CV, publication list, and the names and addresses of at least two referees, preferably via electronic mail, to:

Lucio Colombi Ciacchi (lucio@izbs.uni-karlsruhe.de)
Fraunhofer Institut für Werkstoffmechanik,
Wöhlerstr. 11, 79108 Freiburg, Germany.
Tel: +49 761 5142113.

For further information click at:
`http://www.izbs.uni-karlsruhe.de/Stellenangebote/postdoc_biomaterials_izbs.html`

PhD Position

Fraunhofer Institute for Mechanics of Materials (IWM), Freiburg, Germany

A PhD position is immediately available jointly at the Fraunhofer Institute for Mechanics of Materials (IWM), Freiburg and at the Institute for Reliability of Systems and Devices (izbs), University of Karlsruhe, Germany, within the newly established independent junior research group "Biomaterials Interfaces".

The successful candidate will use a combination of first-principles (DFT-based) and classical molecular dynamics techniques to study atomistically the physical and chemical interactions between oxidised metal and semiconductor surfaces and selected protein fragments (oligopeptides). She/He will investigate how intrinsic materials properties and/or extrinsic factors (e.g. mechanical stress) affect the surface chemistry and how this, in turn, influences the adsorption of oligopeptides and their specific cell adhesion functionalities. Training in the areas of first-principles and classical molecular dynamics, surface physics and biomaterials chemistry will be provided.

The appointment will be for three years. Remuneration will be offered according to the German TVöD salary scheme.

Applicants should possess a solid background in (bio)physics, (bio)chemistry, materials science, engineering or related disciplines. Existing expertise in atomistic modelling, DFT-based simulations, solid state physics or surface chemistry is welcome but not essential. Knowledge of the German language is not required.

Interested candidates should send their applications including CV and academic record, preferably via electronic mail, to:

Lucio Colombi Ciacchi (lucio@izbs.uni-karlsruhe.de)
Fraunhofer Institut für Werkstoffmechanik,
Wöhlerstr. 11, 79108 Freiburg, Germany.
Tel: +49 761 5142113.

For further information click at:
`http://www.izbs.uni-karlsruhe.de/Stellenangebote/phd_biomaterials_izbs.html`

PhD Studentship

Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

At the Theory Department of the Max Planck Institute of Microstructure Physics in Halle (Saale), Germany, a **position for a PhD student** in the field of **Spin-dependent Transport in Nanostructures** is available immediately.

The successful candidate will work on the electronic and magnetic structure of low-dimensional systems and on electronic transport in nanostructures. Besides a knowledge of condensed-matter theory, experience with electronic-structure calculations and computer programming (C++ preferred) is required. It is expected that the candidate will carry on ongoing projects on current problems in spintronics and will develop further existing computer program packages. Collaboration with colleagues from both theoretical and experimental physics is desirable. The graduation will be at the Martin Luther University Halle-Wittenberg.

The Max Planck Institute of Microstructure Physics is an internationally highly competitive institution. The Theory Department is directed by Prof. Patrick Bruno. For details, see `http://www.mpi-halle.mpg.de/~theory`. The MPI is embedded in a stimulating environment, comprising in particular the International Max Planck Research School for Science and Technology of Nanostructures and the Martin Luther University Halle-Wittenberg.

The Max Planck Society is interested in increasing the proportion of women among its scientists and strongly encourages women to apply to the position. Applications from disabled candidates are encouraged and will be given priority in case of equal qualification.

An application, to be sent by electronic mail as a single pdf file to Dr. Jürgen Henk, should include a cover letter, a resume, a statement of qualifications relevant for the position, as well as names and complete addresses of two referees. Applications will be scanned until the position is filled.

Contact: Dr. rer. nat. habil. Jürgen Henk, Theory Department, Max Planck Institute of Microstructure Physics, Weinberg 2, D-06198 Halle (Saale), Germany. Electronic address: `henk@mpi-halle.de`.

Postdoctoral Position

Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

At the Theory Department of the Max Planck Institute of Microstructure Physics in Halle (Saale), Germany, a **post-doctoral position** in the field of **Multiferroic Systems** is available immediately.

The successful candidate will work on the electronic and magnetic structure of biferroic interfaces and on electronic transport in multiferroic tunnel junctions. Besides a profound knowledge of condensed-matter theory, experience with first-principles electronic-structure calculations, electronic transport theory, and computer programming (C++ preferred) is required. It is expected that the candidate will develop further existing state-of-the-art first-principles computer program packages (based on relativistic multiple-scattering theory) and apply these to problems at the frontier of spintronics. Further, collaboration in ongoing theoretical projects on properties of low-dimensional systems and cooperation with colleagues from experimental physics is highly desirable.

The Max Planck Institute of Microstructure Physics is an internationally highly competitive institution. The Theory Department is directed by Prof. Patrick Bruno. For details, see `http://www.mpi-halle.mpg.de/~theory`. The MPI is embedded in a stimulating environment, comprising in particular the International Max Planck Research School for Science and Technology of Nanostructures and the Martin Luther University Halle-Wittenberg.

The Max Planck Society is interested in increasing the proportion of women among its scientists and strongly encourages women to apply to the position. Applications from disabled candidates are encouraged and will be given priority in case of equal qualification.

An application, to be sent by electronic mail as a single pdf file to Dr. Jürgen Henk, should include a cover letter, a resume, lists of publications and selected presentations, a statement of qualifications relevant for the position, as well as names and complete addresses of three referees. Applications will be scanned until the position is filled.

Contact: Dr. rer. nat. habil. Jürgen Henk, Theory Department, Max Planck Institute of Microstructure Physics, Weinberg 2, D-06198 Halle (Saale), Germany. Electronic address: `henk@mpi-halle.de`.

CRISP Postdoctoral Fellowship
Center for Research on Interface Structure and Phenomena (CRISP)
Yale University, USA

The Center for Research on Interface Structure and Phenomena (CRISP) at Yale, an NSF Materials Research Science and Engineering Center (MRSEC), has established a new interdisciplinary research position at the postdoctoral level. The focus is on performing theoretical studies of the structural, mechanical, chemical, and electronic properties of complex oxides and their interfaces by both applying state-of-the-art methods and developing novel approaches for dealing with electronic correlations. In addition, the theoretical effort will connect with parallel experimental efforts within the MRSEC.

The candidate should have a Ph.D. in materials physics or a related field and have prior experience with first principles calculations of material properties. A curriculum vitae and three letters of recommendation should be sent to CRISP@yale.edu, or to CRISP, Yale University, 401 Becton Center, P.O. Box 208284, New Haven, CT 06520-8284. Recommenders should send letters directly under separate cover. The deadline for receipt of all material is January 29, 2007. For further information or questions, please contact Prof. Sohrab Ismail-Beigi (sohrab.ismail-beigi@yale.edu) or Prof. John Tully (john.tully@yale.edu).

Yale is an affirmative action/equal opportunity employer and encourages applications from qualified women and minority group members.

Postdoctoral Position

Simulations of Interaction between Defects and Interstitial solutes in Iron

Department of Materials Science, Delft University of Technology, The Netherlands

Applications are invited for a post-doctoral position that is expected to become available as early as February 2007 but a later starting date is possible. The ideal candidate has experience in computational materials science. A PhD in materials science, physics, theoretical chemistry, or related field is required. The position is focused on developing and applying effective potentials for interstitial solutes in iron, particularly carbon, nitrogen and boron. The effective potentials are derived from electronic density functional calculations. The potentials will be used for large scale MD simulations of the interaction between solutes and extended defects such as dislocations and twins. You will be interacting with a group of PhD students, other staff, and industry so that good verbal communication skills in English are essential. The department of Materials Science at Delft University of Technology offers an exciting research environment with excellent facilities and working conditions. Candidates should send (preferably in pdf format by e-mail) a detailed resume including an outline of their research achievements and interests as well as contact information for academic references to

Marcel Sluiter
Virtual Materials Lab
Department of Materials Science
Delft University of Technology
Mekelweg 2, 2628 CD Delft
The Netherlands

Email:M.H.F.Sluiter@tudelft.nl

Consideration of candidates will begin immediately and continue until the position is filled.

Postdoctoral Position

University of Nebraska-Lincoln, USA

A postdoctoral position in computational materials science is immediately available in the group of Prof. Kirill Belashchenko at the University of Nebraska-Lincoln. The research work will involve applications of electronic structure theory to different problems in materials and nanoscience. The initial appointment will be for one year, with a possible extension upon mutual agreement and availability of funding. Candidates must have a doctoral degree in physics and experience in electronic structure calculations. Interested candidates should send an application letter, CV, list of publications, and contact information for at least two references to Kirill Belashchenko (preferably by email in PDF format).

Review of applications will begin immediately and continue until the position is filled. UNL is committed to a pluralistic campus community through EEO/AA. We assure reasonable accommodation under ADA/504; contact Beth Farleigh at (402)472-2770 for assistance.

Kirill Belashchenko
Assistant Professor
Department of Physics and Astronomy
University of Nebraska-Lincoln, Lincoln
NE 68588-0111

Phone: (402)472-2396
Fax: (402)472-2879
Email: kdbel@unlserve.unl.edu

Ph. D Position

Institute for Reliability of Systems and Devices (IZBS), University of Karlsruhe, Kaiserstr. 12, Karlsruhe, Germany

A PhD position is available in the area of atomistic modeling of materials. The research work will maily focus on the development of analytic bond-order potentials for materials with covalent and mixed metallic-covalent atomic bonding and their application in large-scale atomistic simulations. The analytic bond-order potentials are novel many-body interatomic potentials that have been recently derived by coarse-graining the tight-binding electronic structure and they provide accurate and computationally efficient means to study complex phenomena such as growth of amorphous films, nanotribology, and behavior of extended defects. The research work will be performed under the direction of Prof. Peter Gumbsch and Dr. Matous Mrovec in collaboration with the Fraunhofer Institute for Mechanics of Materials (IWM) in Freiburg and the Department of Materials, University of Oxford, UK.

Candidates are expected to possess a solid background in materials science, physics or engineering. Existing expertise in the atomistic modeling of materials, DFT-based simulations, solid state physics represents a strong advantage. Knowledge of the German language is advantageous but not required.

Interested candidates should send their applications including CV and academic record, preferably via electronic mail, to:

Matous Mrovec (matous.mrovec@iwm.fhg.de)
Fraunhofer Institut fuer Werkstoffmechanik,
Woehlerstr. 11, 79108 Freiburg, Germany
Tel: +49 (0)761 5142-347.

POSTDOCTORAL RESEARCH FELLOWSHIPS

DEMOCRITOS National Simulation Center,
SISSA International School for Advanced Studies,
and ELETTRA Synchrotron Light Laboratory Trieste, Italy

Two postdoctoral positions are available immediately at the DEMOCRITOS National Simulation Center (http://www.democritos.it) in the following areas:

1) Development and application of ab-initio methods for the study of the catalytic properties of metal nanoclusters supported by oxide surfaces

2) Development and implementation of novel theoretical methods for the first-principles simulation of optical spectra of nanostructured materials.

The successful candidates will join a newly established theoretical research group based at the ELETTRA synchrotron radiation center

(http://www.elettra.trieste.it)

with strong links with the SISSA condensed-matter theory group

(http://www.sissa.it/cm/).

He/She is expected to develop strong links with the local experimental groups and to be able to work in a highly multidisciplinary environment.

Candidates must hold a PhD in Physics, Chemistry, Materials Science, or related discipline, and have good knowledge of electronic structure theory. Previous experience with first-principles methods as well as scientific programming is required. Additionally, for position 2, some acquaintance with many-body theory and/or time- dependent density-functional theory, though not strictly necessary, would be an advantage. Initial appointment is for two years and consideration of candidates will begin immediately.

Interested applicants should submit a CV, including list of publications and name (with e-mail address) of two referees to Stefano Baroni (baroni@sissa.it) and to

Stefano Fabris (fabris@democritos.it) (for position 1) or Paolo Umari (umari@democritos.it) (for position 2).

INFM-DEMOCRITOS National Simulation Center and SISSA International School for Advanced Studies via Beirut 2-4, I-34014 Trieste, Italy.

Professor in Physics

TEKNILLINEN KORKEAKOULU
TEKNISKA HÖGSKOLAN
HELSINKI UNIVERSITY OF TECHNOLOGY, FINLAND

The field of the professorship is specified to be Experimental physics. The post will be filled either permanently or initially for a period of five years. The post is placed at the Department of Engineering Physics and Mathematics (http://fysiikka.tkk.fi).

Additional information is available from academy professor Risto Nieminen email: risto.nieminen@tkk.fi, and administrative manager Dr. Riikka Mäki-Ontto, tel: +358-9-451 3000, email: riikka.maki-ontto@tkk.fi.

The particulars of nomination, which contain information about the position and application procedures, are available from the Registrar's Office of the University, tel: +358-9-451 2030. The particulars of nomination can also be found on the TKK website

http://www.tkk.fi/Yksikot/Hallitus/Virat/index.html.

The application is to be addressed to the Council of Helsinki University of Technology. The closing date for applications is 31st of January 2007 before 3.45 p.m.

The street address of the Registrar's Office is Otakaari 1, Espoo, and the postal address is Helsinki University of Technology, P.O.Box 1000, FI-02015 TKK, Finland. Applications can also be delivered by fax or e-mail before the application deadline 31st January 2007 (TKK, Registrar's Office, fax: +358-9-451 2017, email: kirjaamo@tkk.fi). The details about sending the document electronically can be found in the particulars of nomination.

Postdoctoral Position
Institut Francais du Pétrole (IFP), France

”First-principles simulation of heterogeneous catalytic reactions mediated by supported metallic nano-aggregates”

A post-doctoral position is available in the Applied Chemistry and Physical Chemistry R&D Division at Institut Francais du Pétrole (IFP Rueil-Malmaison, Paris suburb) The position is now open, and funded for 18 months from starting date.

The candidate should hold a PhD defended less than one year ago, and be skilled in DFT based calculations applied to solids and surfaces. This theoretical work is part of a new project funded by the French National Research Agency (ANR) within the framework of the High Performance Computing and Simulation Program. The other partners are academic laboratories with first class expertise in electronic structure calculation and applied mathematics. The project will involve advanced transition state search methods. The rather sophisticated systems considered will aim at realistic representations of industrial catalysts in usage in petroleum refining and petrochemistry.

Motivated candidates are invited to send their CV including a list of publications and a brief description of research experience to:

Professor Hervé Toulhoat
Deputy Scientific Director
Director of Doctoral Studies
Institut Francais du Pétrole
Tel.: +33 1 47 52 73 50
Fax: +33 1 47 52 70 36
E-mail: herve.toulhoat@ifp.fr

Web page: `http://www.ifp.fr/IFP/fr/formation/professeurs/Herve_TOULHOAT.html`

Computational Chemistry and Catalysis web site: http://gdrdmq.ifp.fr

Job opening: Postdoctoral Position in Computational Nano-physics

Center for Atomic-scale Materials Design (CAMD), Lyngby, Denmark

The Center for Atomic-scale Materials Design (CAMD) is offering a postdoctoral position in the field of electron transport in nano-scale devices. Applicants should have background in theoretical physics or quantum chemistry at the PhD level. Basic knowledge of many-body theory and/or electronic structure theory as well as some programming experience is preferred.

CAMD is a theoretical materials science research center located at the Technical University of Denmark north of Copenhagen. We offer a dynamic and international working environment with research activities within several areas of theoretical and computational solid state physics including surface science, catalysis, biophysics, and molecular electronics. We have extensive experience in electronic structure theory and its applications, and our two state-of-the-art DFT codes, the plane- wave pseudopotential code ”Dacapo” and the real space PAW code ”GPAW”, are under continuous development Our Linux supercomputer cluster, Niflheim, containing 900+ CPUs provides optimum conditions for both small-and large-scale computer simulations.

The applicant will work with the application and development of a new method for modelling (correlated) electron transport in nano-scale electronic devices utilizing techniques from many body theory (within the Keldysh framework) and density functional theory (DFT). More standard methods like DFT-based transport as well as MD simulations of nano-contacts will also be applied to investigate electronic and mechanical properties of systems of experimental interest. The monthly salary will be around 32,700 DKK (approx. 4360 Euro). The position is for one year with a possibility for extension to two years, and is available now.

Applications including CV and list of publications should be sent to Assistant Professor Kristian Thygesen (thygesen@fysik.dtu.dk) by February 15, 2007.

Postdoctoral Research Fellow

”Computational modelling in extreme conditions physics”

School of Physics, The University of Edinburgh, Scotland

A position is available for a computational modeller to work in the Extreme Conditions Physics Group of the School of Physics (part of the Scottish Universities Physics Alliance, SUPA) in the Centre for Science at Extreme Conditions. This follows the 3.5M renewal of the group's EPSRC Programme Grant.

You will undertake ab-initio total-energy calculations using both static and dynamic methods to provide insight into the behaviour and physical properties of elements and simple molecular systems under extreme conditions working closely with the group's experimental programme. You will also be involved in developing new methods for finding trial structures for unknown crystals, making use of experimental data to assist in full solution of as-yet unsolved diffraction patterns.

The group has established world-leading programmes in extreme conditions electronic structure calculations and in experimental structural studies using both x-ray and neutron diffraction, and also in extreme conditions spectroscopic and transport properties measurements.

`http://www.jobs.ed.ac.uk/vacancies/`
    `index.cfm?fuseaction=vacancies.detail&vacancy_ref=3006794`

Postdoctoral Position
University of California, Davis

A postdoctoral position in theoretical condensed matter physics is available immediately at the Physics Department of University of California, Davis. This project is part of a research program that focuses on the development of a better phenomenological and microscopic understanding of the novel heavy fermion 1-1-5 based superconductors. It involves developing and applying to these materials new methods for calculating electronic properties of strongly correlated systems using combinations of density functional and dynamical mean field based methods, while working in close collaboration with leading experimentalists in this area.

We are looking for an exceptionally able and energetic individual who will join the UC Davis theoretical research group led by Daniel Cox, David Pines, and Sergey Savrasov, which has close ties to the heavy electron experimental groups at UC Irvine, Michigan, UIUC, and Los Alamos National Laboratory.

Prospective applicants should hold a PhD or equivalent degree in physics. Experience with density functional and many body calculations, Quantum Monte Carlo, numerical methods and programming using Fortran, Java and/or C++ is highly desired.

Interested candidates should email their resumes and the names of at least two references to Sergey Savrasov (savrasov@physics.ucdavis.edu).

For additional information about the Department of Physics at UC Davis, please visit our web site

http://www.physics.ucdavis.edu.

The University of California is an Affirmative Action, Equal Opportunity Employer.

Material Scientist or Physicist
Paul Scherrer Institut, Switzerland

At the Paul Scherrer Institut, Switzerland (www.psi.ch), a position for a Material Scientist or Physicist is available to perform atomistic simulations on materials of interest as nuclear fuel in present and future reactor technologies. The position is for an experienced scientist with good programming skills. Good knowledge about Molecular Dynamics and Kinetic Monte Carlo approaches are also strongly desirable. This offer is for a permanent position that will be re-discussed with the management every three years on the basis of goals and achievements.

The Paul Scherrer Institut is a centre for multi-disciplinary research and one of the world's leading user laboratories. With its 1200 employees it belongs as an autonomous institution to the Swiss ETH domain and concentrates its activities on solid-state research and material sciences, elementary particle physics, energy and environmental research as well as on biology and medicine.

Fuel modelling in the STARS project, operated within the Laboratory for Reactor Physics and Systems Behaviour (LRS) at PSI, is devoted to the application of computational tools to investigate the behaviour of nuclear fuel under normal and accident operating conditions. To increase the understanding of fuel thermal-mechanical behaviour at a microstructural level, we are developing a multiscale computational methodology which will address fundamental issues of current nuclear fuels and in a longer perspective, of advanced fuels for future reactor design. At the initial stage, the project will focus on the mechanisms of deformation and mass transport in crystalline UO_2 as well as in the presence of point and extended defects (vacancies, interstitials, grain boundaries). The investigation will be based on both the Molecular Dynamics (MD) and kinetic Monte Carlo (KMC) approach.

Research tasks for the candidate:

To develop together with present staff - a computational methodology to investigate thermal-mechanical and transport properties in crystalline and polycrystalline materials such as UO_2. In particular, you will address compelling scientific issues such as the diffusion mechanism at GB in UO_2 and in a first phase will contribute KMC- based modeling to the calculation of the diffusion coefficient in polycrystalline UO_2.

This offer is for a three year permanent position, available immediately.

Candidate's profile:

Minimum requirements are a Ph.D. degree in Materials Science or Physics. Hands-on experience with Kinetic Monte Carlo Methods codes as well as good programming skills and experience with parallel computation are required. Knowledge of other atomistic techniques, such as Molecular Dynamics and ab-initio are desirable. Self motivation and the ability to work both independently and as part of a team are essential requirements. Communication skills in English are requested.

For further information please contact Mr. Martin Zimmermann, ph.+41 (0)56 310 27 33, e-mail: martin.zimmermann@psi.ch or Dr. Marcella Iannuzzi, Tel. +41 (0)56 310 2784, e-mail: marcella.iannuzzi-mauri@psi.ch.

Please send your application to: Paul Scherrer Institut, Human Resources, Mrs. H. Habersaat, ref. code 4103-04, 5232 Villigen PSI, Switzerland.

`http://pa.web.psi.ch/pages/offenestellen/d/index_offenestellen.php?id=621&was=stelle`

Postdoctoral or Ph. D. Positions

Atomistic Modelling and Design of Materials

University of Leoben, Austria

At the Chair of Atomistic Modelling and Design of Materials at the University of Leoben, Austria

(http://www.uni-leoben.at/amadm)

three postdoctoral or Ph. D. positions are available immediately. The research areas are carbon nanostructures and metal alloys.

The nanotube project is dealing with a new generation of pea-pods, composed of a photoactive organic semiconductor (pea) and a single-wall nanotube (pod). It is embedded into a small EU-wide network involving chemists, as well as experimental and theoretical physicists. For more information, have a look at

http://www.loima.fmns.rug.nl/Naphod.html.

Two of the positions are dedicated to ab-initio investigations of steel and other metallic alloys towards the design of materials with desired physical properties. The University of Leoben has long-standing experience in metal physics and metallurgy combining expertise in simulations and experiments with a strong industrial background. The present projects also aim at strengthening the link between the atomistic modelling and continuum- mechanical approaches.

The candidates should have experience in ab-initio calculations based on density functional theory and a strong background in theoretical solid state physics. Expertise in FORTRAN90 is desired. Highly talented students and postdocs who enjoy working in a team, collaborate with experimentalists, and contribute to the friendly atmosphere of the group are most welcome!

Applications should be sent via email to Claudia Ambrosch-Draxl (cad@mu-leoben.at) together with two letters of recommendation.

Claudia Ambrosch-Draxl
Chair of Atomistic Modelling and Design of Materials
Department Materials Physics, University of Leoben
Erzherzog Johann - Strae 3, A-8700 Leoben
http://www.mu-leoben.at/amadm
Phone: +43 3842 402 4400
Fax: +43 3842 402 4402 Fax

***** Posdoctoral position(s), University of the Basque Country, Spain

Posdoctoral Position(s)

Materials Science Department, University of the Basque Country (ETSF and Nano-bio Spectroscopy Group)

We are looking for highly qualified candidates from physics, chemistry, and materials science. Type of appointment: contract as visiting research professor for two years. The work to be done by the postdoc(s) will be related to the one of the following topics:

* Optical properties of biochromophores, role of environment and Excited state dynamics. (within the NANO-ERA Chemistry programme)

* Response functions of one-dimensional systems: excitonic effects. Low dimensionality effects, screening and role of packing.

* Time-dependent density functional approach to molecular transport. Description of the stationary state and AC-current. Photon induced transport and combined electron/ion dynamics with current and/or external electromagnetic field.

The postdoc(s) will be fully-involved in the activities of the nanoGune research center

(http://www.nanogune.eu)

as part of the strategic area "Basic research of nanostructures and low-dimensional structures" as well as in the establishing of the ETSF related to the node in San Sebastian (in close collaboration with the NANOQUANTA nodes in Palaiseau, Berlin, Rome, Milan, Louvain, Lund and York, see

http://www.nanoquanta.es).

Further information on the research of our group as well as a publication list can be found in the group web page: http://dipc.ehu.es/arubio

We consider applications until the positions are filled. Candidates should send their curriculum vitae (including list of publications), description of their research activity and interests, and 2 to 3 recommendation letters to:

Prof. Angel Rubio
Dpto. Fisica de Materiales, Facultad de Quimicas
U. Pais Vasco, Centro Mixto CSIC-UPV/EHU and
Donostia International Physics Center (DIPC)
Apdo. 1072, 20018 San Sebastian/Donostia. Spain
Mail : Angel.Rubio@ehu.es
http://dipc.ehu.es/arubio

European Theoretical Spectroscopy Facility (ETSF)
http://www.etsf.eu
http://www.etsf.es

Two postdoctoral positions
University of Cincinnati/Oak Ridge National Laboratory
in "Computational Correlated Electronic Systems"
Sponsored by DOE/SciDAC

Applicants should have a PhD in Physics, Applied Math, Computer Science or a related field with a strong emphasis in high performance computing, and an interest in numerical linear algebra and the development and application of new Computational methods for correlated electronic many body systems. Individuals with a strong background in quantum Monte Carlo, or diagrammatic numerical many body methods such as FLEX or Parquet equations are preferred. Applications must be made electronically, at www.jobsatuc.com, job numbers 26UC1397 and 26UC1398, and should include a CV, publication list, copies of up to three relevant articles, and three letters of recommendation. We will begin evaluating applications immediately, and expect the positions to be filled no later than late Sept. 2007. The University of Cincinati is an Equal Opportunity/Affirmative Action Employer. Women and Minority candidates are encouraged to apply.

Fully-funded PhD Studentship

Project title: "Quantum mechanical modelling of semiconductor nanocrystals"

University of Southampton, School of Chemistry, U. K.

We are looking for an exceptional, highly motivated student for this project which will offer training in state-of-the-art methods for atomistic simulations and their application to study some of the most promising materials in nanotechnology. It will involve the computational modelling of structural, electronic and optical properties of semiconductor nanocrystals at the atomic level. The size of these crystals is such that a plethora of quantum phenomena are observed which can be exploited in nanoscale manipulations of light and electrons. This is currently a very active research area as the miniaturisation of electronic devices is approaching atomic dimensions and there is great demand for predicting nanostructures with controlled physical properties that can be produced by chemically well defined procedures. The simulations will involve linear-scaling ab initio quantum mechanical calculations with the ONETEP program as well as other approaches based on finite element methods. Depending on the inclinations of the student, the project can also involve some development of theory and computer code for the simulations. The project will be carried out in the group of Dr Chris-Kriton Skylaris in the School of Chemistry in collaboration with the group of Dr Pavlos Lagoudakis in the School of Physics and Astronomy.

This studentship is open to candidates from EU countries and will be advertised until it is filled.

For enquiries, please contact Dr Chris-Kriton Skylaris (cks@soton.ac.uk).

Post-doctoral position in theoretical semiconductor physics

FACULTY OF SCIENCE

DEPARTMENT OF PHYSICS AND ASTRONOMY, ARRHUS, DENMARK

The Department of Physics and Astronomy (www.phys.au.dk) invites applications for a 2- year position in theoretical semiconductor physics. The position is open from April 1, 2007. The desired area of specialization is fundamental properties of diluted magnetic semiconductors and experience in this field is required.

Applications must be in English and include a curriculum vitae, a complete list of publications, a statement of future research plans and information about research activities, teaching qualifications and management experience, all in 4 copies (see http://www.nat.au.dk/stilling/cv for the recommended level of detail). If the applicant wants other material to be considered in the evaluation (publications and other documentation of research and teaching qualifications, as well as management experience) such material must be clearly specified and must either be enclosed in hardcopy (3 copies) or must be available electronically.

The Faculty refers to the Ministerial Order No. 170 of 17.03.2005 (http://www.au.dk/da/21-05.htm) on the appointment of teaching and research staff at the universities under the Ministry of Science, Technology and Innovation.

Salary depends on seniority as agreed between the Danish Ministry of Finance and the Confederation of Professional Unions.

Applications should be addressed to The Faculty of Science, University of Aarhus, Ny Munkegade, Building 1520, DK-8000 Aarhus C, Denmark, and marked 212/5-167.

The deadline for receipt of all applications is February 7, 2007, at 12,00 noon.

Further information can be obtained from Assoc. Prof. Axel Svane, Department of Physics and Astronomy, University of Aarhus, Ny Munkegade, Building 1520, DK-8000 Aarhus C. Tel: +45 8942 3678, e-mail: svane@phys.au.dk.

POSTDOCTORAL POSITION

Theory and Modeling Research Division at Linköping University, Sweden

A postdoctoral position in theoretical condensed matter physics is available at the Department of Physics, Chemistry and Biology, Linköping University, Sweden.

The theory group is participating in the development of the user-friendly simulation packages for the electronic structure calculations, molecular dynamics and statistical mechanics simulations of the phase stabilities of materials. The goal for the project is to develop further our competence in atomistic simulations at the nanoscale, to integrate theory and experiments, and to transfer the competence to experimental groups investigating structural properties of complex materials, hard thin film coatings, multilayer systems relevant for X-ray mirrors, alloy wide-gap nitrides, etc.

We are looking for a strong candidate with PhD in physics and substantial experience in Density Functional calculations and/or statistical mechanics/molecular dynamics simulations. An expertise in the field of first-principles simulations for strongly correlated materials, including methodological developments and applications is highly desirable. Demonstrated ability of programming in Fortran and /or C++ are requested. Interested candidates should submit their CV, including names of at least two references and the list of publication, before February 28, 2007 to

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POSTDOCTORAL POSITION

LAWRENCE BERKELEY NATIONAL LABORATORY, U.S.A.

POSITION SUMMARY: The scientific computing group in the computational research division at LBNL has two immediate openings for Postdoctoral Fellows in nanoscale electronic structure calculations. These positions are funded by DOE nanoscience initiatives. The group uses large parallel machines at the national energy research scientific computing center (NERSC, http://www.nersc.gov). The scientific computing group (http://hpcrd.lbl.gov/html/SCG.html) is a diverse group working on various aspects of scientific computation. More information about our research activities can be found at http://crd.lbl.gov/ linwang.

DUTIES: The successful candidate will work with Dr. Lin-Wang Wang in (1) methodology developments of the charge patching method or other linear scaling method for large nanosystem and organic system electronic structure and total energy calculations; (2) theoretical studies of physics phenomena in nanosystems and molecular electronics, including electronic structures, optical properties, and charge transports.

QUALIFICATIONS: Required – A Ph.D degree in physics or related fields. Experience in electronic structure calculations and code programming. Desired – Experience in methodology developments and nanostructure calculations.

NOTES: This is a one year term position with the possibility of renewal for up to three years. Salary for post-doctoral positions depends on years of post-degree experience and are competitive with market.

HOW TO APPLY: All applicants must apply online,

http://jobs.lbl.gov/LBNLCareers

(search for job keyword "20264"), or go directly to:

`http://jobs.lbl.gov/LBNLCareers/details.asp?jid=20264&p=1`

For more information, you can contact:

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lwwang@lbl.gov

LBNL is an equal opportunity/affirmative action employer.

6 Abstracts

Interfacial structure and half-metallic ferromagnetism in Co_2MnSi-based magnetic tunnel junctions

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Abstract

X-ray absorption (XAS) and x-ray magnetic circular dichroism (XMCD) techniques are utilized to explore the ferromagnetic/barrier interface in Co_2MnSi full Heusler alloy magnetic tunnel junctions. Structural and magnetic properties of the interface region are studied as a function of the degree of site disorder in the alloy and for different degrees of barrier oxidation. Photoelectron scattering features that depend upon the degree of $L2_1$ ordering are observed in the XAS spectra. Additionally, the moments per $3d$ hole for Co and Mn atoms are found to be a sensitive function of both the degree of $L2_1$ ordering and the barrier oxidation state. Significantly, a multiplet structure is observed in the XMCD spectra that indicates a degree of localization of the moments and may result from the half-metallic ferromagnetism (HMF) in the alloy. The magnitude of this multiplet structure appears to vary with preparation conditions and could be utilized to ascertain the role of the constituent atoms in producing the HMF, and to examine methods for preserving the half-metallic state after barrier preparation. The changes in the magnetic structure caused by barrier oxidation could be reversed by inserting a thin Mg interface layer in order to suppress the oxidation of Mn in the Co_2MnSi layer.

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Extremely large spin-polarization in Co_2MnSi based magnetic tunnel junctions

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Abstract

We fabricated Co_2MnSi/(Mg)/Al-O/CoFe MTJs using UHV magnetron sputtering. The interfacial chemical bond between Co_2MnSi and Al-O was intensively optimized by the changing plasma oxidation time for Al-O and by inserting an Mg layer. The Mg inserted layer between Co_2MnSi and Al-O effectively suppressed the generation of interfacial magnetic impurities. Finally, we successfully observed a giant TMR ratio of 203% at 2 K in the MTJ with a 1.0 nm-Mg layer inserted. The spin-polarization for Co_2MnSi estimated from this TMR ratio was 0.97-1.00, which indicated that an almost perfect spin-polarized state was achieved. We also investigated the relationship between TMR ratio and the site-ordering level of Co_2MnSi. As a result, we found that an $L2_1$-ordering state is not necessary to achieve high spin-polarization for Co_2MnSi in MTJs.

To be published in Journal of Magnetic Society of Japan (2007)
Preprint available from: g.vanderlaan@dl.ac.uk.

γ-TaON: A New Metastable Polymorph of Tantalum Oxynitride

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Abstract

A new metastable, low-density polymorph of tantalum oxynitride (γ-TaON) has been prepared by the ammonolysis of tantalum oxide. γ-TaON crystallizes in the VO_2(B)-type structure. At temperatures around 900 °C a transformation to baddeleyite-type β-TaON is observed. Quantum chemical calculations (atomic orbitals and plane-wave basis sets; pure GGA and Hartree–Fock/DFT hybrid functionals, in particular PW1PW; ultra-soft pseudopotentials and PAW) confirm the metastability of the new polymorph because it is ca. 20 kJ/mol less stable than β-TaON. In addition, the electronic-structure calculations allow to distinguish between the O and N atoms and to refine their atomic positions. The distribution of the O and N atoms over the four anionic sites is in harmony with Pauling's second rule of crystal chemistry.

(In press at Angew. Chem. Int. Ed.)
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Soft x-ray resonant magnetic scattering from magnetic nanostructures

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Abstract

Confinement of the dimensions of a magnetic structure to a microscopic scale can lead to intriguing phenomena that are not encountered in bulk materials. Controlled thin film deposition and lithographic patterning have been established as the technology promoters in magnetism research ever since the engineering of multilayers of magnetic and non-magnetic metals has led to the discovery of new phenomena such as inter-layer exchange coupling, giant magnetoresistance (GMR) and non-volatile magnetic random access memory (MRAM). Several different methods are available to characterize the magnetic properties of thin films and confined magnetic structures. Among these, magneto-optical methods are most prominent and have the advantage that they can be performed under applied magnetic and electric fields. However, magneto-optics in the visible region lacks element specificity and moreover the ultimate spatial resolution is limited by the wavelength. The discovery of strong magneto-optical effects in the core level x-ray absorption edges of magnetic elements has opened up new possibilities, such as the separation of the spin and orbital part of the magnetic moments and the determination of the magnetic anisotropy. Such techniques strongly benefit from the advent of synchrotron radiation undulator devices offering variable linear and circular polarization with tuneable photon energy.

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Manuscript available from: g.vanderlaan@dl.ac.uk

Spin- and charge-ordering in oxygen-vacancy-ordered mixed-valence $Sr_4Fe_4O_{11}$

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Abstract

The structural, electronic, and magnetic properties of the oxygen vacancy-ordered mixed-valence $Sr_4Fe_4O_{11}$ phase have been investigated using spin-polarized electronic-structure total-energy calculations. The optimized structural parameters obtained from accurate total-energy calculations are found to be in very good agreement with low-temperature neutron-diffraction findings. Among the different spin configurations considered for $Sr_4Fe_4O_{11}$, the *G*-type antiferromagnetic configuration is found to represent the magnetic ground state. The calculations show finite magnetic moments at both Fe sites and this is against the conclusions arrived at from Mössbauer and low-temperature powder neutron-diffraction measurements, but consistent with the results from magnetization measurements. The present study clearly show that one of the magnetic sub-lattices is frustrated and hence $Sr_4Fe_4O_{11}$ can be considered as a phase-separated system with one phase in the *G*-type antiferromagnetic state and the other in a spin-glass-like state. Our theoretical results show unambiguously that the Fe atoms with the square pyramidal environment has a lower oxidation state than that in the octahedral co-ordination. However, the presence of covalent interaction between Fe and the neighboring oxygen atoms makes the actual oxidation state considerably smaller than the formal oxidation state of 3+ and 4+ for square-pyramidal and octahedral co-ordination, respectively. $Sr_4Fe_4O_{11}$ is found to be semiconducting in the antiferromagnetic ground state.

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doi: 10.1103/PhysRevB.74.054422
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First-principles investigations of the MMgH$_3$ (M = Li, Na, K, Rb, Cs) series

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Abstract

The structural stability of the MMgH$_3$ (M = Li, Na, K, Rb, Cs) series has been investigated using the density-functional projector-augmented-wave method within the generalized-gradient approximation. Among the 24 structural arrangements used as inputs for the structural optimization calculations, the experimentally known frameworks are successfully reproduced, and positional and unit-cell parameters are found to be in good agreement with the experimental findings. The crystal structure of LiMgH$_3$ has been predicted, the most stable arrangement being the trigonal LiTaO$_3$-type ($R3c$) structure, which contains highly distorted octahedra. The formation energy for all members of the MMgH$_3$ series is investigated along different reaction pathways. The electronic density of states reveals that the MMgH$_3$ compounds are wide-band-gap insulators. Analyses of chemical bonding in terms of charge density, charge transfer, electron-localization function, Born effective charge, and Mulliken population show that these hydrides are basically saline hydrides similar to the parent alkali-/alkaline-earth mono-/di-hydrides.

(J. Alloys Compd. (2006), doi:10.1016/j.jallcom.2006.10.163 (2007).)
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A new series of high hydrogen content hydrogen-storage materials - a theoretical prediction

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Abstract

The structural phase stability of $NaBeH_3$, $NaMgH_3$, and $Ca(BH_4)_2$ has been investigated using density-functional projector-augmented-wave method within the generalized-gradient approximation. Among the various structural arrangements used as inputs for the structural optimization calculations, the experimentally known frameworks are successfully reproduced for $NaMgH_3$ and the positional and unit-cell parameters are found to be in good agreement with the experimental finding. The crystal structure of $NaBeH_3$ and $Ca(BH_4)_2$ has been predicted. The electronic density of states reveal that all the considered compounds are wide-band-gap insulators.

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Structural phase stability studies on MBeH$_3$ (M = Li, Na, K, Rb, Cs) from density functional calculations

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Abstract

Density-functional-theory calculations within the generalized-gradient approximation are used to establish the ground-state structure, equilibrium structural parameters, and electronic structure for $MBeH_3$ phases. Among 24 structural arrangements used as inputs for structural optimization calculations, the crystal structures of $MBeH_3$ phases have been predicted. At ambient condition $LiBeH_3$ and $NaBeH_3$ crystallize in perovskite related orthorhombic and cubic structures, respectively. The remaining phases $KBeH_3$, $RbBeH_3$ and $CsBeH_3$ crystalize in a monoclinic structure. In the predicted phases one can store up to 15.93 wt.% of hydrogen. The formation energy for the $MBeH_3$ phases have been investigated along different reaction pathways. The electronic structures reveal that all these phases are insulators with estimated band gap varying between 1.79 and 3.44 eV.

(Submitted for publication in Phys. Rev. B (2007).)
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Origin of magnetoelectric behavior in $BiFeO_3$

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Abstract

The magnetoelectric behavior of $BiFeO_3$ has been explored on the basis of accurate density functional calculations. We are able to predict structural, electronic, magnetic, and ferroelectric properties of $BiFeO_3$ correctly without including any strong correlation effect in the calculation. Unlike earlier calculations, the equilibrium structural parameters are found to be in very good agreement with the experimental findings. In particular, the present calculation correctly reproduced experimentally-observed elongation of cubic perovskite-like lattice along the [111] direction. At high pressure we predicted a pressure-induced structural transition from rhombohedral ($R3c$) to an orthorhombic ($Pnma$) structure. The total energy calculations at expanded lattice show two lower energy ferroelectric phases (with monoclinic Cm and tetragonal $P4mm$ structures), closer in energy to the ground state phase. Spin-polarized band-structure calculations show that $BiFeO_3$ will be an insulator in A- and G-type antiferromagnetic phases and a metal in C-type antiferromagnetic, ferromagnetic configurations, and in the nonmagnetic state. Chemical bonding in $BiFeO_3$ has been analyzed using partial density-of-states, charge density, charge transfer, electron localization function, Born-effective-charge tensor, and crystal orbital Hamiltonian population analyses. Our electron localization function analysis shows that stereochemically active lone-pair electrons are present at the Bi sites which are responsible for displacements of the Bi atoms from the centro-symmetric to the noncentrosymmetric structure and hence the ferroelectricity. A large ferroelectric polarization of $88.7\,\mu C/cm^2$ is predicted in accordance with recent experimental findings, but differing by an order of magnitude from earlier experimental values. The strong spontaneous polarization is related to the large values of the Born-effective-charges at the Bi sites along with their large displacement along the [111] direction of the cubic perovskite-type reference structure. Our polarization analysis shows that partial contributions to polarization from the Fe and O atoms almost cancel each other and the net polarization present in $BiFeO_3$ mainly ($> 98\%$) originates from Bi atoms. We found that the large scatter in experimentally reported polarization values in $BiFeO_3$ is associated with the large anisotropy in the spontaneous polarization.

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DOI: 10.1103/PhysRevB.74.224412
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Self-interaction in Green's-function theory of the hydrogen atom

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Abstract

Atomic hydrogen provides a unique test case for computational electronic structure methods, since its electronic excitation energies are known analytically. With only one electron, hydrogen contains no electronic correlation and is therefore particularly susceptible to spurious self-interaction errors introduced by certain computational methods. In this paper we focus on many-body perturbation-theory (MBPT) in Hedin's *GW* approximation. While the Hartree-Fock and the exact MBPT self-energy are free of self-interaction, the correlation part of the *GW* self-energy does not have this property. Here we use atomic hydrogen as a benchmark system for *GW* and show that the self-interaction part of the *GW* self-energy, while non-zero, is small. The effect of calculating the *GW* self-energy from exact wavefunctions and eigenvalues, as distinct from those from the local-density approximation, is also illuminating.

(submitted to Phys. Rev. A)
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Ab initio study of the half-metal to metal transition in strained magnetite

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Abstract

Using density-functional theory, we investigate the stability of the half-metallic ground state of magnetite under different strain conditions. The effects of volume relaxation and internal degrees of freedom are fully taken into account. For hydrostatic compression, planar strain in the (001) plane and uniaxial strain along the [001] direction we derive quantitative limits beyond which magnetite becomes metallic. As a major new result, we identify the bond length between the octahedrally coordinated iron atoms and their neighbouring oxygen atoms as the main characteristic parameter, and we show that the transition occurs if external strain reduces this interatomic distance from 2.06 Å in equilibrium to below a critical value of 1.99 Å. Based on this criterion we also argue that planar strain due to epitaxial growth does not lead to a metallic state for magnetite films grown on (111)-oriented substrates.

(published: New Journal of Physics **9**, 5 (2007))
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Ab initio calculations of transport properties of epitaxial (Ga,Mn)As systems

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Abstract

We calculate transport properties of thin (Ga, Mn)As layers sandwiched between two Cr (001) leads as a function of Mn concentration and layer thickness, special attention is paid to the spin polarization of the current-perpendicular-to-plane (CPP) conductance. We compare our results to bulk properties. Doping GaAs with Mn increases occurrence of As antisites on the Ga sublattice. Our method allows us to take into account even very small defect concentration changes, and we show that it may have a dramatic impact on the CPP spin polarization, apart from its already known influence on Curie temperatures.

(Accepted for publication in J. Magn. Magn. Mat.)
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Spin-orbital physics emerging from interacting O_2 molecules in KO_2

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Abstract

The alkali hyperoxide KO_2 is a molecular analog of strongly-correlated systems, comprising of orbitally degenerate magnetic O_2^- ions. Using first-principles electronic structure calculations, we set up an effective spin-orbital model for the low-energy *molecular* orbitals and argue that many anomalous properties of KO_2 replicate the status of its orbital system in various temperature regimes.

Manuscript available from Solovyev.Igor@nims.go.jp or directly from cond-mat/0612475.

Ruby, metals, and MgO as alternative pressure scales: A semiempirical description of shock-wave, ultrasonic, x-ray, and thermochemical data at high temperatures and pressures

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Abstract

We have constructed semiempirical equations of state of Al, Au, Cu, Pt, Ta, and W, which within experimental error bars describe the available shock-wave, ultrasonic, x-ray, and thermochemical data in the temperature range from 10-20 K up to the melting temperature and to compression x=V/V0=0.5-0.7. The comparison of the calculated room-temperature isotherms for these metals with quasihydrostatic measurements supports recently proposed ruby pressure scales. We recommend a new ruby pressure scale calibration. The cross check on independent data confirms the obtained PVT equations of state of Ag, Al, Au, Cu, Pt, Ta, W, MgO, and diamond. The equations of state of these materials obtained here provide accurate and versatile means for calibrating pressure at all temperatures below the melting point. Furthermore, they can be used for accurate tabulation of thermodynamic properties (heat capacities, entropies) of these reference substances in a wide P-T range.

(Phys. Rev. B75, art. 024115 (2007))

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Crystal structure transformations in SiO_2 from classical and ab initio metadynamics

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Abstract

Silica is the main component of the Earth's crust and is also of great relevance in many branches of materials science and technology. Its phase diagram is rather intricate and exhibits many different crystalline phases. The reported propensity to amorphization and the strong influence on the outcome of the initial structure and of the pressurization protocol indicate the presence of metastability and large kinetic barriers. As a consequence, theory is also faced with great difficulties and our understanding of the complex transformation mechanisms is still very sketchy despite a large number of simulations. Here, we introduce a substantial improvement of the metadynamics method, which finally brings simulations in close agreement with experiments. We unveil the subtle and non-intuitive stepwise mechanism of the pressure-induced transformation of fourfold-coordinated a-quartz into sixfold-coordinated stishovite at room temperature. We also predict that on compression fourfoldcoordinated coesite will transform into the post-stishovite a-PbO_2-type phase. The new method is far more efficient than previous methods, and for the first time the study of complex structural phase transitions with many intermediates is within the reach of molecular dynamics simulations. This insight will help in designing new experimental protocols capable of steering the system towards the desired transition.

(Nature Materials 5, 623-626 (2006))
Manuscripts available from: Prof. Artem R. Oganov (a.oganov@mat.ethz.ch)

Valence and spin states of iron impurities in mantle-forming silicates

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Abstract

Using ab initio simulations, we investigated the valence and spin states of iron impurities in the perovskite (Pv) and postperovskite (PPv) polymorphs of $MgSiO_3$. In agreement with the previous experimental work, we find a valence disproportionation reaction: $3Fe^{2+}=2Fe^{3+}Fe$ (metal). This exothermic reaction results in the predominance of Fe^{3+} impurities in lower mantle silicates and produces free metallic iron. It occurs both in Pv and PPv, Al-free and Al-rich, at all lower mantle pressures. This reaction provides a possible mechanism for the growth of the Earth's core and core-mantle chemical equilibration. In the presence of Al^{3+}, iron forms Fe^{3+}-Al^{3+} coupled substitutions in Pv, but separate Fe^{3+}-Fe^{3+} and Al^{3+}-Al^{3+} substitutions in PPv. Only the high-spin state is found for Fe2+ impurities at all mantle pressures, while Fe^{3+} impurities on the Si-site are low-spin at all pressures in both phases. Fe^{3+} impurities on the Mg-site are in the high-spin state in PPv at all mantle pressures, but in Pv we predict a high-spin - low-spin transition. The pressure at which this transition occurs strongly depends on the Al^{3+} content and according to our calculations increases from 76 GPa for Al-free to 134 GPa for aluminous Pv; this reconciles many of the previous experimental results. Our findings have implications for the chemical evolution of the Earth and for the radiative conductivity and dynamics of the D" layer.

(Earth Planet. Sci. Lett. 249, 436-443)
Contact person: Prof. Artem R. Oganov (a.oganov@mat.ethz.ch)

Crystal structure prediction using evolutionary algorithms: principles and applications

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ETH Zurich, Wolfgang-Pauli-Strasse 10, Zurich 8093, Switzerland

Abstract

We have developed an efficient and reliable methodology for crystal structure prediction, merging ab initio total-energy calculations and a specifically devised evolutionary algorithm. This method allows one to predict the most stable crystal structure and a number of low-energy metastable structures for a given compound at any P-T conditions without requiring any experimental input. Extremely high (nearly 100%) success rate has been observed in a few tens of tests done so far, including ionic, covalent, metallic, and molecular structures with up to 20 atoms in the unit cell. We have been able to resolve some important problems in high-pressure crystallography and report a number of new high-pressure crystal structures. Physical reasons for the success of this methodology are discussed.

(J. Chem. Phys. 124, art. 244704 (2006))
Contact person: Prof. Artem R. Oganov (a.oganov@mat.ethz.ch)

USPEX - evolutionary crystal structure prediction

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Abstract

We approach the problem of computational crystal structure prediction, implementing an evolutionary algorithm USPEX (Universal Structure Predictor: Evolutionary Xtallography). Starting from chemical composition we have tested USPEX on numerous systems (with up to 80 atoms in the unit cell) for which the stable structure is known and have observed a success rate of nearly 100%, simultaneously finding large sets of competitive metastable structures. Here focus is on implementation and discussion of our method.

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7 SCIENTIFIC HIGHLIGHT OF THE MONTH: "Exciting prospects for solids: Exact-exchange based functionals meet quasiparticle energy calculations"

Exciting prospects for solids: Exact-exchange based functionals meet quasiparticle energy calculations

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Abstract

Focussing on spectroscopic aspects of semiconductors and insulators we will illustrate how quasiparticle energy calculations in the G_0W_0 approximation can be successfully combined with density-functional theory calculations in the exact-exchange optimised effective potential approach (OEPx) to achieve a first principles description of the electronic structure that overcomes the limitations of local or gradiant corrected DFT functionals (LDA and GGA).

Contents

1 Introduction

Density functional theory (DFT) has contributed significantly to our present understanding of a wide range of materials and their properties. As quantum-mechanical theory of the density and the total energy it provides an atomistic description from first principles and is, in the local-density or generalized gradient approximation (LDA and GGA), applicable to polyatomic systems containing up to several thousand atoms. However, a combination of three factors limits the applicability of LDA and GGA to a range of important materials and interesting phenomena. They are approximate (jellium-based) exchange-correlation functionals, which suffer from incomplete cancellation of artificial self-interaction and lack the discontinuity of the exchange-correlation potential with respect to the number of electrons. As a consequence the Kohn-Sham (KS) single-particle eigenvalue band gap for semiconductors and insulators underestimates the quasiparticle band gap as measured by the difference of ionisation energy (via photoemission spectroscopy (PES)) and electron affinity (via inverse PES (IPES)). This reduces the predictive power for materials whose band gap is not know from experiment and poses a problem for calculations where the value of the band gap is of importance, e.g. the energies of surface and defect states. Moreover, the incomplete cancellation of the self-interaction questions the applicability to systems with localized defect states, strong magnetic interactions or materials with localized d or f states, such as II-VI compounds, group-III-nitrides, transition metal oxides or lanthanides, actinides and their compounds. It is therefore desirable to have a more advanced electronic structure approach that overcomes these shortcomings of LDA/GGA and provides a reliable description of ground state total energies as well as electronic excitation spectra.

Many-body perturbation theory in the GW approach [1] has developed into the method of choice for describing the energies of electronic excitation spectra of solids (in the following referred to as quasiparticle energies) [2, 3]. For conceptual as well as computational reasons, however, it is currently not feasible to compute total energies for systems of interest. Exact-exchange based DFT functionals on the other hand give access to total energies and thus atomic structures. In addition they largely or in certain cases completely remove the self-interaction error. Exact-exchange in the optimized effective potential (OEP) approach (OEPx or EXX or OEPx(cLDA) when LDA correlation is added (see also Section 3)) has become the most prominent approach of this kind. It is self-interaction free and greatly improves the comparison of KS eigenvalue differences with quasiparticle excitations for solids, with respect to Hartree-Fock [4–15].

Alternatively, the removal of the self-interaction can be approached by applying the exchange-operator in a non-local fashion within the framework of the generalized Kohn-Sham (GKS) scheme [16] or by using other non-local schemes such as the self-interaction corrected LDA SIC-LDA) method [17]. In the former case the bare exchange interaction is empirically screened and combined with a local potential (commonly referred to as hybrid functional). The most widely used hybrid functionals are PBE0 [18–20], B3LYP [21–23], HSE [24–26], screened exchange (sX-LDA) [16, 27]. With the exception of SIC and sX-LDA hybrid functionals are only slowly being applied to solids. Almost no experience exist in their combination with GW calculations and we will thus defer a discussion of their properties to the outlook.

Combining exact-exchange based DFT calculations with quasiparticle energy calculations [10, 11, 28–30] offers several advantages and will be an important step towards reaching a thorough understanding of the aforementioned systems. Up to now most GW calculations are performed non self-consistently as a single perturbation to an LDA ground state. Unlike the Kohn-Sham scheme in DFT (see Section 3.1) the GW approach in MBPT is not governed by a closed set of equations. Iterating beyond the first order (G_0W_0) in the exact theory would introduce higher order interactions at every step that lead beyond the GW approximation, while iterating the set of GW equations ignoring these contributions generally worsens the good agreement achieved after the first iteration (A more detail account of self-consistency in GW will be given in Section 2.2). ¿From the view point of perturbation theory it would thus be advantageous to use OEPx

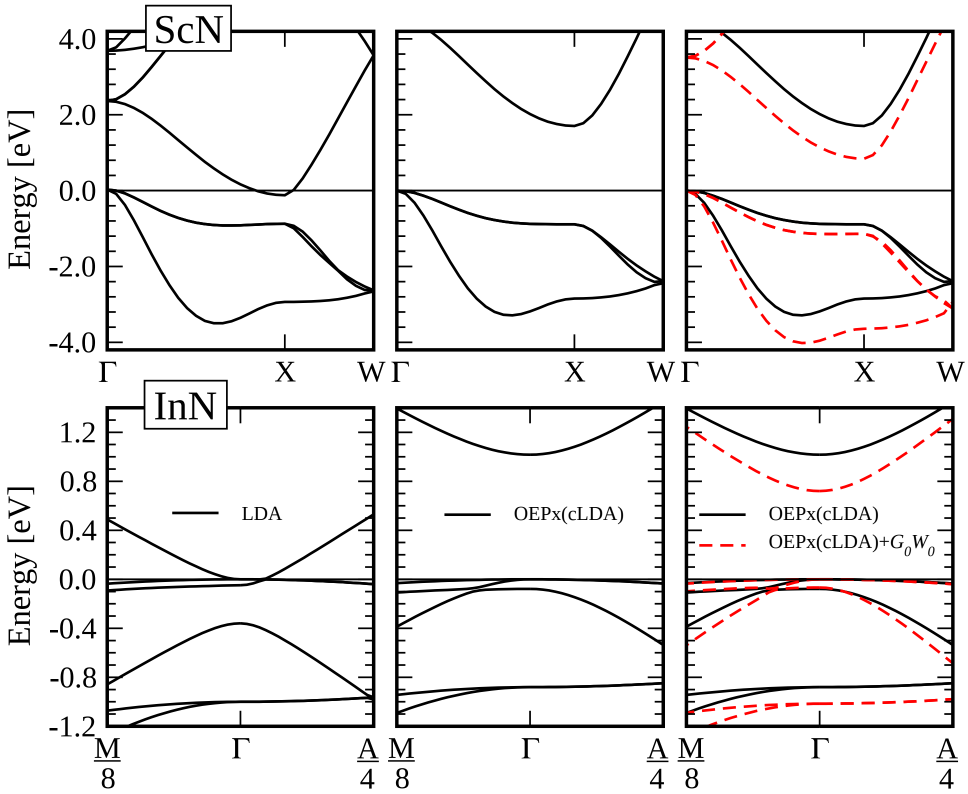

Figure 1: LDA KS calculations incorrectly predict wurtzite InN (bottom) to be a metal with the wrong band ordering at the Γ point. ScN (top) is erroneously described as semimetal. In OEPx(cLDA) the band gap opens and InN and ScN correctly become semiconductors, thus providing a more suitable starting point for subsequent quasiparticle energy calculations in the G_0W_0 approximation. All calculations are performed at the experimental lattice constants.

instead of the LDA[1] as a starting point for G_0W_0 calculations, because the corresponding Kohn-Sham spectrum is closer to the quasiparticle spectrum, thus requiring a smaller correction. This becomes particularly important for materials where LDA severely underestimates the band gap like in ZnO (by ≈80% for the wurtzite phase, LDA (at exp. lattice const.): 0.7 eV, exp: 3.4 eV) or incorrectly predicts a (semi)metallic state as e.g. in germanium (Ge), indium nitride (InN) and scandium nitride (ScN) (see Fig. 1 and 3).

In this Ψ_k-highlight we will focus only on spectroscopic aspects of semiconductors and insulators and illustrate how quasiparticle energy calculations can be successfully combined with OEPx(cLDA) calculations to achieve a first principles description of the electronic structure that overcomes the limitations of LDA and GGA. Figure 1 summarises the structure and the message of this paper. For the example of InN and ScN we demonstrate how OEPx(cLDA) opens the band gap and correctly predicts them to be semiconductors thus providing a better starting point for subsequent G_0W_0 calculations. The quasiparticle corrections to the OEPx(cLDA) band structure are considerable – also for Gallium nitride (GaN) and selected II-VI compounds – bringing the quasiparticle band structure into good agreement with direct and inverse pho-

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[1] All G_0W_0 calculations in this work are performed at the experimental lattice constants.

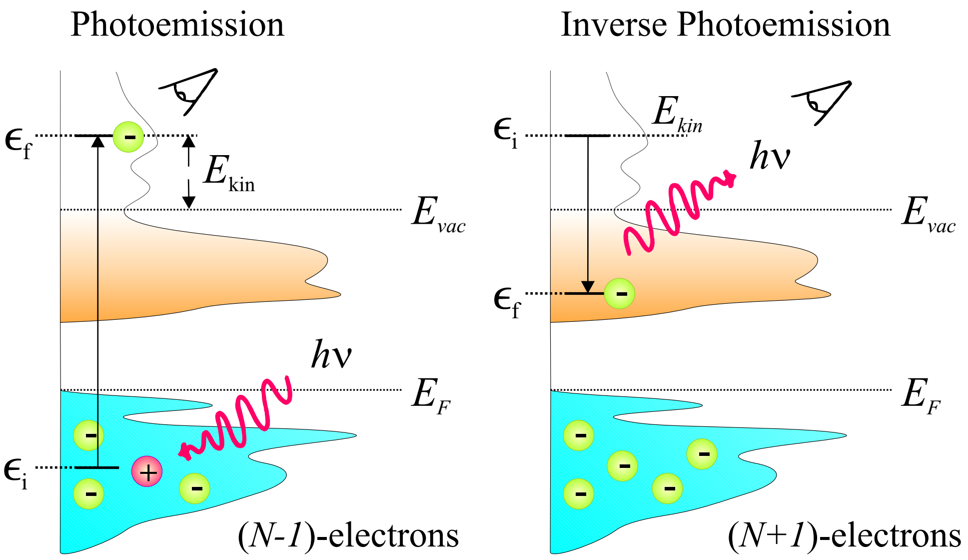

Figure 2: Schematic of the photoemission (PES) and inverse photoemission (IPES) process. In PES (left) an electron is excited from an occupied valence state (lower shaded region) into the continuum (white region) starting above the vacuum level E_{vac} by an incoming photon. In IPES (right) an injected electron with kinetic energy $\epsilon_i = E_{kin}$ undergoes a radiative transition into an unoccupied state (upper shaded region) thus emitting a photon in the process.

toemission data. Our OEPx based G_0W_0 calculations have helped to identify the band gap of InN and ScN and to clarify the source for the puzzling, wide interval of experimentally observed band gaps [29, 30].

The article is structured as follows. In Section 2.2 we will briefly review the connection between the single particle Green's function and photo-electron spectroscopy and then introduce the GW method. The exact-exchange approach will be described in Section 3. In Section 4 we will demonstrate why and how the combination of OEPx(cLDA) and GW provides an improved description of the electronic structure. Finally we will conclude in Section 6 and give an outlook in Section 7.

2 Quasiparticle energy calculations

2.1 Photo-electron spectroscopy and the quasiparticle concept

In photo-electron spectroscopy (PES) [31–33] electrons are ejected from a sample upon irradiation with visible or ultraviolet light (UPS) or with X-rays (XPS), as sketched in the left panel of Fig. 2. Energies $\epsilon_i = h\nu - E_{kin}$ and lifetimes of holes can be reconstructed from the photon energy $h\nu$ and the kinetic energy E_{kin} of the photoelectrons that reach the detector[2]. By inverting the photoemission process, as schematically shown in the right panel of Fig. 2, the unoccupied states can be probed. An incident electron with energy E_{kin} is scattered in the

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[2] Throughout this article the top of the valence bands is chosen as energy zero.

sample emitting *bremsstrahlung.* Eventually it will undergo a radiative transition into a lower-lying unoccupied state, emitting a photon that carries the transition energy $h\nu$. The energy of the final, unoccupied state can be deduced from the measured photon energy according to $\epsilon_f = E_{kin} - h\nu$. This technique is commonly referred to as inverse photoemission spectroscopy (IPES) or bremsstrahlung isochromat spectroscopy (BIS) [34–36].

The experimental observable in photoemission spectroscopy is the photocurrent. Since the energy dependence of the transition matrix elements is usually weak and smooth, structures in the photoemission spectrum can be associated with features in the density of states (DOS), i.e. the imaginary part of the one-particle Green's function[3] [3,37]

$$A(\mathbf{r},\mathbf{r}';\epsilon) = \frac{1}{\pi}\mathrm{Im}\, G(\mathbf{r},\mathbf{r}';\epsilon). \tag{1}$$

Peaks due to shake ups and shake downs found in XPS are not described by the GW approximation and will therefore not be addressed here.

The Green's function is the solution to the many-body Hamiltonian

$$\hat{H}(\mathbf{r},\mathbf{r}';\epsilon) = \hat{h}_0(\mathbf{r}) + \Sigma(\mathbf{r},\mathbf{r}';\epsilon) \tag{2}$$

written here in single-particle form where all electron-electron interaction terms are rolled up in the non-local, energy dependent self-energy Σ. The remaining contributions are given by[4] $\hat{h}_0(\mathbf{r}) = -\frac{1}{2}\nabla^2 + v_{ext}(\mathbf{r})$. The external potential v_{ext} is due to the nuclei, after the Born-Oppenheimer approximation of stationary nuclei is taken. The photocurrent is then the surface weighted integral over the diagonal part of the spectral function $A(\mathbf{r},\mathbf{r}';\epsilon)$. We note, however, that with respect to the measured intensities a photoemission spectrum is a noticeably distorted spectral function that in addition is weighted over the momentum components normal to the surface ($\mathbf{k}_\perp$). In particular when selection rules and the energy dependence of the transition matrix elements become important certain peaks in the spectral function may be significantly reduced or may even disappear completely.

The excitation of a non-interacting or a bare particle would give rise to a delta peak in the spectral function. When the electron-electron interaction is turned on, the electrons can no longer be regarded as independent particles. As a consequence the matrix elements of the spectral function $A_{n\mathbf{k}}(\epsilon)$ will contain contributions from many non-vanishing transition amplitudes. If these contributions merge into a clearly identifiable peak that appears to be derived from a single delta-peak broadened by the electron-electron interaction

$$A_{n\mathbf{k}}(\epsilon) \approx \frac{Z_{n\mathbf{k}}}{\epsilon - (\epsilon_{n\mathbf{k}} + i\Gamma_{n\mathbf{k}})} \tag{3}$$

this structure can be interpreted as single-particle like excitation – the *quasiparticle*[5]. The broadening of the quasiparticle peak in the spectral function is associated with the lifetime $\tau_{n\mathbf{k}} =$

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[3] The $\mathbf{r}$ and $\mathbf{r}'$ dependence can easily be transformed into a $\mathbf{k}$ dependence. Furthermore only the spin unpolarised situation is discussed here. For the present discussion a summation over the spin indices in the Green's function is therefore assumed.

[4] Atomic units $4\pi\epsilon_0 = h = e = m_e = 1$, where e and m_e are the charge and mass of an electron, respectively, will be used in the remainder of this article. If not otherwise stated energies are measured in hartrees and length in bohr.

[5] A *quasielectron* is a new quantum mechanical entity that combines an electron with its surrounding polarisation cloud. The same concept applies to holes.

$2/\Gamma_{n\mathbf{k}}$ of the excitation due to electron-electron scattering, whereas the area underneath the peak is interpreted as the renormalisation $Z_{n\mathbf{k}}$ of the quasiparticle. This renormalisation factor quantifies the reduction in spectral weight due to electron-electron exchange and correlation effects compared to an independent electron.

A computational description of the quasiparticle band structure thus requires the calculation of the Green's function and the self-energy.

2.2 The *GW* formalism

An exact solution to equation (2) is given by Hedin's set of coupled integro-differential equations [1]

$$P(1,2) = -i \int G(1,3)G(4,1)\Gamma(3,4,2)d(3,4) \tag{4}$$

$$W(1,2) = v(1,2) + \int v(1,3)P(3,4)W(4,2)d(3,4) \tag{5}$$

$$\Sigma(1,2) = i \int G(1,3)\Gamma(3,2,4)W(4,1)d(3,4) \tag{6}$$

$$\Gamma(1,2,3) = \delta(1,2)\delta(1,3) + \int \frac{\delta\Sigma(1,2)}{\delta G(4,5)} G(4,6)G(7,5)\Gamma(6,7,3)d(4,5,6,7) \tag{7}$$

where the notation $1 \equiv (\mathbf{r}_1, t_1, \sigma_1)$ is used to denote a triple of space, time and spin variables[6]. Accordingly $\int d(1)$ is a shorthand notation for the integration in all three variables of the triple. In eq. 4-7 P is the polarisability, W the screened and v the bare Coulomb interaction and Γ the so called vertex function. By means of Dyson's equation

$$G^{-1}(1,2) = G_0^{-1}(1,2) - \Sigma(1,2), \tag{8}$$

which links the non-interacting system with Green's function G_0 to the fully interacting one (G) via the self-energy Σ, Hedin's equations could in principle be solved self-consistently starting from a given G_0. However, the functional derivative in eq. (7) does not permit a numeric solution, requiring approximations for the vertex function in practise.

Starting from a non-interacting system the initial self-energy is zero and the vertex function becomes $\Gamma(1,2,3) = \delta(1,2)\delta(1,3)$. The first iteration yields Hedin's GW approximation for the self-energy as a function of energy

$$\Sigma_{\mathrm{GW}}(\mathbf{r},\mathbf{r}';\epsilon) = \frac{i}{2\pi} \int_{-\infty}^{\infty} d\epsilon' e^{i\epsilon'\delta} G(\mathbf{r},\mathbf{r}';\epsilon+\epsilon')W(\mathbf{r},\mathbf{r}';\epsilon') \tag{9}$$

where δ is an infinitesimal, positive time. The physical interpretation of the self-energy corresponds to the energy contribution of the quasiparticle that arises from the response of the system due to quasiparticle's own presence. The interaction of the electrons in the system is mediated by the screened Coulomb interaction rather than the bare one. In the GW formalism W (eq. 5) is calculated in the Random-Phase approximation (i.e. quasiholes and quasielectrons

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[6] Throughout this article we will only consider systems without any explicit spin dependence and will therefore omit the spin index in the following.

do not interact with each other) from the irreducible polarizability[7] (eq. 4). For the zeroth order Green's function G_0 a Kohn-Sham Green's function G_{KS}

$$G_0(\mathbf{r},\mathbf{r}',\epsilon) = G_{\mathrm{KS}}(\mathbf{r},\mathbf{r}',\epsilon) = \lim_{\delta\to 0^+}\sum_{n\mathbf{k}} \frac{\phi_{n\mathbf{k}}(\mathbf{r})\phi^*_{n\mathbf{k}}(\mathbf{r}')}{\epsilon - (\epsilon_{n\mathbf{k}} + i\delta\,\mathrm{sgn}(E_{\mathrm{F}} - \epsilon_{n\mathbf{k}}))} \tag{10}$$

is typically taken where E_{F} defines the Fermi level.

Keeping Γ fixed (to delta functions) removes the functional derivative from eq. 4-7. These *GW* equations could then in principle be solved self-consistently via Dyson's equation (8). However this issue is still a matter of debate [38–42]. Unlike in DFT, a self-consistent solution of the full set of Hedin's equations would go beyond the *GW* approximation and successively introduce higher order electron-electron interactions through the vertex function with every iteration step. Solving the *GW* equations self-consistently is therefore inconsistent if no higher order electron-electron interactions are included. It was first observed for the homogeneous electron gas [43] that the spectral features broaden with increasing number of iterations in the self-consistency cycle. Similarly, for closed shell atoms the good agreement with experiment for the ionization energy after the first iteration is lost upon iterating the equations to self-consistency [40]. Imposing self-consistency in an approximate fashion [42, 44–46] is not unique and different methods yield different results. Since the controversies regarding self-consistency within *GW* have not been resolved conclusively, yet, the majority of all *GW* calculations is performed employing the zeroth order in the self-energy (G_0W_0). This, however, introduces a dependence of the starting Green's function on the self-energy and thus the quasiparticle energies, which can have a noticeable influence, as we will demonstrate in this article.

To obtain the quasiparticle band structure we solve the quasiparticle equation[8]

$$\left[-\frac{\nabla^2}{2} + v_{ext}(\mathbf{r}) + v_H(\mathbf{r})\right]\psi_{n\mathbf{q}}(\mathbf{r}) + \int d\mathbf{r}'\Sigma(\mathbf{r},\mathbf{r}';\epsilon_{n\mathbf{q}})\psi_{n\mathbf{q}}(\mathbf{r}') = \epsilon_{n\mathbf{q}}\psi_{n\mathbf{q}}(\mathbf{r}) \tag{11}$$

with the *GW* self-energy ($\Sigma = \Sigma_{\mathrm{GW}}$) by a approximating the quasiparticle wavefunctions ($\psi_{n\mathbf{q}}$) with the Kohn-Sham ones ($\phi_{n\mathbf{q}}$) [47]. The corrections to the Kohn-Sham eigenvalues ($\epsilon^{\mathrm{KS}}_{n\mathbf{q}}$) are then give by

$$\epsilon^{qp}_{n\mathbf{q}} = \epsilon^{\mathrm{KS}}_{n\mathbf{q}} + \langle\phi_{n\mathbf{q}}|\Sigma_{\mathrm{GW}}(\epsilon^{qp}_{n\mathbf{q}}) - v_{xc}|\phi_{n\mathbf{q}}\rangle \tag{12}$$

where $\langle\phi_{n\mathbf{q}}||\phi_{n\mathbf{q}}\rangle$ denotes matrix elements with respect to the wavefunctions $\phi_{n\mathbf{q}}$ of the preceding Kohn-Sham DFT calculation with exchange-correlation potential v_{xc}.

3 Exact-exchange based DFT

3.1 DFT and the Kohn-Sham band structure

With regard to quasiparticle band structures Kohn-Sham DFT calculations not only serve as a starting point for G_0W_0 calculations, they are frequently used to interpret quasiparticle spectra due to the similarity of the Kohn-Sham

$$\left[-\frac{\nabla^2}{2} + v_{eff}[n(\mathbf{r})](\mathbf{r})\right]\phi_i(\mathbf{r}) = \epsilon_i\phi_i(\mathbf{r}) \tag{13}$$

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[7] With $\Gamma(1,2,3) = \delta(1,2)\delta(1,3)$ eq. 4 reduces to the much simpler form $\chi_0 = -iGG$.

[8] Note that the Hartree potential $v_H(\mathbf{r})$ defined in the next Section has been separated from the self-energy.

with the quasiparticle equation (11) and because the correction can often be expressed in terms of first-order perturbation theory (eq. (12)). To illustrate the difference between the Kohn-Sham and the quasiparticle picture and to lay the foundations for the discussion of the exact-exchange OEP approach we will briefly review the Kohn-Sham DFT scheme.

The central quantities in DFT are the electron density $n(\mathbf{r})$ and the total energy E_{tot}. The latter is a functional of the former and attains its minimum at the exact ground state density, as proven by Hohenberg and Kohn [48]. This formalism was turned into a tractable computational scheme by Kohn and Sham [49], by assuming that the system of interacting particles can be mapped onto a fictitious system of non-interacting particles moving in an effective local potential $v_{eff}(\mathbf{r})$ that reproduce the same density as the many-body problem of interest. The electron density

$$n(\mathbf{r}) = \sum_i^{occ} |\phi_i(\mathbf{r})|^2 \tag{14}$$

is composed of the occupied Kohn-Sham orbitals $\phi_i(\mathbf{r})$ that are solutions of the Kohn-Sham equation 13.

In analogy to Hartree theory Kohn and Sham divided the total energy into known contributions such as the kinetic energy of the non-interacting particles T_s, the Hartree energy

$$E_H[n] = \frac{1}{2}\int d\mathbf{r}\, n(\mathbf{r})v_H(\mathbf{r}) = \frac{1}{2}\sum_{ij}^{occ}\iint d\mathbf{r}\, d\mathbf{r}'\, \frac{\phi_i^*(\mathbf{r})\phi_i(\mathbf{r})\phi_j^*(\mathbf{r}')\phi_j(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} \quad , \tag{15}$$

the external energy

$$E_{ext}[n] = \int d\mathbf{r}\, n(\mathbf{r})v_{ext}(\mathbf{r}) \quad , \tag{16}$$

and an unknown remainder. This last term includes all electron-electron interactions beyond the Hartree mean-field and is defined as the exchange-correlation energy

$$E_{xc}[n] = E_{tot}[n] - T_s[n] - E_{ext}[n] - E_H[n] \quad . \tag{17}$$

Performing the variation with respect to the density then yields the effective potential

$$v_{eff}[n](\mathbf{r}) = v_{ext}(\mathbf{r}) + v_H[n](\mathbf{r}) + v_{xc}[n](\mathbf{r}) \quad , \tag{18}$$

where each term in the sum is obtained as functional derivative of the corresponding energy expression. Since the exact form of the exchange-correlation functional is unknown[9] suitable approximations have to be found in practice.

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[9] To be more precise: according to the Hohenberg-Kohn theorem E_{xc} is a unique functional of the density. However, this does not necessarily imply that E_{xc} can be written in a closed mathematical form as functional of the density. Analogous examples are T_e (the kinetic energy of interacting electrons) and T_s. For the latter one may write down a series expansion, but this does not converge properly. Its evaluation therefore requires the detour via the Kohn-Sham formalism. In fact this exact evaluation of T_s is the strongest reason for using the Kohn-Sham scheme. As far as the exact v_{xc} is concerned it can be expressed in terms of the Green's function and the self-energy via the Sham-Schlüter equation in the context of many-body perturbation theory. Alternatively an exact representation of v_{xc} can be obtained in Görling-Levy perturbation theory [50].

3.2 LDA and self-interaction

Probably the most popular and efficient approximation for the exchange-correlation energy to-date is the local-density approximation (LDA) [49]

$$E_{xc}^{\mathrm{LDA}}[n] = \int d\mathbf{r}\; n(\mathbf{r})\epsilon_{xc}^{\mathrm{HEG}}(n)\big|_{n=n(\mathbf{r})} \tag{19}$$

where $\epsilon_{xc}^{\mathrm{HEG}}(n(\mathbf{r}))$ is the exchange-correlation energy density of the homogeneous electron gas (HEG). Whilst the LDA[10] describes even inhomogeneous systems with startling success in many cases, it does so at the expense of a non-physical electron self-interaction. This is introduced by the Hartree term, that contains a spurious interaction of an electron with itself since the sum in eq. 15 includes all occupied states. Although the LDA fulfills the sum rule and thus correctly removes the self-interaction with respect to the particle number, the shape of the exchange-correlation hole is not correct[11]. Applying the definition of Perdew and Zunger [17] the ensuing self-interaction error can be quantified for every state as

$$\delta_i = \frac{1}{2}\iint d\mathbf{r}\, d\mathbf{r}' \frac{|\phi_i(\mathbf{r})|^2|\phi_i(\mathbf{r}')|^2}{|\mathbf{r}-\mathbf{r}'|} + E_{xc}[|\phi_i(\mathbf{r})|^2], \tag{20}$$

which follows directly from eq. 14 and 15 for $i = j$. The self-interaction error is largest for localised states and has a tendency to delocalise the electronic wavefunction [28, 52], a point that we will return to later.

Perdew and Zunger proposed to subtract the sum over all self-interaction contributions δ_i for all occupied states from the expression for the total energy in the LDA (or alternatively the spin-dependent LDA) [17]

$$E_{xc}^{\mathrm{SIC-LDA}}[n] = E_{xc}^{\mathrm{LDA}}[n] - \sum_i^{occ} \delta_i \quad . \tag{21}$$

The expression for the self-interaction corrected LDA (SIC-LDA) total energy can be minimised according to the variational principle. However, since the energy functional is now orbital dependent, the computational simplicity of the LDA is lost, making SIC-LDA calculations computationally much more demanding than LDA or GGA calculations. Moreover, this form of self-interaction correction vanishes for completely delocalised states, which makes a direct application of this formalism to Bloch states in solids difficult.

It is this self-interaction error in the LDA that is responsible for the fact that InN, ScN and also Ge are incorrectly predicted to be (semi)metals (cf Fig. 1 and Fig. 3) and that the band gap of GaN and II-VI compounds is underestimated severely as shown in Fig. 3. Removing the self-interaction error, as done for instance in the exact-exchange approach, alleviates this problem, despite the fact that the valence and conduction band edge are give by Bloch states, as we will demonstrate in the following.

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[10] The statements made in this subsection apply also to the generalised gradient approximation.

[11] Only the spherical average of the exchange-correlation hole enters in the expression for the total energy [51], which, at least partly, explains the remarkable success of the LDA

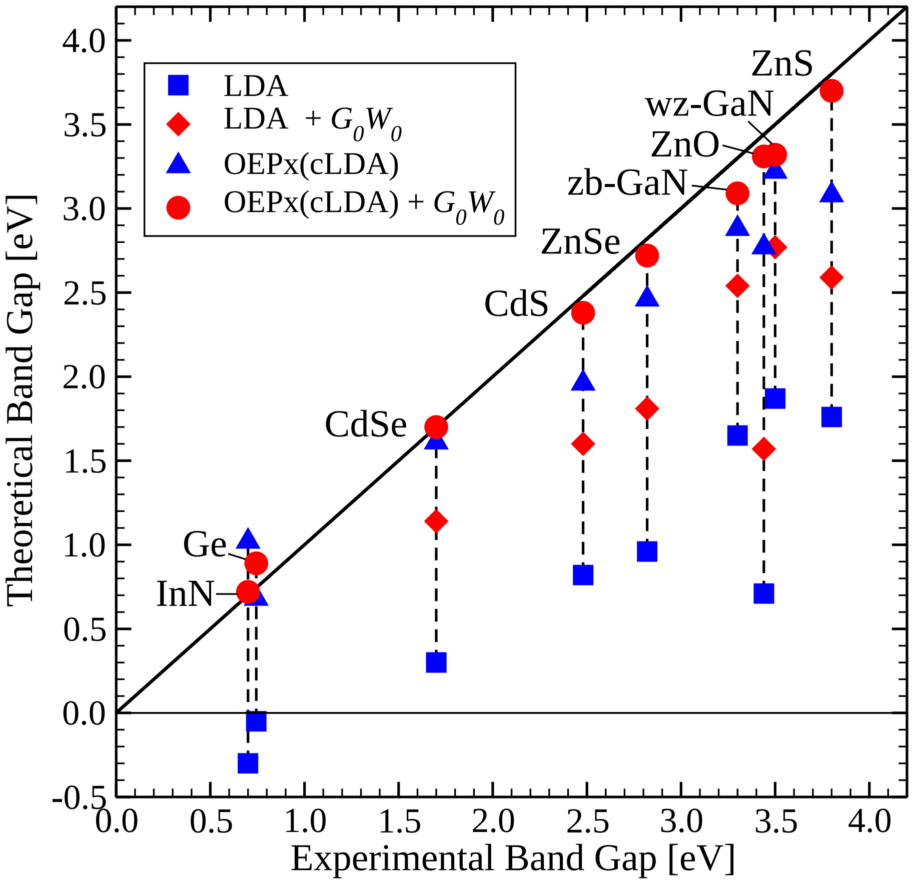

Figure 3: Theoretical versus experimental band gaps: the OEPx(cLDA) based schemes systematically open the band gap compared to the LDA based calculations. Our OEPx(cLDA)+G_0W_0 calculations with the cation d-electrons included as valence electrons agree very well with the experimental values. (For ZnO an estimate of 0.2 eV was added to the zinc-blende values in order to compare to the experimental results for wurtzite.)

3.3 The OEP method and exact-exchange

Following Kohn and Sham's concept of dividing the total energy into known and unknown contributions the exact-exchange energy E_x

$$E_x = -\frac{1}{2}\sum_{ij}^{occ}\iint d\mathbf{r}\, d\mathbf{r}'\frac{\phi_i^*(\mathbf{r})\phi_j(\mathbf{r})\phi_j^*(\mathbf{r}')\phi_i(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} \tag{22}$$

can be isolated from E_{xc} leaving only the correlation part E_c to be approximated. In the exact-exchange only approach this correlation term is omitted[12] so that the total energy becomes

$$E_{tot}^{\text{x-only}} = T_s + E_{ext} + E_H + E_x \quad . \tag{23}$$

For occupied states the exact-exchange term cancels exactly with the corresponding term in the Hartree potential for $i = j$ and thus removes the self-interaction $\delta_i = 0$ in the Perdew-Zunger sense.

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[12] Later in this section we will reintroduce the correlation energy in an approximate form that is commonly used in connection with exact-exchange DFT calculations.

Applying the variational principle to eq. 22 and 23 with respect to the orbitals $\phi_i(\mathbf{r})$ yields the Hartree-Fock approach. Like in the *GW* approach the eigenenergies and wavefunctions are solutions to a quasiparticle equation – all be it with a hermitian operator

$$\left[-\frac{\nabla^2}{2} + v_{ext}(\mathbf{r}) + v_H(\mathbf{r})\right] \phi_i(\mathbf{r}) + \int d\mathbf{r}' \Sigma_x(\mathbf{r}, \mathbf{r}')\phi_i(\mathbf{r}') = \epsilon_i \phi_i(\mathbf{r}) \quad . \tag{24}$$

Σ_x is the non-local Fock-operator or exchange self-energy

$$\Sigma_x(\mathbf{r}, \mathbf{r}') = -\sum_j^{occ} \frac{\phi_j(\mathbf{r})\phi_j^*(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \quad . \tag{25}$$

To stay within the framework of density-functional theory the variation of eq. 22 with respect to the density has to be performed instead. This can be done analytically for the exact-exchange energy expression (eq. 22) in the optimised effective potential approach [53–56]. and yields the local exchange potential

$$v_x^{\mathrm{OEP}}(\mathbf{r}) = \int d\mathbf{r}' \sum_i^{occ} \sum_j^{unocc} \left[\langle \phi_i | \Sigma_x | \phi_j \rangle \frac{\phi_j(\mathbf{r}')\phi_i(\mathbf{r}')}{\epsilon_i - \epsilon_j} + c.c.\right] \chi_0^{-1}(\mathbf{r}', \mathbf{r}) \quad . \tag{26}$$

$\langle \phi_i | \Sigma_x | \phi_j \rangle$ are the matrix elements of the exchange self-energy of eq. 25 and $\chi_0^{-1}(\mathbf{r}, \mathbf{r}')$ is the inverse of the static independent particle polarisability

$$\chi_0(\mathbf{r}, \mathbf{r}') = \sum_k^{occ} \sum_{n \neq k}^{\infty} \frac{\phi_k^*(\mathbf{r})\phi_n(\mathbf{r})\phi_n^*(\mathbf{r}')\phi_k(\mathbf{r}')}{\epsilon_k - \epsilon_n} + \text{c.c.} \quad . \tag{27}$$

The exchange-potential v_x can be thought of as the best local potential approximating the non-local Fock operator [53]. It is important to emphasise, however, that by construction the total energy in Hartree-Fock is always lower (or at most equal) and thus better than in the OEPx formalism [57], because the energy minimisation in the optimised effective potential method is subject to the constraint of the wavefunctions being solutions to the Kohn-Sham equation (13). The eigenvalues of the OEPx formalism for the unoccupied states, on the other hand, are closer to the photo-electron excitation energies for the materials presented in this article than the Hartree-Fock single-particle energies. The difference is due to the derivative discontinuity of the exchange potential (see Section 4.2 and 7). In the exchange-only case the discontinuity is particularly large [8, 9, 58, 59], because the conduction band states are poorly accounted for in Hartree-Fock. They are subject to a different potential than the valence states, since the Fock-operator contains the self-interaction correction only for the valence electrons. In OEPx, on the other hand, the Kohn-Sham valence and conduction band states are governed by the same effective potential, which exhibits the correct asymptotic behaviour ($1/r$ decay for large distances in finite systems). The contribution arising from the discontinuity can be calculated separately [16] and when added to the OEPx band gaps the Hartree Fock band gaps are recovered [59].

In OEPx calculations *local* correlation is frequently added by including the LDA correlation energy in the expression of the total energy (eq. 23). Here we follow the parametrization of Perdew and Zunger [17] for the correlation energy density $\epsilon_c^{\mathrm{HEG}}[n]$ of the homogeneous electron gas based on the data of Ceperley and Alder [60]. This combination will in the following be denoted OEPx(cLDA).

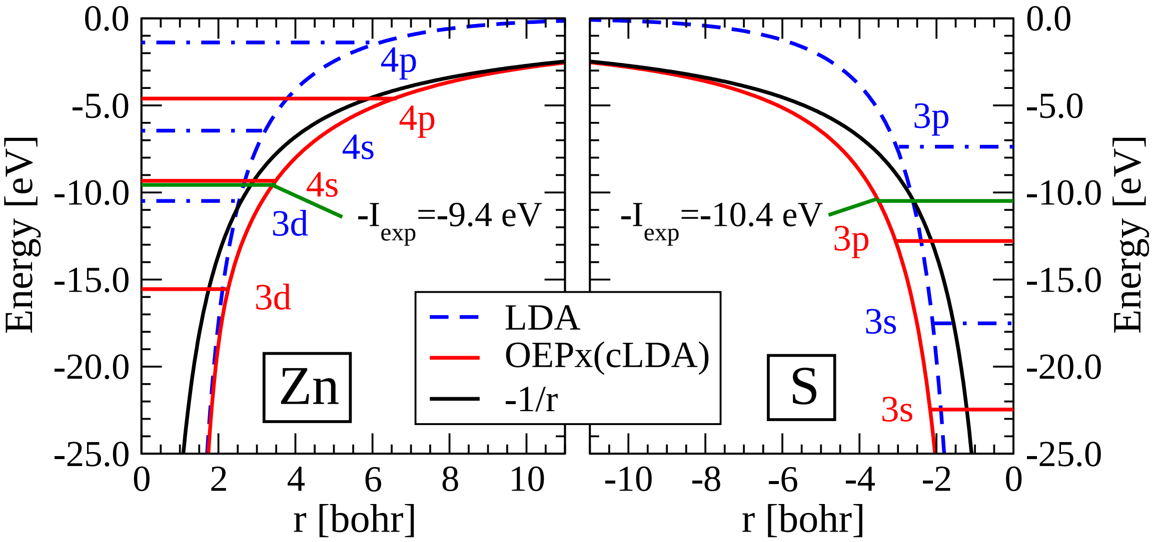

Figure 4: Effective Kohn-Sham potential for the neutral Zn (left panel) and S (right panel) atom: the OEPx(cLDA) potential (red curves) reproduces the correct asymptotic decay $-e^2/r$ (black curves), whereas the LDA (blue, dashed curves) decays exponentially and thus underbinds the electrons. The atomic Kohn-Sham eigenvalues (shown as horizontal lines) are lowered in the OEPx(cLDA) approach compared to the LDA resulting in better agreement with the experimentally measured ionisation potentials (green horizontal lines).

4 OEPx(cLDA)+G_0W_0

4.1 From the atom to the solid

Having established that the removal of the self-interaction in the OEPx(cLDA)-KS approach is the distinguishing feature compared to KS-LDA or KS-GGA calculations we will now illustrate how this leads to an opening of the band gap in solids. For this it is illuminating to start from the eigenvalues of the isolated atoms, depicted in Fig. 4 and 5. The Kohn-Sham potential in OEPx(cLDA) (red curves) is essentially self-interaction free and follows the correct asymptotic $-e^2/r$ potential outside the atom (black curves), whereas the LDA potential (blue, dashed curves) decays exponentially fast. The strong underbinding of the electrons inherent to the LDA is thus greatly reduced in the OEPx(cLDA) approach resulting in a downward shift of all atomic states. The valence electrons of the anion (N, O, S, Se) that take part in the chemical bonding are localised stronger than the respective valence electrons in the cation (Al, Ga, In, Sc, Zn, Cd). The stronger localisation leads to a larger self-interaction correction resulting in a net relative downwards shift of the anion levels with respect to the relevant cation levels (see Fig. 5).

The same behaviour is also observed in SIC-LDA calculations [61] as Tab. 1 demonstrates. In fact for the anions O, S and Se and the 4(5)s in Zn(Cd) Kohn-Sham eigenvalues obtained in the SIC-LDA calculations agree closely with the OEPx(cLDA) ones. The most striking differences are found for the semicore d-electrons, which are significantly lower in SIC-LDA than in OEPx(cLDA). This is due to the fact that the potential in SIC-LDA is non-local. Localising

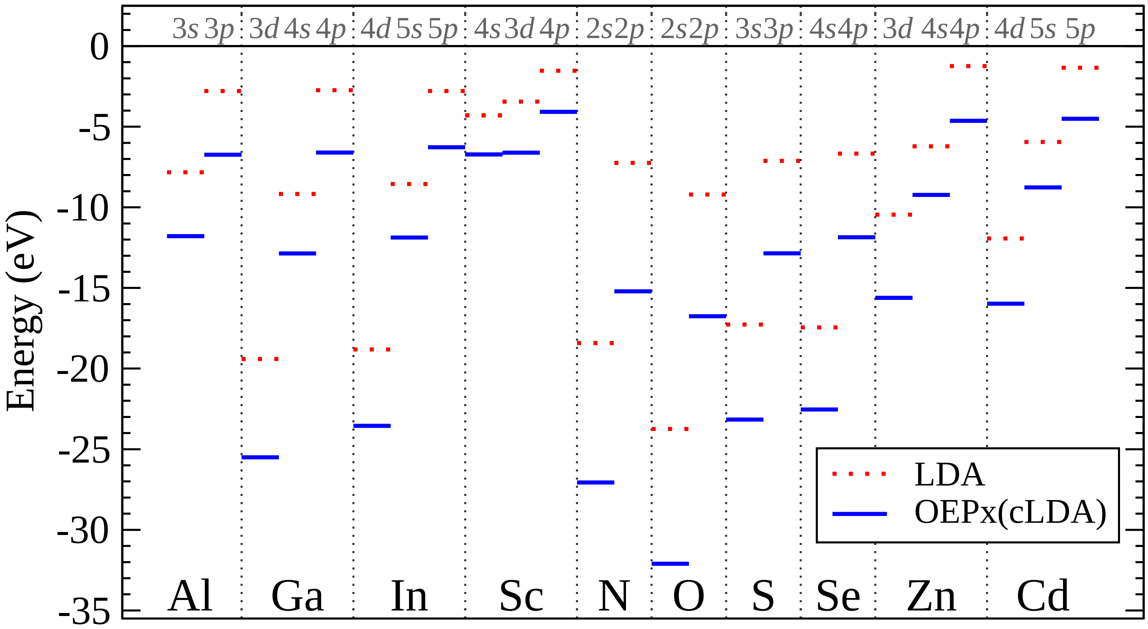

Figure 5: Kohn-Sham eigenenergies of the isolated atoms: The removal of the self-interaction in the OEPx(cLDA) (solid lines) leads to a systematic lowering of all atomic levels compared to the LDA (dotted lines). The downward shift is larger for more localised atomic states, such as the outer valence *s* and *p* states in N, O, S and Se.

	Zn		Cd		O		S		Se	
	4*s*	3*d*	5*s*	4*d*	2*p*	2*s*	3*p*	3*s*	4*p*	4*s*
Exp.	-9.4	-17.2	-9.0	-17.6	-13.6	-28.5	-10.4	-20.3	-9.8	-20.2
LDA	-6.2	-10.4	-6.0	-11.9	-9.2	-23.8	-7.1	-17.3	-6.7	-17.5
SIC-LDA	-9.3	-20.0	-8.9	-18.9	-16.5	-31.0	-11.4	-22.4	-10.5	-22.3
OEPx(cLDA)	-9.2	-15.6	-8.8	-16.0	-16.8	-32.1	-12.9	-23.2	-11.9	-22.5
OEP(SIC-LDA)	-9.1	-15.0	-8.4	-15.8	-17.9	-32.8	-12.1	-22.2	-11.1	-21.3

Table 1: Kohn-Sham eigenvalue spectrum of selected isolated atoms. For the valence states SIC-LDA [61], OEPx(cLDA) and OEP calculations for the SIC-LDA functional (OEP(SIC-LDA)) [62] yield very similar energies and improve significantly on the LDA compared to experiment [63] (remaining difference in the two SIC formulations are due to different parametrisation of the LDA functional). For the semicore *d*-electrons (as well as the lower states not shown here) calculations with a local potential (OEP) give higher eigenvalues than their non-local counterpart.

it by means of the optimised effective potential approach denoted here by OEP(SIC-LDA) [62] leads to a significantly narrower spectrum, i.e. the *d*-electrons (as well as all lower lying states) are moved up in energy. The same behaviour has also been observed for the local/non-local pair of OEPx(cLDA)/Hartree-Fock [64]. The Kohn-Sham energies for both the valence as well as the semicore *d*-electrons then agree quite well in OEPx(cLDA) and OEP(SIC-LDA).

Taking the example of ScN we demonstrate that the large relative shift of the atomic anion state (N 2*p*) also translates to the solid[13]. In ScN the scandium atom donates its two 4*s* and single

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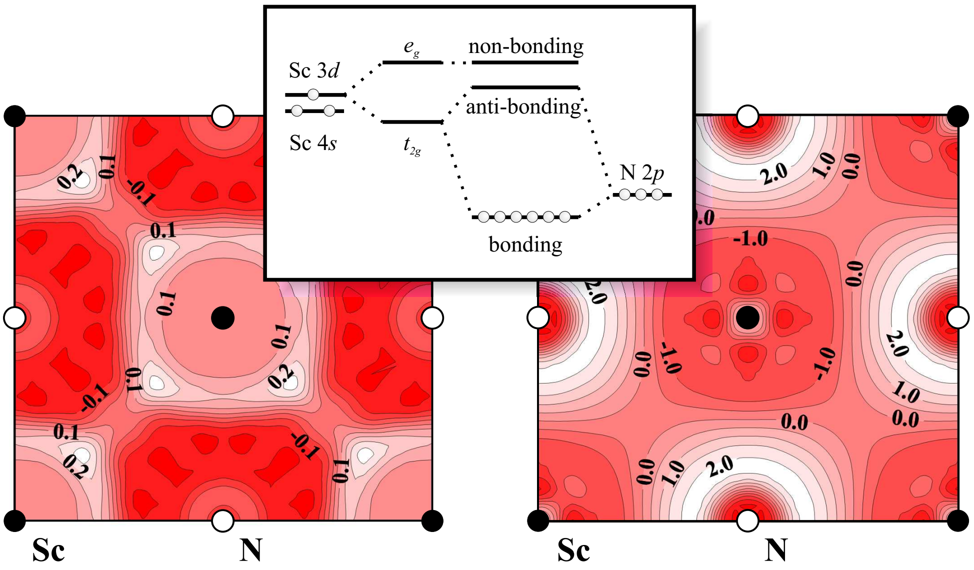
[13] This was for example realised by Vogl *et al.* who therefore suggested to use SIC-LDA pseudopotentials in reg-



(a) **(b)**

Figure 6: The difference between (a) the OEPx(cLDA) and LDA exchange potentials (in Hartree atomic units) and (b) the valence electron densities (in electrons/unit cell) of ScN (the 1*s* of N and the 1*s*, 2*s* and 2*p* of Sc form the core of the pseudopotentials) for one of the square faces of the conventional rock-salt unit cell. Black circles denote Sc and white circles N atoms. The inset shows the bonding scheme together with the electron filling.

3*d* electron to the nitrogen atom (cf. Fig 6). The five *d* states of Sc hybridize with the three valence *p* states of the neighboring N atoms in the rock-salt structure of ScN, forming three *p*-like bonding, three *d*-like anti-bonding t_{2g} and two *d*-like non-bonding e_g bands at the Γ point[14]. The upper three valence bands correspond to the bonding states and originate mainly from the N 2*p* states with some admixture of the Sc 3*d* states, while the lowest conduction band are the anti-bonding t_{2g} states with Sc 3*d* character. The two bands derived from the non-bonding e_g states are more than 1 eV higher in energy [30].

Inspection of the difference between the exchange potential in OEPx(cLDA) ($v_x^{\mathrm{OEPx(cLDA)}}$) and LDA ($v_x^{\mathrm{LDA}}$) shown in Fig. 6(a) reveals that the large relative shift of the atomic anion state (N 2*p*) indeed translates to the solid. Fig. 6(a) illustrates that $v_x^{\mathrm{OEPx(cLDA)}}$ is significantly less attractive than v_x^{LDA} in the Sc regions and more attractive around the N atoms. This difference in v_x leads to a significant electron density redistribution (shown in Fig. 6(b)). The electron transfer from the Sc to the N regions gives rise to an increase in the electron negativity difference between the cation and the anion and thus increases the bond polarisation. This in turn, leads to

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ular LDA calculations for the solid [61,65,66]. While this idea is appealing, using a different exchange-correlation functionals for the pseudopotential than for the solid has to be taken with care. In fact LDA calculations performed with OEPx(cLDA) pseudopotentials, that include the *d*-electrons but not the remaining semicore states in the valence, yield greatly improved *d*-electron binding energies in GaN and InN, whereas OEPx(cLDA) calculations using the same OEPx(cLDA) pseudopotentials worsens the results again [15].

[14] At other points in the Brioulline zone the 4*s* states hybridise, too, which does not change the argument, since the relative shift of the 4*s* and the 3*d* to the N 2*p* level are almost identical.

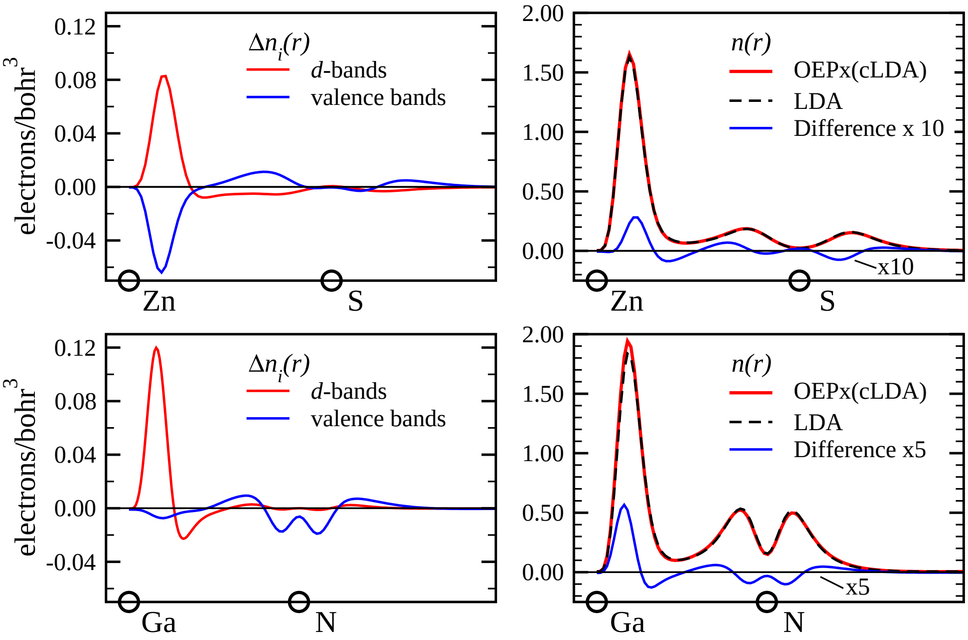

Figure 7: Comparison between OEPx(cLDA) and LDA results for the electron density and the partial densities difference for ZnS (top) and GaN (bottom). Positive density differences indicate an accumulation in OEPx(cLDA). *Left hand side*: partial density differences ($\Delta n_i(\mathbf{r})$) along the [111] direction through the unit cell. The removal of the self-interaction in OEPx(cLDA) leads to a stronger localisation of the d-electrons (red) on the Zn and Ga atoms. In GaN this localisation is not accompanied by a delocalisation of the valence electrons as in ZnS leading to a visible change in the electron density (*Right hand side*: OEPx(cLDA) (red), LDA (black dashed) and their difference (blue line – magnified by a factor of 10 for ZnS and 5 for GaN)).

an opening of the band gap – consistent with our OEPx(cLDA) band structure calculations [30].

In the II-VI compounds and group-III-nitrides this mechanism is also responsible for an opening of the band gap in OEPx(cLDA) compared to LDA, but it is complemented by a contribution arising from the coupling between the anion semicore d-electrons and the 2p-electrons of the anion. Contrary to ScN the cation d-shell of these compounds is fully filled and the bonding and anti-bonding bands are formed between the 2p state of the anion and the highest occupied s states of the respective cation. The bands derived from the d-electrons fall energetically between the anion 2p and 2s bands. The difference in the partial electron densities plotted along the [111] direction for the example of ZnS and GaN in Fig. 7 reveals that the removal of the self-interaction leads to a localisation of the d-electrons in OEPx(cLDA) compared to LDA. In ZnS, where the d-bands are energetically much closer to the p-derived valence bands than in GaN, the stronger localisation of the d-electrons is accompanied by a delocalisation of the p electrons into the bonding region [28]. The localisation of the d electrons reduces the strength of the pd repulsion and the valence bands are lowered in energy leading to a further opening of the band gap in OEPx(cLDA) [15].

4.2 Discontinuity and the band gap

In the Kohn-Sham formalism the system of interacting electrons is mapped onto a fictitious system of non-interacting electrons as alluded to in Section 3. Even in exact DFT only the eigenvalue corresponding to the highest occupied Kohn-Sham state of a finite system can be rigorously assigned to the ionisation potential. For an extended system with well defined chemical potential (Fermi level) this is equivalent to stating that the electron chemical potential in DFT is the same as the true one [49, 67–69]. In comparison to quasiparticle energies this leads to an important difference between the Kohn-Sham and the quasiparticle band structure. This is best illustrated for the example of the band gap of semiconductors and insulators. It can be expressed in terms of total energy differences of the N- and $(N \pm 1)$-particle system ($E_{gap} = E(N+1) - 2E(N) + E(N-1)$). Alternatively, the band gap can be entirely formulated in terms of KS eigenvalues as the difference between the electron affinity and the ionisation potential:

$$\begin{aligned} E_{gap} &= \epsilon^{\mathrm{KS}}_{N+1}(N+1) - \epsilon^{\mathrm{KS}}_{N}(N) && (28) \\ &= \underbrace{\epsilon^{\mathrm{KS}}_{N+1}(N+1) - \epsilon^{\mathrm{KS}}_{N+1}(N)}_{\Delta_{xc}} + \underbrace{\epsilon^{\mathrm{KS}}_{N+1}(N) - \epsilon^{\mathrm{KS}}_{N}(N)}_{E^{\mathrm{KS}}_{gap}} && (29) \end{aligned}$$

Here $\epsilon^{\mathrm{KS}}_i(N)$ denotes the ith Kohn-Sham state of an N-particle system. E^{KS}_{gap} is the eigenvalue gap of a Kohn-Sham calculation for the N-particle system, given by the difference between the highest occupied and lowest unoccupied state. In a solid, in which $N \gg 1$, the addition of an extra electron only induces an infinitesimal change of the density. Therefore, the two Kohn-Sham potentials must be practically the same inside the solid up to a constant shift and, consequently, the Kohn-Sham wavefunctions do not change. The energy difference Δ_{xc} can then only arise from a spatially constant discontinuity of the exchange-correlation energy upon changing the particle number

$$\Delta_{xc} = \left(\left.\frac{\delta E_{xc}[n]}{\delta n(\mathbf{r})}\right|_{N+1} - \left.\frac{\delta E_{xc}[n]}{\delta n(\mathbf{r})}\right|_{N} \right) + \mathcal{O}\left(\frac{1}{N}\right), \qquad (30)$$

since changes in the Hartree potential will be negligible for $\Delta n(\mathbf{r}) \to 0$ [70–72]. This also implies that the dispersion of bands will not be affected by the discontinuity. The conduction bands will merely be shifted relative to the valence bands.

Whether the (considerable) underestimation of the LDA Kohn-Sham band gaps reported here and elsewhere is a deficiency of the LDA itself or a fundamental property of the Kohn-Sham approach has been a longstanding debate. Similarly it has been argued that the band gaps in the OEPx approach should be larger than the true Kohn-Sham gap, since correlation is omitted [9]. The exchange-correlation energy in the LDA is a continuous function of the density with respect changes in the particle number and will thus not exhibit a discontinuity even if the band gap were calculated by means of total energy differences. The OEPx formalism, on the other hand, exhibits a derivative discontinuity [69, 73], which would be taken into account if the excitation energies were calculated by computing total energy differences between the N and the $N \pm 1$ electron system [73]. In KS-DFT, however, this derivative discontinuity does not enter the calculation. The opening of the band gap discussed in Section 4.1 is therefore due to the removal of the self-interaction and not the derivative discontinuity. Adding the corresponding derivative discontinuity would make the band gaps comparable to the Hartree-Fock ones [9,58,74], which are significantly too large.

Further substance to the notion that even the exact Kohn-Sham potential would give rise to a band gap underestimation was first given by Gunnarsson and Schönhammer [75,76] and Godby *et al.* [72,77] and recently by Grüning *et al.* [74]. Gunnarsson and Schönhammer derive their conclusions from an exactly solvable, Hubbard-like model, whereas Godby *et al.* and Grüning *et al.* use the Sham-Schlüter formalism to generate the local exchange-correlation potential that corresponds to the G_0W_0 self-energy. They show that the resulting potential closely resembles that of the LDA. Also the Kohn-Sham eigenvalue differences are very similar to the LDA ones, whereas G_0W_0 calculations, that incorporate information of the $(N \pm 1)$-particle system in a natural way (see Section 2), generally give quasiparticle band gaps to within 0.1-0.2 eV [2].

The good agreement of the OEPx(cLDA) band gaps with experiment reported previously [7–13] can therefore be regarded as fortuitous. In fact inspection of Fig. 3 shows that the OEPx(cLDA) band gaps are lower than the experimental ones [28] and hence lower than those of previous studies. This is due to the fact, that unlike in the earlier work we have explicitly included the *d*-electrons as valence electrons in our pseudopotential calculations. The *pd* repulsion pushes the valence bands up in energy and shrinks the gap [15]. All-electron OEPx calculations for CdS and ZnS, on the other hand, report band gaps considerably higher than the experimental ones [58]. The origin of this discrepancy between pseudopotential and all-electron calculations is still under debate. A band gap underestimation in OEPx has also been observed for noble gas solids and insulators [14,58,59].

Table 2 illustrates for four representative compounds that adding LDA correlation to the OEPx approach has a marginal effect on the calculated band gaps (and the band structure in general – not shown). ZnO is the only material of those studied here where the difference exceeds 0.1 eV. The remaining difference between OEPx(cLDA) and experiment is recovered to a large degree in the G_0W_0 quasiparticle energy calculations as Tab. 2 and Fig. 3 demonstrate.

The advantage of the OEPx(cLDA)-G_0W_0 approach is that it proves to be sufficient to include the *d*-electrons of the cation explicitly as valence electrons in the pseudopotential without having to include the remaining electrons of the semicore shell, provided OEPx(cLDA) pseudopotentials [90] are used. Taking GaN as an example the 3*s*-electrons in the Gallium atom are approximately 130 eV and the 3*p*-electrons approximately 80 eV lower than the 3*d* states. Resolving these more strongly localized 3*s* and 3*p*-electrons in GaN with a plane-wave basis set will hence require significantly higher plane-wave cutoffs than the 70 Ry required for the *d*-electrons [28]. In a pseudopotential framework it would thus make sense to explicitly include the *d*-electrons of the cations in the II-VI compounds and group-III-nitrides as valence electrons, but to freeze the chemically inert semicore *s* and *p*-electrons in the core of the pseudopotential. However, due to the large spatial overlap of the atomic semicore *s* and *p* with the *d* wavefunctions, core-valence exchange-correlation is large in these compounds. As a consequence core-valence exchange-correlation is treated inconsistently when going from LDA to LDA-G_0W_0, if pseudopotentials are used in this fashion, because the exchange-correlation self-energy in the GW approach acts on the *d*-electrons in the solid, but cannot act on the *s* and *p*-electrons in the semicore shell, too. The result is a severe underestimation of the LDA-G_0W_0 band gaps (cf. red

DFT	PP	Conf.	*GW*	ZnO	ZnS	CdS	GaN
OEPx	OEPx	*d*		2.34	2.94	1.84	2.76
OEPx(cLDA)	OEPx(cLDA)	*d*		2.57	3.08	1.96	2.88
OEPx	OEPx	*d*	*GW*	3.07	3.62	2.36	3.09
OEPx(cLDA)	OEPx(cLDA)	*d*	*GW*	3.11	3.70	2.39	3.09
Experiment				(3.44)	3.80	2.48	3.30
LDA	LDA	no *d*'s	*GW*		3.98[a]	2.83[a]	3.10[b]
OEPx(cLDA)	OEPx(cLDA)	no *d*'s			3.74[c]		3.46[d]
OEPx(cLDA)	OEPx(cLDA)	no *d*'s	*GW*				3.49[e]
LDA	LDA	*d*	*GW*			1.50[f]	
LDA	LDA	*d*-shell	*GW*		3.64[g]		
LDA	LDA	*d*-shell	*GW*		3.50[h]	2.45[h]	2.88[h]
LDA	LDA	*d*-shell	*GW*		3.38[i]	2.11[i]	
LDA	FP	all e^-	*GW*	(2.44)[j]	3.24[k]		(3.03)[k]
LDA	ASA	all e^-	*GW*	(4.06)[m]	3.97[l]		(3.25)[m]

Table 2: DFT and quasiparticle band gaps in eV for ZnO, ZnS, CdS, and GaN in the zinc-blende structure sorted in increasing energy from top to the experimental values. The first column lists the DFT scheme and the second column denoted PP the pseudopotential used. For all-electron calculations this column denotes if the atomic sphere approximation (ASA) or the full potential (FP) was employed. "Conf." refers to the configurations of the (pseudo)atoms: *d*-electrons included (*d*), as described in the previous section, valence only (no *d*'s), *d*-electrons and their respective shell included (*d*-shell) and all-electron (all e^-). Experimental results are taken from: ZnO [78], ZnS [79], CdS [80], GaN [81] and the OEPx(cLDA) and *GW* data from: [a]Ref. [82], [b]Ref. [83], [c]Ref. [13], [d]Ref. [8], [e]Ref. [10], [f]Ref. [84], [g]Ref. [44], [h]Ref. [85], [i]Ref. [45] [j]Ref. [86], [k]Ref. [87], [l]Ref. [88], [m]Ref. [89]. Numbers in round brackets refer to wurtzite structures. In Ref. with superscript [a] and [b] a model dielectric function was employed and in [e,f,g,h] a plasmon pole model was used.

diamonds in Fig. 3) and *d*-bands that are pushed energetically into the *p*-derived valence bands in the II-VI compounds [28, 84, 85] (cf. Tab. 3). The only way to remedy this problem within LDA-G_0W_0 is to free the electrons in question by performing all-electron G_0W_0 calculations [87] or by using pseudopotentials that include the entire shell as valence electrons [44, 84, 85], which in the latter case introduces formidably high plane-wave cutoffs. If, on the other hand, OEPx or OEPx(cLDA) is used for the ground state calculation, then the exchange self-energy already acts on the semicore *s* and *p* states in the generation of the pseudopotential. Since the exchange self-energy can be linearly decomposed into a core and a valence contribution no non-linear core corrections [91] arise in the Hartree-Fock case and they are expected to be small for OEPx(cLDA) pseudopotentials [9]. When going from OEPx(cLDA) to OEPx(cLDA)-G_0W_0 core-valence exchange is therefore treated consistently, even when the semicore *s* and *p*-electrons are frozen in the core, as long as OEPx(cLDA) pseudopotentials are used [28].

For InN the LDA starting point is so problematic (cf. Fig. 1), that even subsequent all-electron G_0W_0 calculations only open the gap to 0.02 - 0.05 eV [87, 92]. Here the importance of removing the self-interaction from the ground state calculation has been demonstrated before by combining SIC-LDA calculations with G_0W_0 calculations (all be it in a rather approximate way by adjusting the *pd* repulsion and combining calculations with and without the 4*d*-electrons in the core of the In pseudopotential) [93, 94]. The OEPx(cLDA) approach, on the other hand, predicts InN to be a semiconductor, as Fig. 1 illustrates, and G_0W_0 calculations can be applied without further approximations. The size of the band gap of InN and the origins for the considerable spread in the experimental data has been a matter of intense debate over the last years. For wurtzite InN our value of 0.7 eV [29] strongly supports recent experimental findings [95–97]. The OEPx(cLDA)-G_0W_0 calculations further helped to clarify the source for wide interval of experimentally observed band gaps [29].

For ScN, an emerging versatile material for promising technological applications, the electronic band structure has also been difficult to access experimentally, due to growth related problems. Similar to the metallic state in InN the strong self-interaction effects in the LDA predict ScN to be semimetallic (cf. Fig. 1) preventing a direct application of the LDA-G_0W_0 approach. By using OEPx(cLDA) the ground state becomes essentially self-interaction free. This leads to an opening of the band gap and a suitable semiconducting starting point for G_0W_0 calculations [30], as described in Section 4.1. The OEPx(cLDA)-G_0W_0 calculations lower the OEPx(cLDA) band gap of 1.7 eV down to 0.8 eV [30], clearly supporting recent experimental findings of an indirect gap of 0.9 ± 0.1 eV [98].

While LDA based G_0W_0 calculations generally open the band gap from the underestimated LDA value, we observe here that the G_0W_0 corrections lower the respective gaps of InN and ScN obtained in the OEPx(cLDA) approach. Since G_0W_0 falls into the realm of perturbation theory (cf. eq. 12) this is not unusual, because it is not *a priori* clear if the quasiparticle corrections are positive or negative.

4.3 *d*-electron binding energies

A known problem for the group-III-nitrides and the II-VI compounds is, that both KS-LDA and LDA based G_0W_0 calculations underestimate the binding of the *d*-bands – regardless of whether applied in a pseudopotential or an all-electron fashion. For four selected compounds the center of the *d*-bands[15] is listed in Tab. 3 for the different computational schemes. For ZnS and CdS OEPx(cLDA) and OEPx(cLDA)-G_0W_0 produce essentially the same *d*-electron binding energies. Only in ZnO quasiparticle corrections are found to lower the *d*-states by 1.5 eV compared to OEPx(cLDA), further reducing the *pd* coupling. Again, adding LDA correlation to the OEPx has essentially no effect on the energies of the bands (energy differences < 0.2 eV).

At first side it appears surprising that the removal of the self-interaction does not automatically lower the position of the *d*-bands in OEPx(cLDA). Closer inspection of the atomic eigenvalues in Fig. 5 reveals, however, that the cation *d* states are shifted down by almost the same amount as the anion *p* states. The relative position to the *p* states, from which the top of the valence band is derived, therefore remains similar in OEPx(cLDA) and LDA [15]. A recent all-electron OEPx study (line 20) has reported values for ZnS and CdS in better agreement with experiment [58]. However, it is not clear at the present stage, why for CdS the difference to our pseudopotential calculations (line 5) is merely 0.6 eV, whereas for ZnS it amounts to 2.2 eV. It is also not clear if the discrepancy is caused by core-valence linearisation in the pseudopotentials, the use of pseudo- rather than all-electron wavefunctions in the Fock-operator or if it has an entirely

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[15] This has been obtained by averaging over the *d*-bands at the Γ-point.

	DFT	PP	Conf.	*GW*	ZnO	ZnS	CdS	GaN
1	LDA	LDA	d	*GW*	4.29	4.30	6.17	13.05
2	OEPx(cLDA)	LDA	d	*GW*	4.98	5.02	6.40	13.58
3	OEPx(cLDA)	LDA	d		4.36	5.33	6.54	12.75
4	LDA	LDA	d		5.20	6.32	7.72	14.25
5	OEPx	OEPx	d		5.12	6.91	7.57	14.85
6	OEPx(cLDA)	OEPx(cLDA)	d		5.20	7.05	7.61	15.02
7	OEPx	OEPx	d	*GW*	6.68	6.97	7.66	16.12
8	OEPx(cLDA)	OEPx(cLDA)	d	*GW*	6.87	7.08	7.75	16.15
9	Experiment				(9.00)[a]	8.97[a]	9.20[b]	17.70[c]
						9.03[d]	9.50[d]	
10	LDA	LDA	d	*GW*			5.20[e]	
11	LDA	LDA	d-shell	*GW*		7.40[f]		
12	LDA	LDA	d-shell	*GW*		6.40[g]	8.10[g]	15.70[g]
13	LDA	LDA	d-shell	*GW*		6.87[h]	8.15[h]	
14	LDA	LDA	d-shell	*SAT*		7.90[g]	9.10[i]	17.30[g]
15	LDA	LDA	d-shell	$G'W'\Gamma$		8.02[h]	8.99[h]	
16	LDA+U	LDA				8.78[j]		
17	LDA+U	LDA	d-shell	*GW*		7.10[j]		
18	LDA	FP	all e^-	*GW*	6.16[k]	7.10[l]	8.20[l]	(16.40)[l]
19	LDA	ASA	all e^-	*GW*	(5.94)[m]	8.33[n]		(17.60)[m]
20	OEPx	FP	all e^-			9.1[o]	8.2[o]	

Table 3: d-electron binding energies referenced to the top of the valence band: The layout is the same as in Table 2. Experimental values taken from: [a]Ref. [99], [b]Ref. [100] [c]Ref. [101], [d]Ref. [102], and the *GW* data from: [e]Ref. [84], [f]Ref. [44], [g]Ref. [85], [h]Ref. [45] [i]Ref. [103], [j]Ref. [104] (U=8 eV, J=1 eV), [k]Ref. [86], [l]Ref. [87], [m]Ref. [89], [n]Ref. [88]. [o]Ref. [58]. *SAT* denotes *GW* calculations including plasmon satellites, $G'W'\Gamma$ calculations performed with eigenvalue self-consistency and addition of a vertex contribution. Numbers in round brackets refer to wurtzite structures. In Ref. denoted by [d,e,f] a plasmon pole model was used.

different origin.

Overall the binding energies obtained with our OEPx(cLDA)-G_0W_0 approach agree well with other available *GW* calculations (line 11, 12, 13 and 18 in Tab. 3), but are still about 2 eV at variance with experiment. The reason for this could be twofold. Either interactions beyond the *GW* approximation are required for describing the excitation of these semicore d-electrons and/or one needs to go beyond even OEPx(cLDA) as a starting point. Evidence that both self-consistency in the *GW* calculations as well as the inclusion of vertex effects (see Section 2.2) introduce changes in the right direction was recently given by Fleszar and Hanke. Starting from an LDA ground state only eigenvalues (and not also the wavefunction) were iterated to self-consistency in the *GW* calculations. This increases the binding energy by 0.4-0.5 eV in ZnS and CdS [45]. Adding a vertex function that makes the calculation "consistent" with the LDA starting point[16] increases the binding energy by additional 0.4-0.5 eV (line 15) [45]. Similar conclusions were drawn from a recent study, in which G_0W_0 calculations were based on LDA+U ground states [104]. With increasing U the binding energy of the d-state increases linearly in

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[16] Strictly spoken Σ is only zero on the first iteration when starting from Hartree theory (see Section 2.2). Starting from a Kohn-Sham calculations implies that $\Sigma = v_{xc}$ in which case the vertex function is not given by delta functions, but can still be analytically derived [105].

the LDA+U calculations (line 16) as the *pd* hybridisation reduces. The G_0W_0 calculations, however, prove to be insensitive to this change and shift the d-bands up again, close to their LDA-G_0W_0 positions (line 17) [104]. Previously Rohlfing *et al.* had devised a way to go beyond the GW approximation by including plasmon satellites in the Green's function, denoted here by SAT (line 14 in Tab. 3). Although the SAT improves on the d-electron binding energies it considerably over corrects the band gap and the valence part of the bandstructure [103]. Work towards a consistent description of excitation energies of the semicore d-electrons in these materials is clearly required in the future.

5 Comments on self-sonsistency

As alluded to in Section 2.2 it is still a matter of debate how to perform self-consistent GW calculations for quasiparticle band structures. For the non self-consistent G_0W_0 scheme our calculations for the selected II-VI compounds and GaN have revealed a significant dependence on the starting point, if the semicore d-electrons, but not the remaining electrons of the semicore shell, are explicitly taken into account in the pseudopotential calculation. Since for the reasons given in Section 4.2 no meaningful comparison can be made between LDA-G_0W_0 and OEPx(cLDA)-G_0W_0 calculations unless the entire semicore shell is included in the calculation these systems are not suitable to assess the influence of the starting point on the quasiparticle band structure. In ScN, on the other hand, the semicore d-shell of Sc is not fully filled and the remaining s and p-electrons in the semicore shell are much higher in energy. Taking GaN as an example the $3s$-electrons in the Scandium atom are approximately 100 eV and the $3p$-electrons approximately 60 eV higher than in Gallium. Resolving the $3s$ and $3p$-derived bands in ScN with plane-waves thus only requires a cutoff of 80 Ry [30] and makes ScN an ideal candidate for constructing a comparison between LDA-G_0W_0 and OEPx(cLDA)-G_0W_0 calculations.

However, the *negative* LDA band gap (see Table 4) impedes the direct application of the LDA-G_0W_0 formalism with our GW code, since in its current implementation [106–108] a clear separation between conduction and valence bands is required. Therefore, an indirect approach is adopted. First, LDA-G_0W_0 calculations are performed at a lattice constant (a_0 = 4.75 Å) larger than the experimental one (a_0 = 4.50 Å), where the fundamental band gap in the LDA is small but positive. We then use the LDA *volume* deformation potentials (which agrees well with the OEPx(cLDA)-G_0W_0 one) to determine the corresponding LDA-G_0W_0 band gaps at the equilibrium lattice constant[17].

We find (Tab. 4) that the LDA-G_0W_0 and OEPx(cLDA)-G_0W_0 calculations, starting from the two extremes (negative band gap in LDA, 0.8 eV overestimation in OEPx(cLDA)), yield quasiparticle band gaps that agree to within 0.3 eV. Since the LDA-based calculations are close to the limit of metallic screening, whereas the OEPx(cLDA)-based calculations form the opposite extreme of starting from a completely self-interaction free exchange-correlation functional, we expect the results of a self-consistent GW calculation to fall in the range between the LDA-G_0W_0 and OEPx(cLDA)-G_0W_0 calculations. ¿From these results we estimate the maximum

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[17] This approach is in principle not limited to band gaps and can equally well be applied to the full band structure for example.

Approach	$E_g^{\Gamma-\Gamma}$	$E_g^{\Gamma-X}$	E_g^{X-X}
OEPx(cLDA)-G_0W_0	3.51	0.84	1.98
LDA-G_0W_0	3.71	1.14	2.06
$[G_0W_0]_{\text{average}}$	3.62	0.99	2.02
OEPx(cLDA)	4.53	1.70	2.59
GGA	2.43	−0.03	0.87
LDA	2.34	−0.15	0.75
Experiment			
Ref. [12]	∼3.8	1.30	2.40
Ref. [98]		0.9±0.1	2.15

Table 4: Calculated and experimental band gaps (E_g) of ScN (in eV). $[G_0W_0]_{\text{average}}$ denotes the arithmetic average between the OEPx(cLDA)-G_0W_0 and LDA-G_0W_0 results (see text).

error bar associated with omitting self-consistency in GW to be the difference from the arithmetic averages for $E_g^{\Gamma-\Gamma}$, $E_g^{\Gamma-X}$ and E_g^{X-X} (reported in Tab. 4) to the largest deviation between LDA-G_0W_0 and OEPx(cLDA)-G_0W_0, i.e. of the order of 0.15 eV for ScN [30].

6 Conclusions

We have presented the combination of quasiparticle energy calculations in the G_0W_0 approximation with DFT calculations in the OEPx(cLDA) approach. Using OEPx(cLDA) instead of LDA or GGA removes the inherent self-interaction of the latter from the ground state calculation. Starting from the individual atoms we have illustrated how this leads to an opening of the Kohn-Sham band gap in the solid. In the spirit of perturbation theory OEPx(cLDA) thus provides a more suitable starting point for G_0W_0 calculations than LDA or GGA. For materials for which LDA erroneously predict a (semi)metallic state, like e.g. InN or ScN, OEPx(cLDA) yields a semiconducting ground state, which unlike in the LDA case, permits a direct application of the G_0W_0 approximation. For the II-VI compounds and group-III-nitrides presented here as an example the band gaps in the OEPx(cLDA)-G_0W_0 approach are in excellent agreement with experiment and the position of the semicore *d*-electron bands no worse than in previous LDA-G_0W_0 calculations.

7 Outlook

A disadvantage of the OEPx approach is that it yields total energies and thus structural properties comparable to Hartree-Fock. For the materials considered here all lattice parameters are know from experiment, but this might not be the case for surfaces, defects, nanostructures or new materials. Then, in cases where LDA and GGA are known to fail, exchange-correlation functionals that appropriately include exact-exchange are needed.

In the most pragmatic approach a portion of Hartree-Fock exchange is mixed with a different portion of local exchange and correlation[18]

$$E_{xc}^{hyb} = E_{xc}^{\mathrm{DFT}} + \alpha(E_x^{\mathrm{HF}} - E_x^{\mathrm{DFT}}) \tag{31}$$

as already alluded to in the introduction. Prominent examples of these so called hybrid functionals are PBE0 [18–20] and B3LYP [21–23]. The factor α in front of the Hartree-Fock exact-exchange term is equivalent to a constant static screening function. Alternatively a more complex screening function can be chosen as in the screened-exchange (sX-LDA) approach [16, 27, 109, 110], the Σ-GKS scheme [111] or the Heyd-Scuseria-Ernzerhof (HSE) functional [24–26]. An appealing feature of the hybrid functional approach is that it incorporates part of the derivative continuity of the exchange-correlation energy [16]. For semiconductors and insulators this leads to a much improved description of band gaps [26, 59, 109, 110, 112–114]. However, the dependence on universal (i.e. material independent) parameters questions their *ab initio* character.

While ample experience with hybrid functionals exists in the quantum chemistry community applications to solids are only slowly emerging and many issues remain open or controversial i.e. computational efficiency, basis sets, pseudopotentials or the choice of functional. In particular with regard to structural properties systematic studies are required. Conversely, GW quasiparticle energy calculations have traditionally fallen into the realm of condensed matter physics, but also here similar open questions remain (most of which have not been explicitly discussed in this article) including that of self-consistency. Again in a more pragmatic fashion the latter has recently been approached by mapping the G_0W_0 self-energy back onto a static, non-local potential. The eigenenergies and eigenfunctions of the new non-local, hermitian Hamiltonian then serve as input for the next G_0W_0 cycle until self-consistency is reached [42, 115]. This approach shows promising success for different types of materials ranging from semiconductors, insulators and metals to transition metal oxides and f-electron systems [42, 115–117].

Although the G_0W_0 approximation is currently the state of the art approach to calculate defect levels for solids from first principles it has so far only been applied in a few cases [118–122]. Ideally the GW calculations would encompass the determination of the defect structure, too, but so far GW total energy calculations, that would be required for this task, are still in their infancy. Meanwhile a combination of exact-exchange-based DFT functionals (for the structural properties) with quasiparticle energy calculations in the GW approach and the G_0W_0 approximation (for the spectral properties) offer the possibility to develop a better understanding of defect properties untainted by structural artefacts caused by the self-interaction or the band gap underestimation of Kohn-Sham. First G_0W_0 calculations for bulk semiconductors based on hybrid functionals appear to be promising [116, 123], but more work along these lines is needed in the future.

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[18] Some hybrid schemes introduce additional parameters for mixing different portions of the local-spin density (LSD) functional with GGA exchange and correlation in the E_{xc}^{DFT} term [20].

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