Report on the 13^{th} NANOQUANTA-ETSF Workshop on Electronic Excitations: Theoretical Spectroscopy and Quantum Transport

Pugnochiuso (Italy), 22-27 September 2008

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The aim of the workshop was to assess the present status, the latest achievements, and future perspectives of ab-initio theories for the calculation of electronic excitations in condensed matter and nanostructures. Furthermore, the related topic of quantum transport was a strong focus of the meeting. 114 researchers participated in the workshop. 18 invited talks, 26 contributed talks, 34 posters were presented during the 5-day workshop.

The first session dealt with "DFT Development and Application". In a keynote lecture, Leeor Kronik (Weizann Institute) talked about the importance of self-interaction correction and the performance of hybrid functionals for the proper description of localized states in organic semiconductors. Volker Blum (Berlin) presented the progress of the Fritz-Haber Institute in the development of "DFT-and-beyond" code based on numerical localized orbitals. Applications of this code for calculation of surface reconstruction at gold and platinum surfaces were presented by Paula Havu (Berlin). Matthieu Verstraete (Louvain-la-Neuve) gave convincing evidence for the influence of spin-orbit coupling on the phonon-dispersion of lead. Jelena Sjakste (Ecole Polytechnique) presented an ab-initio approach for electron-phonon scattering in semiconducting nanostructures.

The session on "Strongly Correlated Systems" was introduced by Richard Martin (University of Illinois and Stanford University) who compared the performance of the GW-approximation and of dynamical mean-field theory (DMFT) for the description of strong correlations. Nicole Helbig (San Sebastián) presented the explicit calculation of the exchange-correlation potential for a simple model system (one-dimensional hydrogen molecule). Franca Manghi (Modena) described a "3-Body-Scattering-Theory" for the description of electron-electron correlation in narrow-band systems. Sangeeta Sharma (Berlin) presented a new exchange correlation functional which captures the correct band-gap behavior for weakly and strongly correlated insulators in the framework of density-matrix functional theory.

There were three sessions on "quantum transport", demonstrating the growing interest of this field in the electronic-excitation community. The sessions comprised 5 keynote lectures: Massimiliano Di Ventra (UC San Diego) gave a general introduction to the field and discussed the viscous nature of the electron liquid giving rise to non-linear effects. Stefano Sanvito presented the `Smeagol` non equilibrium electronic transport code and its application to magnetic tunnel junctions. Alexej Bagrets (Karlsruhe, replacing Ferdinand Evers) talked about the limitations of local approximations to the exact XC-functional and a way to overcome these limitations by constructing a functional using the density matrix renormalization group. Jeff Neaton (Berkeley) reviewed transport measurements on single-molecule junctions and gave evidence for the quantitative performance of DFT-based methods including a band-gap correction. Frank Ortmann (Jena) presented an ab-initio framework for the calculation of the temperature-dependent mobility tensors in wide-band and narrow-band semiconductors. Contributed talks in the quantum transport session covered memory effects (Robert van Leeuwen, Univ. of Jyväskylä; Stefan Kurth, San Sebastian), applications to graphene nanostructures and carbon nanotubes (Didier Mayou, Grenoble; Zeila Zanolli and Simon Dubois, Louvain-la-Neuve), GW and hybrid functional corrections (Andrea Ferretti, Modena), and the use of a stroboscopic wavepacket basis (Peter Bokes, Bratislava).

As in previous years, a keynote lecture was given by an experimentalist: Pierre Malet (Grenoble) reviewed scanning tunneling microscopy and presented quantum interferences in graphene.

The session on "Optical Properties" contained contributions on Ge and Si nanocrystals (Margherita Marsili, Rome; Roberto Guerra, Modena) and porphyrin oligomers (Conor Hogan, Rome), the dynamic structure factor of metals (Marco Cazzaniga, Milan), and the linear density response function within the time-dependent exact-exchange approximation (Maria Hellgren, Lund). Applications to graphene were discussed by Christine Giorgetti (Ecole Polytechnique) and Paolo Trevisanutto (Grenoble).

A large session was devoted to "Theory Developments". Double excitations were discussed in the presentations of Pina Romaniello (Ecole Polytechnique) and Davide Sangalli (Milan). Ilya Tokatly (San Sebastian) introduced the concept of quantum stress as an alternative tool to visualize chemical bonding. Eleonora Luppi (Ecole Polytechnique) discussed the role of local-field effects in the second-order harmonic generation in crystalline semiconductors. Gianluca Stefanucci (Rome) presented a real-time view of electron spin manipulation in quantum dots. New ways to overcome the difficulty of treating f-electron systems in GW were presented by Hong Jiang (Fritz Haber Institute).

In a session on "Methods and Implementations", recent progress in code developments was discussed. Myrta Grüning (Louvain-la-Neuve) presented a computational approach to calculate the real spectrum of non-Hermitian Hamiltonians. Fabien Bruneval (CEA) showed how to obtain precise GW-bands with only a few empty states. Adriano Mosca Conte (Rome) demonstrated how many-body theory can be extended to large systems in the framework of the QM/MM approach. Frank Fuchs (Jena) talked about an efficient $O(N^2)$ approach for the solution of the Bethe-Salpeter equation.

The last session of the workshop was devoted to "applications of many-body perturbation theory". Riad Shaltaf (Louvain-la-Neuve) explained how band offsets in quantum-hetero structures can be calculated within the GW approximation. Christoph Friedrich (Jülich) presented efficient all-electron GW calculations of complex materials. Arno Schindlmayr (Paderborn) discussed spin excitations from first principles. Marc Puig van Friesen (Lund) talked about non-equilibrium dynamics in strongly correlated clusters. The session ended with a presentation of Michael Rohlfing (Osnabrück) on exchange and correlation effects on adsorbation energies at metal surfaces.

Besides the oral presentations, much attention was given to the poster presentations. A time-slot of three hours was devoted to the poster session and posters could be discussed during all the coffee breaks of the 5-day workshop.

The next ETSF workshop on electronic excitations will take place in Evora, Portugal, September 15-19, 2009.

13th ETSF/ Nanoquanta Conference

Theoretical Spectroscopy and Quantum Transport

The aim of this workshop is to assess the present status, the latest achievements and future perspectives of *ab initio* theories for the calculation of excitations, excited state properties and spectroscopy in condensed matter and nanostructures, systems with increasing complexity, and materials of technological and biological interest.

The workshop belongs to a series of annual events organized by the Nanoquanta European Network of Excellence and with the participation of the coordinator of the American projects FEFF and CSMN.

The workshop covers theoretical approaches (GW-Approximation, Bethe-Salpeter-Equation, Time-Dependent Density Functional Theory), its numerical implementations, and application to systems of technological interest. In close relation to electronic excitations, quantum transport, i.e., electron transport through molecules and nanostructures, has become an important topic of our annual workshop. The workshop brings together theory and code developers with code users and with experimentalists who seek an understanding of their spectra with the help of *ab initio* calculations.

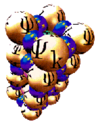
The workshop series has its origin in the NANOQUANTA European Network of Excellence and preceeding European Research and Training Networks EPSI, EXCAM and NANOPHASE dealing with electronic excitations. In the future, it will become the annual workshop of the European Theoretical Spectroscopy Facility (ETSF).

ETSF

European Theoretical Spectroscopy Facility

nanoquanta

Network of Excellence



Pugnochiuso is situated on the coast between Vieste (20 Km) and Mattinata, in the heart of the National Park of Gargano on the borders of the Umbrian Forest. The road which leads to the resort curls around hairpin bends among olive trees, maritime pines and terraces with sea views of unique beauty.

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Programme

Monday 22	
09:00	Nanoquanta Steering Committee IT s
16:00	Registration
	Nanoquanta Steering Committee
20:00	Dinner

Tuesday 23	
09:00	Welcome
09:30	Kronik
10:00	Verstraete
10:20	Caffeine Break
10:50	Sjakste
11:10	Blum
11:30	Havu
12:00	Lunch
14:30	Martin
15:00	Helbig
15:20	Caffeine Break
15:50	Manghi
16:20	Sharma
16:50	Poster Session +
18:30	Aperitivo
20:00	Dinner

Wednesday 24	
09:00	Di Ventra
09:30	Mayou
10:00	Zanolli
10:20	Caffeine Break
10:40	Sanvito
11:10	Bokes
11:40	Ferretti
12:00	Lunch
14:30	Mallet
15:00	Marsili
15:20	Hellgren
15:40	Caffeine Break
16:10	Cazzaniga
16:30	Guerra
16:50	Hogan
17:10	YR Meeting
20:00	Social Dinner

Thursday 25	
09:00	Evers
09:30	Dubois
09:50	Kurth
10:10	Caffeine Break
10:40	Neaton
11:10	Ortmann
11:40	van Leeuwen
12:00	Lunch
14:30	Romaniello
15:00	Tokatly
15:20	Caffeine Break
15:50	Luppi
16:10	Sangalli
16:30	Stefanucci
16:50	Nanoquanta Meeting
20:00	Dinner

Friday 26	
09:00	Giorgetti
09:30	Jiang
09:50	Trevisanutto
10:10	Caffeine Break
10:40	Grüning
11:00	Bruneval
11:20	Mosca Conte
11:50	Fuchs
12:10	Lunch
15:00	Shaltaf
15:30	Neaton
15:50	Caffeine Break
20:00	Dinner

Saturday 27	
09:00	Friedrich
09:20	Schindlmayr
09:50	Caffeine Break
10:20	Puig von Friesen
10:50	Rohlfing
11:10	Farewell
	Departures
12:00	Lunch
	Departures
14:30	Nanoquanta Steering Committee
20:00	Dinner

30’ talks are in blue, 20’ talks are in black

Monday 22 September

17:00-20:00 Registration

Tuesday 23 September

09:00 Welcome

9:30-10:20 DFT Development and Application (I)

09:30 Leoor Kronik (*Weizmann Institute of Physics, Rehovoth, Israel*)
Theoretical photoemission spectroscopy of organic semiconductors

10:00 Verstraete (*Universidad del Pais Vasco, San Sebastián*)
Density Functional Perturbation Theory with Spin-Orbit Coupling: Phonon Band-Structure of Lead

10:20-10:50 Caffeine Break

10:50-11:50 DFT Development and Application (II)

10:50 Elena Sjakste (*Ecole Polytechnique, Palaiseau, France*)
Electron-phonon scattering in semiconducting nanostructures: ab initio approach

11:10 Volker Blum (*Fritz Haber Institute of the Max Planck Society, Berlin*)
"DFT and beyond" with localized orbitals: FHI-aims

11:30 Paula Havu (*Fritz Haber Institute of the Max Planck Society, Berlin*)
How Au(100) and Pt(100) reorganize themselves: Large-scale surface reconstructions studied by all-electron DFT

12:00-14:30 Lunch Break

14:30-16:40 Strongly Correlated System

14:30 Richard Martin (*University of Illinois, Urbana Champaign*)
Combining concepts and methods of GW and DMFT for calculation of spectra and thermodynamics of interacting electron systems

15:00 Nicole Helbig (*Universidad del Pais Vasco, San Sebastián*)
Kohn-Sham potential of strongly correlated systems

15:20-15:50 Caffeine Break

15:50 Franca Manghi (*Università di Modena e Reggio Emilia*)
e-e correlations in narrow-band systems

16:20 Sangeeta Sharma (*Freie Universität, Berlin*)
Reduced Density Matrix Functional for Many-Electron Systems

16:50-18:30 Poster Session

18:30 Aperitivo

Wednesday 24 September

9:00-10:10 Quantum Transport (I)

09:00 Massimiliano Di Ventra (*University of California, San Diego*)
Many-body effects in the electrical resistance of nanoscale conductors and fundamental limitations of the Landauer approach

09:30 Didier Mayou (*Institut Néel, Grenoble*)
Efficient algorithms for the simulation of quantum transport in nanodevices: application to graphene

10:00 Zeila Zanolli (*Université Catholique de Louvain*)
Gas sensors based on defective carbon nanotubes

10:20-10:50 Caffeine Break

10:50-12:10 Quantum Transport (II)

10:50 Stefano Sanvito (*Trinity College, Dublin*)
Can DFT quantum transport get it right?

11:20 Peter Bokes (*University of Technology, Bratislava*)
Non-linear and time-dependent quantum transport and its description using stroboscopic wavepacket basis

11:50 Andrea Ferretti (*Università di Modena e Reggio Emilia*)
GW and hybrid functional corrections to the calculation of transport properties in organic systems

12:10-15:00 Lunch Break

14:30-15:00 Experiment

14:30 Pierre Mallet (*Institut Néel, Grenoble*)
Two-dimensional quasiparticles probed by STM: Quantum interferences and wave function symmetry

15:00-17:10 Optical Properties

15:00 Margherita Marsili (*Università di Roma "Tor Vergata"*)
Quantum confinment effects on the electronic and optical properties of Ge nanocrystals

15:20 Maria Hellgren (*Lund University*)
Linear density response function within the time-dependent exact-exchange approximation

15:40-16:10 Caffeine Break

16:10 Marco Cazzaniga (*Università di Milano*)
The dynamic structure factor of simple metals: a study of the electronic correlation in solids

16:30 Roberto Guerra (*Università di Modena e Reggio Emilia*)
Optical properties of Silicon Nanocrystallites in SiO2 matrix: amorphization and size effects

16:50 Conor Hogan (*Università di Roma Tor Vergata*)
Optoelectronic properties of porphyrin oligomers

17:10-20:00 YR Meeting (Strictly-3-hours close doors session - beach forbidden!)

20:00 Social Dinner

Thursday 25 September

09:00-12:00 Quantum Transport (III)

09:00 Ferdinand Evers (*Karlsruhe University, Germany*)
Ab inito calculations for molecular electronics: density functional theories

09:30 Simon Dubois(*Université Catholique de Louvain*) Electronic Transport in zig-zag Graphene Nanoribbons

09:50 Stefan Kurth (*Freie Univeritát, Berlin*)
Time-dependent quantum transport: memory effects and bound-state oscillations

10:10-10:40 Caffeine Break

10:40 Jeff Neaton (*Molecular Foundry, Lawrence Berkeley National Laboratory*)
Understanding the Conductance of Single-Molecule Circuits from First Principles: Links, Length, and Switching

11:10 Frank Ortmann (*Friedrich-Schiller-Universität, Jena*)
Unified Theory of Charge Transport in Wide-Band and Narrow-Band Semiconductors

11:40 Robert van Leeuwen (*University of Jyväskylä, Finland*)
Initial correlations and memory in correlated quantum transport

12:00-15:00 Lunch Break

14:30-16:50 Theory Developments

14:30 Pina Romaniello (*École Polytechnique, Palaiseau*)
Double excitations in finite systems

15:00 Ilya Tokatly (*Universidad del País Vasco, San Sebastián*)
Quantum stress focusing: A new tool for visualizing a chemical structure of matter

15:20-15:50 Caffeine Break

15:50 Eleonora Luppi (*École Polytechnique, Palaiseau*)
Second-order harmonic generation: the effects of local fields.Applications to crystalline semiconductors.

16:10 Davide Sangalli (*Universitá degli Studi di Milano*)
Hounting double excitations

16:30 Gianluca Stefanucci (*Università di Roma Tor Vergata*)
Ultrafast manipulation of electron spins in a double quantum dot device: A real-time view

16:50-20:00 Nanoquanta General Meeting

Friday 26 September

09:30-10:10 Graphene Applications

09:00 Christine Giorgetti (*École Polytechnique, Palaiseau*)
Ab Initio calculations of electronic excitations in carbon nanotubes and graphene layers systems

09:30 Paolo Emilio Trevisanutto (*Institut Néel, Grenoble*)
Ab initio dynamical correlation effects in graphene: disentangling

09:50-10:10 f-electron Applications

09:50 Hong Jiang (*Fritz-Haber-Institut, Berlin*)
f-electron systems from the GW perspective:Applications to Rare Earth Oxides

10:10-10:40 Caffeine Break

10:40-12:10 Methods and Implementations

10:40 Myrta Grüning (*Université Catholique de Louvain*)
Frequency-dependent response of non-Hermitian Hamiltonians with real spectrum: an efficient and robust computational approach

11:00 Fabien Bruneval (*CEA, Gif-sur-Yvette*)
Accurate GW self-energy with only a few empty states

11:20 Adriano Mosca Conte (*Università di Roma Tor Vergata*)
Many-Body extended to QM/MM: applications to Indole in water solution

11:50 Frank Fuchs (*Friedrich-Schiller-University, Jena*)
Efficient O(N^2) approach to solve the Bethe-Salpeter equation for excitonic bound states.

12:10-15:00 Lunch Break

15:00-15:30 MBPT applications

15:00 Riad Shaltaf (*Université Catholique de Louvain*)
Band offsets prediction within many-body perturbation theory

15:30 Jeff Neaton (*Molecular Foundry, Berkeley*)
Brief introduction on the Molecular Foundry

15:50-16:20 Caffeine Break

Saturday 27 September

09:00-11:40 MBPT applications (II)

09:00 Christoph Friedrich (*Forschungszentrum Jülich*)
Efficient all-electron GW calculations of complex semiconductors and metals

09:20 Arno Schindlmayr (*Universität Paderborn*)
Spin excitations in solids from first principles

09:50-10:20 Caffeine Break

10:20 Marc Puig von Friesen (*Lund University*)
Non-equilibrium dynamics in strongly correlated clusters

10:50 Michael Rohlfing (*University of Osnabrueck*)
Binding energy of adsorbates on a noble-metal surface: exchange and correlation effects

11:10 Farewell and Lunch

14:30-20:00 Nanoquanta Steering Committee

Oral Contributions

Theoretical photoemission spectroscopy of organic semiconductors

Leeor Kronik

Department of Materials and Interfaces, Weizmann Institute of Science, Rehovoth 76100, Israel

Abstract

The combination of ultraviolet photoemission spectroscopy and density functional theory is an important approach to elucidating the electronic structure of a given material. In recent years, it has become a particularly popular tool for studying organic semiconductors and their interfaces with (metallic or semiconducting) inorganic substrates ? topics of great importance in organic electronics.

In this talk, I will review our recent progress in understanding the strengths, limitations, and true predictive power of such analyses. In particular, I will emphasize the role of self-interaction errors; the pros and cons of different functionals, including hybrid functionals and orbital-dependent approaches; and general criteria for selecting the best-suited theoretical approach and evaluating its accuracy. This is achieved through a comparison of density functional approaches with experimental photoemission spectra, as well as with many-body perturbation theory, for a variety of prototypical semiconducting organic molecules and organic/inorganic interfaces.

Density Functional Perturbation Theory with Spin-Orbit Coupling: Phonon Band Structure of Lead

Matthieu J. Verstraete[1,3,4], Marc Torrent[2], François Jollet[2], Gilles Zérah[2], Xavier Gonze[3]

(1) ETSF / Dept Fisica de Materiales Universidad del Pais Vasco
(2) Commissariat à l'énergie atomique, F-91680 Bruyères-le-Châtel, France
(3) Unité PCPM and CERMIN, Université Catholique de Louvain, Louvain-la-Neuve, Belgium
(4) Physics Dept University of York, UK

Abstract

The effect of spin-orbit coupling on phonon band structures can be profound for materials containing heavy elements. We describe our implementation of Density Functional Perturbation Theory with the spin-orbit interaction, for norm-conserving pseudopotentials. We show that the spin-orbit effect on the phonon frequency at the X point in face-centered cubic Pb is very large: it explains the discrepancy between calculated and experimental frequencies previously observed by De Gironcoli [Phys. Rev. B **53**, (1996) R7575]. Several technical issues (the exchange-correlation functional, the presence of semicore states, the pseudization scheme, and the real-space range of inter-atomic force constants) are also investigated.

Electron-phonon scattering in semiconducting nanostructures: ab initio approach.

J. Sjakste, N. Vast, V. Tyuterev

Ecole Polytechnique, Laboratoire des Solides Irradiés, CEA-DSM-DRECAM, CNRS, 91128 Palaiseau, France

Abstract

The interaction of excited electrons with phonons plays a central role for electronic and transport properties at the nanoscale. It is the dominant process limiting the excitation lifetime at medium excitation energies. Despite its importance, a reliable ab initio approach to describe the electron-phonon interaction in nanostructures is still lacking. The theoretical models used in the past, which are often oversimplified, need revisiting and justification. Recently, we developed a fully ab initio approach to calculate the electron-phonon constants and scattering times for collisions of carriers in the conduction band with short-wavelength phonons [1,2]. We applied it to the deexcitation of hot electrons in GaAs [1,2], and to the lifetime of the direct exciton in GaP and GaAs [2,3], all in excellent agreement with experiments. Our next goal is to study the effects of nanostructuring on the electron-phonon coupling constants. We will discuss our first results for the the electron-phonon coupling constants in GaAs/AlAs superlattices.

[1] J. Sjakste, N. Vast, V. Tyuterev, Phys. Rev. Lett. **99**, 236405 (2007).

[2] J. Sjakste, V. Tyuterev, N. Vast, Appl. Phys. A **86**, 301 (2007).

[3] J. Sjakste, V. Tyuterev, N. Vast, Phys. Rev. B **74**, 235216 (2006).

“DFT and beyond” with localized orbitals: FHI-aims

Volker Blum, Ralf Gehrke, Felix Hanke, Paula Havu, Ville Havu, Xinguo Ren, Patrick Rinke, Karsten Reuter, Matthias Scheffler

Fritz Haber Institute of the Max Planck Society Theory Department Faradayweg 4-6, D-14195 Berlin, Germany

Abstract

We describe the Fritz Haber Institute “*ab initio* molecular simulations” (FHI-aims) computer code package for an efficient, accurate all-electron, full-potential “density functional theory and beyond” description of molecules and periodic systems, including (for molecular systems) capabilities for hybrid functionals, Hartree-Fock based MP2 perturbation theory, and GW corrections. FHI-aims is based on localized, numerically tabulated atom-centered orbitals with preconstructed, hierarchical basis sets, allowing to move from a fast qualitative description up to meV-level total energy convergence with the basis set. A low-communication parallelization of all real-space grid based algorithms and ScaLapack-based linear algebra for all matrix operations guarantee efficient scaling (CPU time and memory) up to massively parallel computer systems with thousands of CPUs. These capabilities are here demonstrated by investigating the stability of secondary structure (helices) formed in Polyalanine peptides, and its implications for spectroscopy.

How Au(100) and Pt(100) reorganize themselves: Large-scale surface reconstructions studied by all-electron DFT

Paula Havu[1], Ville Havu[1], Patrick Rinke[1,2], Volker Blum[1], and Matthias Scheffler[1,2]

1) Fritz-Haber-Institut, Berlin, Germany
2)University of California at Santa Barbara, CA 93106, USA

Abstract

Pt(100) and Au(100) show large-scale quasi-hexagonal surface reconstructions, often called (5×20), that are intimately linked to the external surface conditions (temperature, absorbates, electrochemistry). Using density-functional theory (DFT), these classic reconstruction have been studied by (5×1) approximants in the past, but not, to our knowledge, in the true large-scale reconstructed form. We here use the new numerical atom centered orbital based all-electron, full-potential code FHI-aims [1] to determine the full (5×20)-reconstructed surface structure, relying on (1) good scaling of CPU time and memory with system size, (2) the fully parallel nature of the code, and (3) the adaptation of the code for IBM's BlueGene architecture up to thousands of processors with strict per-process memory limits (512 MB). For both surfaces, we investigate the reconstruction energetics and associated electronic structure changes, and conclude that the underlying electronic mechanism is similar for both systems.

Combining concepts and methods of GW and DMFT for calculation of spectra and thermodynamics of interacting electron systems

Richard M. Martin

University of Illinois at Urbana-Champaign and Stanford University

Abstract

The purpose of this talk is to present a set of questions and possible approaches for calculation of excitation spectra and thermodynamics of strongly-coupled electron systems, building upon the virtues of GW and dynamical mean field theory (DMFT). The strong point of DMFT is that it provides a solution of an interacting system in a small selected basis, with other degrees for freedom treated as a dynamical mean field. It is exact both in the limit of a small decoupled system and in the limit of an extended non-interacting problem, and it provides an interpolation in a real problem. In contrast GW is a perturbation approach that treats the full basis. Many people have suggested that the two approaches can be combined to make an accurate ab initio method for real condensed matter problems. In this talk, I want to discuss ways in which DMFT can be used to add vertex corrections in GW and to provide a description of low energy excitations that are critical for thermodynamics and phase transitions. Conversely, GW can provide ways to incorporate realistic frequency-dependent interactions in DMFT and eliminate (or at least reduce) the problem that the results of present DMFT calculations depend upon the choice of basis. Finally, we can consider open questions of how to go beyond mean-field methods, e.g., to include fluctuations important for critical phenomena and other properties.

Kohn-Sham potential of strongly correlated systems

N. Helbig, I.V. Tokatly, A. Rubio

Universidad del Pais Vasco, San Sebastian, Spain

Abstract

The description of strongly correlated systems within density functional theory has proven very difficult in the past. The dissociation of molecules, even the most simple hydrogen molecule, cannot be described accurately with any of the presently available density functionals. These problems have led to a discussion of what properties the local Kohn-Sham potential has to satisfy in order to correctly describe strongly correlated systems. In order to shine some light onto this discussion, we present calculations for one-dimensional model systems which are exactly solvable. We show that it is indeed possible to describe the dissociation of one-dimensional hydrogen correctly and present the exact Kohn-Sham potential for this system. We also address the question of the asymptotic behavior of the Kohn-Sham potential for different diatomic configurations. This study provides important hints to how static Hubbard correlations are build into the Kohn-Sham system.

e-e correlations in narrow-band systems

Franca Manghi

Università di Modena e Reggio Emilia, Modena, Italy

Abstract

Theoretical description of interacting many particle systems remains one of the grand challenges in condensed-matter physics. The field of strongly correlated electron systems has gained theoretical and experimental interest trough the discovery of new materials and new low dimensional structures where the electron localization is responsible of peculiar properties ensuing from e-e correlations. In this class of systems the strength of the interaction between particles is comparable or larger than the kinetic energy and any theory based on perturbation expansion around the non-interacting state is questionable. The Hubbard model contains the essential physics of highly correlated materials and much effort has been devoted in the last decades to devise solutions of Hubbard Hamiltonian for real materials, where the full complexity of ab-initio band structure is taken into account together with many-body interactions.

In this talk I will describe the approach "3-Body-Scattering-Theory" that we have developed to augment ab-initio band structure with many-body e-e interactions and provide spin- and k-dependent self-energies, hole spectral functions, quasiparticle energies and lifetimes. Applications to some non trivial test cases (1D and 2D Hubbard models) will allow us to address fundamental issues such as the correlation-driven metal-insulator transition in low dimensional solids and the nature of quasi particle excitations. Comparison with other theoretical approaches will also be discussed together with results obtained for real materials.

Reduced Density Matrix Functional for Many-Electron Systems

Sangeeta Sharma

Institut für Theoretische Physik Freie Universität Berlin Arnimallee 14, D-14195 Berlin Germany

Abstract

Reduced density matrix functional theory for the case of solids is presented anda new exchange correlation functional based on a fractional power of the density matrix is introduced. We show that compared to other functionals, this produces more accurate results for both finite systems and the homogeneous electron gas. Moreover, it captures the correct band gap behavior for conventional semiconductors as well as strongly correlated Mott insulators, where a gap is obtained in the absence of any magnetic ordering.

Many-body effects in the electrical resistance of nanoscale conductors and fundamental limitations of the Landauer approach

Massimiliano Di Ventra

Department of Physics, University of California, San Diego

Abstract

I will discuss the fundamental incompleteness of the Landauer multi-terminal formula for the conductance of a nanoscale system due to many-body effects which cannot be treated as contributions to the single-particle transmission probabilities. The physical origin of these effects is related to the viscous nature of the electron liquid which gives rise to non-linear electron flow, electron heating, etc. [1]

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Efficient algorithms for the simulation of quantum transport in nanodevices : application to graphene

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Abstract

We present efficient algorithms, based on recursion like methods, to compute quantum transport either in the Kubo-Greenwood formalism or in the Landauer type approach. The algorithm for the Kubo-Greenwood formalism allows to compute bulk properties and gives access to diffusivity, low frequency conductivity and to characteristics lengthes such as mean-free path, localization length. The algorithm for the Landauer approach allows to treat the problem of contact resistance in a very efficient way. As examples we discuss recent studies of quantum transport in graphene based nanodevices. We discuss in particular the problem of transport in functionalized graphene. We demonstrate also a divergent contact resistance at the Dirac point and analyse this effect and its potential use for nanodevices.

Gas sensors based on defective carbon nanotubes

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Abstract

The quest for gas sensors with high sensitivity and selectivity has recently attracted large interest not only for industrial and environmental monitoring applications, but also for the experimental and theoretical challenges raised by this issue. When gas molcules adsorbs on the surface of a nanodevice, a charge transfer between the molecule and the nanomaterial occurs, thus affecting the device electrical conductivity. In this spirit, devices having carbon nanotubes (CNT) or graphene as sensing element have been recently proposed [1]. To fully exploit the potentialities of CNTs as gas sensors, it is essential to investigate the key features of the gas–CNT interaction. In addition, to provide a realistic description of the problem, it is crucial to consider that CNTs have defects and their electronic and transport properties will be affected accordingly [2]. As the aim of the present work is the study of the gas–CNT interaction at the atomistic level, a first-principles density functional theory (DFT) approach is needed to accurately describe the electronic structure of the nanodevice. Due to the large number of atoms involved in the simulation, a pseudopotential atomic orbitals basis set method [3] has been chosen to investigate the ground state properties of the defective tube. In addition, this method allows one to identify the most likely adsorption configurations via Born-Oppenheimer quantum molecular dynamics. The electronic transport calculations have been performed within the non-equilibrium Green's (NEGF) functions formalism [3]. By following this approach, the interaction between defective CNTs and several gas species has been investigated. Our results suggest that the binding of NO_2 to the tube induces a major change in its electronic quantum conductance. Consequently, defective CNTs are potentially good candidates for the selective detection of gas with high sensitivity.

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Can DFT quantum transport get it right?

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Abstract

Density functional theory combined with the non-equilibrium Green's function method for quantum transport has now become the mainstream method for first principles transport calculation. Despite the fact that some fundamental aspects still remain unsolved, the success of this method has been demonstrated 'on the field' with predictions of I-V characteristics for systems ranging from molecular junctions to atomic point contacts to magnetic tunnel junctions. In this talk I will review the most recent advances of our DFT quantum transport code Smeagol, and in particular I will discuss the possibility of large scale simulations for physical and biological systems. Examples from transport across DNA strands and magnetic molecules will be presented. In the second part of my talk I will instead review some recent progress on time-dependent quantum/classical simulations for open systems. In particular I will present recent results on the current-induced dynamics of domain walls in mono-atomic nanowires.

Non-linear and time-dependent quantum transport and its description using stroboscopic wavepacket basis

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Abstract

We introduce the construction of a orthogonal wavepacket basis set, using the concept of stroboscopic time propagation, tailored to the efficient description of non-equilibrium extended electronic systems. Thanks to three desirable properties of this basis, significant insight is provided into non-equilibrium processes (both time-dependent and steady-state), and reliable physical estimates of various many-electron quantities such as density, current and spin polarization can be obtained. Use of the wavepacket basis provides new results for time-dependent switching-on of the bias in quantum transport, and for current-induced spin accumulation at the edge of a 2D doped semiconductor caused by edge-induced spin-orbit interaction.

GW and hybrid functional corrections to the calculation of transport properties in organic systems

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Abstract

The unprecedented technological development in the microelectronics industry eventually arrives at its ultimate limit when individual electronic devices reach the nanoscale. The design of these systems will rely on a accurate quantum-mechanical ab-initio modeling of electronic transport through them. In the current work, we will focus on organic junctions made by single molecules or self-assemble monolayers (SAM). For these systems, many experimental results exist (see e.g. the case of alkane or aromatic thiols), mainly providing the decay factor of the current as a function of the molecule length or the plain I/V characteristics. Nevertheless, the theoretical description is partly lacking, both for the variety of the molecules considered and for the methods adopted. In this work, we studied the effects of GW corrections and hybrid exchange-correlation functionals on the calculation of beta decay factors, according to a method based on the evanescent states given by the complex band structure formalism. We apply the method to a number of oligomers both aliphatic and aromatic. On the other side, the effects of GW corrections on the full conductance through a prototype system, i.e. benzene dithiol connected to gold, are also investigated.

Quantum confinment effects on the electronic and optical properties of Ge nanocrystals

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Abstract

Recent measurements demonstrate that Ge nanocrystals act as photocurrent generators in an electrochemical cell [1,2], giving the chance of exploiting Ge dots for photovoltaic nanodevices. The experiments show sizable modifications of the electronic gap and of the photocurrent spectra of this system due to quantum confinement effects. The electronic gap, measured by scanning tunneling spectroscopy (STS), increases while decreasing the dot size, in agreement with other experimental results [3]. However the experimental points seem to lie above the electronic gap vs dot radius trend predicted by theory by means of a density functional theory (DFT) based method, namely Δ-SCF [4]. Here we present the results of *ab initio* calculations for the electronic and optical properties of Ge nanocrystals of different sizes. The electronic gap is computed at the GW level within the framework of many-body perturbation theory. This state-of-the-art tool for the determination of the electronic structure of materials gives direct access to the single-particle excitation energies of the system. Our results show that the DFT-based method of Ref. [4] predicts smaller gaps with respect to GW. The optical properties at different dot sizes are computed within both the time-dependent density-functional theory (TDDFT) and the Bethe-Salpeter Equation (BSE) scheme. The comparison with previous TDDFT calculations[5] and between the two methods is presented. A possible link to the photocurrent spectra is discussed.

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Linear density response function within the time-dependent exact-exchange approximation

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Abstract

We have calculated the frequency-dependent exact-exchange (EXX) kernel of time-dependent (TD) density functional theory employing our recently proposed computational method based on cubic splines. Apart from analyzing the kernel itself we have calculated the linear density response function and obtained discrete excitation energies, photoabsorption spectra as well as correlation energies for some spherical spin compensated atoms. As might be expected, the results of the TDEXX approximation are close to those of TD Hartree-Fock theory. In addition, correlation energies obtained by integrating over the strength of the Coulomb interaction turn out to be highly accurate.

The dynamic structure factor of simple metals: a study of the electronic correlation in solids

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Abstract

The study of simple metals is particularly appealing due to their nearly free electron behaviour. The comparison with the simple jellium model gives access to a more detailed knowedge of the origin of what can be observed. IXS spectra can provide information of the effect arising by correlations beyond RPA. In this work we compare experimental and calculated spectra for sodium and aluminum. The presence of the effects due to the real lattice are not enough to explain all the differences in the comparison. We will show that an accurate description of correlations is also required to provide a reasonable agreement, expecially in the case of sodium. TDLDA with the inclusion of lifetime effects provides a good agreement for small and intermediate momentum transfer q, similarly to what is observed for silicon [1], altough for very large q the agreement becomes worse. Moreover this approximation fails in describing a structure, interpreted as plasmon-plasmon peak [2], which can be observed in the experiment. On the other hand, the inclusion of lifetimes as a transition-dependent broadening in the independent-particle polarizability is not rigorous in the linear response framework. For this reason we will discuss the possibility of constructing a kernel able to reproduce the same effect. In this direction we will present the results of a dynamical TDLDA kernel with an imaginary part [3], and different approaches to such a problem.

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Optical properties of Silicon Nanocrystallites in SiO2 matrix: amorphization and size effects

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Abstract

Embedding silicon nanocrystals (Si-nc) in a silica matrix is one way to achieve high optical gain that can be used to fabricate devices as demonstrated by Pavesi and coworkers [1]. This discovery has attracted a lot of interest on this type of systems and produced a lot of debates on their optoelectronic properties and especially on the gain mechanisms. One of the goals of recent works was to answer the fundamental question whether the energy gaps of Si-nc with different sizes, which are embedded in amorphous silica, follow or deviate from the quantum confinement (QC) model that seems essential for the visible light emission in crystalline Si nanostructures. What emerges is that, while QC tend to increase the band-gap, small clusters are more sensitive to defects and surface configurations. Moreover, debated results have been obtained for deviations of the Si-nc from the crystalline geometry towards the amorphic state, in which the Si-nc get heavily distorted, sometimes producing a greater and sometimes a smaller band-gap [2-4]. The question of surface effects, in particular oxidation, has been addressed in the last years. Many theoretical [5,6] and experimental works have demonstrated that the interface properties have dramatic effects on the emission properties. On the other hand, absorption measurements are less sensitive to surface quality and allow for a more direct characterization of the intrinsic structure of Si-nc, being also easier to compute. Therefore, the study of the direct absorption can provide a clear physical understanding of the Si-nc properties, and until now researcher have mainly focused on the interband absorption process, that is the only optical absorption possibility for an intrinsic semiconductor nanocrystal under equilibrium. Some studies already tried to investigate the excited states of Si-nc, which are the basis for the understanding of the emission properties, but any known work included the effect of the host insulating matrix. Here we present first-principles calculations of the structural, electronic and optical properties of different sizes Si-nc embedded in a SiO2 matrix considering both the crystalline and the amorphous case. Our aim is to study the effect of the embedding matrix and of its crystallinity or not on the properties of Si-nc with respect to the free standing system. For these systems we will show the first ab-initio calculation of the optical gain highlighting the effects of QC and of crystalline order and disorder. We will then discuss the experimental findings in the light of our results in order to investigate the possibility and best conditions to obtain gain from in Si-nc dispersed in a silica matrix.

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Ab inito calculations for molecular electronics: density functional theories

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Abstract

Transport calculations for single molecules pose a considerable challenge to a theoretical analysis, because a profound understanding of the hybridization of the molecular many-body state with the electrodes is required. In order to achieve the system sizes necessary it is common practice to combine the density functional theory (DFT) with a Landauer-Buttiker-type formalism. In our presentation, we shall review attempts to justify this approach for the linear conductance. In particular, we present a model calculation which demonstrates that the approach yields quantitative results for the (generalized) interacting resonant level model in the regime of strong Coulomb blockade. The talk closes with a discussion of the impact of the local density approximation in common DFT-functionals on transport coefficients.

Electronic Transport in zig-zag Graphene Nanoribbons

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Abstract

Recent theoretical investigations have pointed out the strong connection between the electronic structure of Graphene nanoribbons (GNRs) and their geometry [1]. Hydrogenated GNRs with armchair shaped edges exhibit a band gap which arises from both quantum confinement and the crucial effect of the edges. In the GNRs with zig-zag shaped edges a gap also appears but because of edge magnetization. This opens a route to promising applications for use in future spin based electronics.

Besides the magnetization due to the borders, defect-mediated magnetism is also expected in carbon structures [2]. Structural defects, in general, give rise to localized electronic states around the Fermi level with possible magnetization. In this work, the effect of point defects on the structural and electronic properties of zig-zag GNRs is investigated using spin-polarized DFT calculations. By analogy to the work of Lethinen et al. [2] on the defect-induced magnetism in graphite, I have considered the vacancy and the carbon adatom. The interaction between the spin ordered edges states and the point defect has been carefully taken into account. The carbon adatom appears to stabilize the parallel spin orientation between the edges giving rise to a spin polarized electronic transmission function around the Fermi level. These results underline the interest of defect-doped zig-zag GNRs for use in future spin-based electronic devices.

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Time-dependent quantum transport: memory effects and bound-state oscillations

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Abstract

Our numerical algorithm for the time propagation of the Schrödinger equation for open systems which is suited for investigation of electron transport with time-dependent density functional theory is used to study time-dependent transport phenomena such as electron pumping, transients and the role of bound states. The presence of at least two bound states in the biased electrode-device-electrode system not only leads to current oscillations which remain undamped in the long-time limit but also to contributions to the non-equilibrium density which depend on the history of the time-dependent bias or gate voltage. The time-dependent formulation leads to a natural definition of occupation numbers for bound states under non-equilibrium conditions both inside and outside the bias window. We also report on recent progress in the implementation of the transport algorithm into the TDDFT software package OCTOPUS.

Understanding the Conductance of Single-Molecule Circuits from First Principles: Links, Length, and Switching

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Abstract

A fundamental challenge in nanoscience is to understand and control electron flow through individual molecules and nanostructures. Recently, the electrical conductance of single-molecule junctions - small aromatics linked to macroscopic gold electrodes by amine and pyridine endgroups - has been reliably and reproducibly measured using modified break junction techniques [1]. These and other recent experiments provide an opportunity to benchmark standard first-principles methods while quantitatively exploring fundamental concepts in molecular-scale charge transport. Here I will describe work [2,3] using a scattering-state technique based on density functional theory (DFT) to understand reported transport measurements of these single-molecule junctions. Using a physically motivated approximate self-energy correction based on GW calculations of a model metal-molecule interface [4], we explore quantitatively how binding geometry, link chemistry, and molecular chain length affect transport properties and, in some cases, lead to novel phenomena, such as mechanically-controlled conductance switching. The importance of many-electron corrections beyond DFT for accurately computing molecular conductance - and understanding experiments - is discussed in the context of our results. This work was supported in part by the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

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Unified Theory of Charge Transport in Wide-Band and Narrow-Band Semiconductors

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Abstract

An intrinsic material property of semiconductors highly relevant for charge transport is the mobility tensor of electrons and holes. In the literature, it is often calculated within one of the two limiting cases: wide bands or narrow bands. In the case of wide-band systems, usually pure band transport is assumed and characterized by a mean velocity of the charge carriers, which depends on the applied electric field and a phenomenological relaxation time. In contrast, for narrow-band materials, hopping is usually considered prevalent and the interaction with lattice vibrations is described within the nonperturbative polaron concept. In this talk, we will present a unified approach to the description of charge transport in both wide-band and narrow-band materials. The unification is based upon the Kubo formalism applied to a Holstein Hamiltonian for electron-phonon coupling. The temperature-dependent mobility tensors are calculated for arbitrary bandwidths. As an analytical result, we obtain a seamless transition from band transport at low temperatures to hopping transport at high temperatures. The results are illustrated for a prototypical model system [1] and a detailed comparison to previous approaches [2,3] is made.

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Initial correlations and memory in correlated quantum transport

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Abstract

We study time-dependent quantum transport in a correlated system by means of time-propagation of the Kadanoff-Baym equations for the nonequilibrium many-body Green function. We consider an initially contacted equilibrium system of a correlated central region coupled to tight-binding leads. Subsequently a time-dependent bias is switched on after which we follow in detail the time-evolution of the system. Important features of the Kadanoff-Baym approach are 1) the possibility of studying the ultrafast dynamics of transients and other time-dependent regimes and 2) the inclusion of exchange and correlation effects in a conserving approximation scheme. We find that initial correlation and memory terms due to many-body interactions have a large effect on the transient currents. Furthermore the value of the steady state current is found to be strongly dependent on the approximation used to treat the electron interactions.

Double excitations in finite systems

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Abstract

Time-dependent density-functional-theory (TDDFT) is widely used in the study of linear response properties of finite systems. However, there are difficulties in properly describing excited states which have double- and higher-excitation character, particularly important in molecules with open-shell ground-state. These states would be described if the exact TDDFT kernel were used; however, within the adiabatic approximation to the exchange-correlation (xc) kernel the calculated excitation energies have strict single-excitation character, and are fewer than the real ones [1]. A frequency-dependent xc kernel could create extra poles in the response function, which would describe states with multiple-excitation character. We introduce a frequency-dependent xc kernel which can reproduce, within TDDFT, double excitations in finite systems. In order to achieve this, we use the Bethe-Salpeter equation with a dynamical screened Coulomb interaction $W(\omega)$ which can describe these excitations [2,3]. We test this kernel on a two-electron model systems, and we find that poles corresponding to double excitations are produced. However, together with the expected energies, another pole corresponding to a unphysical state appears. We attribute this deficiency to the self-screening problem the description of $W(\omega)$ suffers from.

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Quantum stress focusing: A new tool for visualizing a chemical structure of matter

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Abstract

I show that several important concepts of descriptive chemistry, such as atomic shells, bonding electron pairs and lone electron pairs, may be described in terms of *quantum stress focusing*, i.e. the spontaneous formation of highly-stressed regions in an electron gas. This description subsumes previous mathematical constructions, such as the Laplacian of the density and the electron localization function, and provides a new tool for visualizing chemical structure of matter. I also demonstrate that the full stress tensor, defined as the derivative of the energy with respect to a local deformation, can be easily calculated from density functional theory.

Second-order harmonic generation: the effects of local fields.Applications to crystalline semiconductors.

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Abstract

Second harmonic generation (SHG) is a nonlinear optical spectroscopies largely used as a noninvasive tool to study properties of a systems because of its symmetry properties.
SHG is linked to the second-order nonlinear susceptibilities: χ^2, which theoretical determination is very complex and these difficulties have delayed any accurate calculations for many years. We have formulated a derivation for the calculation of the χ^2 for crystals of any simmetry within the TDDFT including the effects of the microscopic response of the electrons: local fields effects. In particular which is the effects of local fields in nonlinear spectroscopy like SHG ? Numerical results will be presented for several materials with zinc-blende structure.

Hounting double excitations

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Abstract

The spectra of isolated systems is known to be described quite well by Casida's approach to Time-Dependent Density-Functional-Theory (TDDFT) for many closed shell systems, within the adiabatic approximation. On the other hand the same framework is known to perform quite poorly for open shell systems. The reason of this deficiency can be found in the failure of the adiabatic approximation in describing double excitations, that leads, therefore, to the break of the spin symmetries of the system[1]. One would encounter analogues deficiencies using the Bethe Salpeter (BSE) approach with a static kernel $W^{RPA}(\omega = 0)$. A solution to the problem is the use of dynamical kernels. This work aims to scrutinize the dynamical structure that the BSE kernel and/or the exchange correlation kernel of TDDFT should have in order to describe double excitations. Following the same line of previous works on the study of removal/addition of electrons [2] and of excited states of nuclei [3,4], we find out that second order Feynman diagrams should be included in the kernel. We show some preliminary tests on model systems. Finally we focus our attention on the dynamic $W^{RPA}(\omega)$ kernel: we give some guidelines for future developments.

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Ultrafast manipulation of electron spins in a double quantum dot device: A real-time view

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Abstract

We consider a double single-level quantum dot system with two embedded and non-aligned spin impurities to manipulate the magnitude and polarization of the electron spin density. The device is attached to semi-infinite one-dimensional leads which are treated exactly. We provide a real-time description of the electron spin dynamics when a sequence of ultrafast voltage pulses acts on the device. The numerical simulations are carried out using a spin generalized and modified version of a recently proposed algorithm for the time propagation of open systems [Phys. Rev. B **72**, 035308 (2005)]. Time-dependent spin accumulations and spin currents are calculated during the entire operating regime which includes spin injection and read-out processes. The full knowledge of the electron dynamics allows us to engineer the transient responses and improve the device performance. An approximate rate equation for the electron spin is also derived and used to discuss the numerical results.

Two-dimensional quasiparticles probed by STM: Quantum interferences and wave function symmetry

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Abstract

In this review, I will demonstrate the power of the scanning tunnelling microscopy (STM) for probing the low energy quasiparticles of two dimensional (2D) or nearly 2D systems lying at surfaces. I will mainly focus on the analysis of the quantum interferences generated by elastic scattering of the 2D quasiparticles off static impurities. Such analysis provides quantitative information about the 2D band structure of the system, and also in some cases gives insights on inelastic scattering processes. I will also present recent experiments evidencing that the symmetry of the quasiparticle wave functions can prevent some impurity-induced elastic scattering processes. This will be illustrated in epitaxial graphene on SiC, for which STM can be used to check the unique quasiparticles symmetry (chirality) predicted for ideal free standing graphene.

f-electron systems from the GW perspective:Applications to Rare Earth Oxides

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Abstract

Understanding the physics of f-electron systems is regarded as a great challenge in condensed-matter physics today. In many of these materials the strong localization of the f-electrons gives rise to large many-body interaction effects and in addition leads to severe self-interaction errors in the local-density approximation (LDA). Many-body perturbation theory in the G_0W_0 approximation can describe screening effects among itinerant electrons accurately, and moreover treat exchange exactly. It is therefore a promising approach for investigating these systems. In this work we first apply the G_0W_0 method to CeO_2 and ThO_2, the "simplest" f-electron systems for which the LDA provides a qualitatively correct description, but underestimates band gaps significantly. For both materials, G_0W_0 based on LDA provides an accurate description for the fundamental valence-conduction band gap and the position of f-states. We further apply the G_0W_0 method to R_2O_3 (R=Ce, Pr, and Nd), in which the f-shell is partially occupied. Since LDA gives qualitatively wrong metallic ground states for these materials, we apply G_0W_0 corrections on top of LDA+U calculations. We find that 1) The density of states of Ce_2O_3 is in good agreement with experiment; 2) Band gaps of R_2O_3 for R=La, Ce, Pr and Nd agree quantitatively with optical experimental results; and 3) The trend observed in the relative position of f-bands is in accord with the phenomenological conjecture derived from high-temperature conductivity experiments. Our work shows that the LDA(+U)-G_0W_0 method can treat both itinerant *spd* bands and localized f-bands accurately for the materials we have considered.

Ab initio dynamical correlation effects in graphene: disentangling kinks

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Abstract

The unconventional electronic structure of graphene, with a massless cone-like band dispersion, has raised increasing interest in view of applications in nano-electronics. The non-interacting valence and conduction linear band dispersions crossing at a single 0-bandgap point can be affected by many-body self-energy effects. Here we perform an *ab initio* numerical calculation of quasiparticle bands in real free-standing graphene by an electronic self-energy in the GW approximation. We find that electron-electron interactions substantially renormalize the bands and raise the Fermi velocity by a 17% with respect to density-functional theory, thus providing an *ab initio* estimate of 1.12 ms^{-1} in good agreement with magnetotransport experiments. Furthermore, close to the Dirac point the linear dispersion is modified by the presence of a kink, as observed in ARPES experiments, which we find as due to low-energy $\pi \rightarrow \pi^*$ single-particle excitations and to the π plasmon. Finally, many-body GW self-energy effects do not open a gap at the Dirac point.

Frequency-dependent response of non-Hermitian Hamiltonians with real spectrum : an efficient and robust computational approach.

Myrta Grüning, Andrea Marini,and Xavier Gonze

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Abstract

Compared to their Hermitian Hamiltonian counterparts, computational methods for the treatment of non-Hermitian Hamiltonians are often less efficient and less stable, sometimes leading to the breakdown of the method. Actually, the fundamental properties of a wide set of non-Hermitian Hamiltonians with real spectrum can yield computational techniques with the same advantages as Hermitian ones. We design a Lanczos-type algorithm for the computation of frequency-dependent responses, and apply it to the optical response of nanostructures and biomolecules.

Accurate GW self-energy with only a few empty states

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Abstract

The GW approximation to the electronic self-energy yields band structures in excellent agreement with experimental data. Unfortunately, this type of calculation is extremely cumbersome even for nowadays computers. The huge number of empty states required both in the calculation of the polarizability and of the self-energy is a major bottleneck in GW calculations. We propose an almost costless scheme, which allows to divide the number of empty states by about a factor of five to reach the same accuracy. The computational cost and the memory requirements are decreased by the same amount, accelerating all calculations from small primitive cells to large supercells.

Many-Body extended to QM/MM: applications to Indole in water solution

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Abstract

We present a method to calculate the optical properties of large scale systems, based on many body perturbation theory combined with quantum-mechanics/molecular-mechanics (QM/MM). We show how to include quasi-particle and excitonic effects for the calculation of optical absorption spectrum in a QM/MM scheme. This method is particularly indicated to calculate optical properties of relevant biological molecules such as chromophores, that are used to probe complex biological processes in vitro and in vivo, yet how the environment tunes their optical properties is far from being fully understood. A well known example is given by indole. We apply therefore this scheme, together with the well established TDDFT approach, to indole in water solution. Our calculations show that the solvent induces a red shift in the main spectral peak of indole, in quantitative agreement with the experiments and point to the importance of performing averages over molecular dynamics configurations for calculating optical properties.

Efficient O(N^2) approach to solve the Bethe-Salpeter equation for excitonic bound states.

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Abstract

The accurate calculation and parameter-free prediction of optical spectra including excitonic effects is highly desirable for both fundamental and applied research. Excitonic effects can be treated in the framework of many-body perturbation theory by solving the Bethe-Salpeter equation (BSE), which can be recast in an eigenvalue problem for the electron-hole Hamiltonian $\hat{H}$. Although for the computation of excitonic optical spectra in an extended frequency range efficient methods are available, the determination and analysis of individual exciton states (e.g. excitonic bound states or dark excitons) still requires the diagonalization of the electron-hole Hamiltonian $\hat{H}$. We present a numerically efficient approach for the calculation of exciton states with quadratically scaling complexity, which significantly diminishes the computational costs compared to the commonly used cubically scaling direct-diagonalization schemes. The accuracy and performance of this approach is demonstrated by solving the BSE numerically for the Wannier-Mott two-band model in k space and the semiconductors MgO and InN. For the convergence with respect to the k-point sampling a general trend is identified, which can be used to extrapolate converged results for the binding energies of the lowest bound states.

Ab Initio calculations of electronic excitations in carbon nanotubes and graphene layers systems

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Abstract

In this talk, I will present some recent ab initio calculations of electronic excitations in carbon nanotubes (CNT), graphene layers and graphite, obtained in our group. The ground state properties are calculated with the AbInit code (1) and the electronic excitations in TDDFT using the DP code (2).

In order to simulate the dispersion of the π plasmon along the tube axis in vertically aligned single wall nanotubes, we calculated the Electron Energy Loss Spectra (EELS) for a graphene layer and a bi-layer graphene system to simulate isolated CNT, and tubes interacting in bundles respectively. The dispersion of EELS for in-plane transfered momenta in graphene reproduced the linear dispersion of the π plasmon along the tube axis for the isolated tubes. The comparison of Independant Particle spectra and spectra including the LFE allowed us to analyze the origin of this behavior (3).

The π plasmon measured in CNT present a second non dispersing structure, which has been attributed to a plasmon arising from momenta transfered perpendicular to the tube axis, and was not present in EEL spectra done in graphene. To account for this double plasmon with different dispersions, we calculated EELS for (3,3) CNT for transfered momenta having different orientations as compared to the tube axis. These spectra show the double plasmon structure with a non dispersing component coming from perpendicular transfered momentum, and a dispersing one. The origin of these two structures will be discussed. This subject has been selected as an ETSF user project.

Since the calculations on the tubes are cumbersome, they are limited to small diameter tubes. Thus we continue the investigation of the possibility to study electronic excitations for large diameter tubes with planar systems. In order to study the interaction between tubes in bundles or walls in multiwalls CNT, I will present some preliminary results on graphene layers systems with different kind of stacking.

(1) http://www.abinit.org

(2) http://dp-code.org

(3) C. Kramberger et al. Phys. Rev. Lett. **100**, 196803 (2008)

Optoelectronic properties of porphyrin oligomers

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Abstract

Porphyrin oligomers (short chains of porphyrins covalently linked by small molecular bridges) are of great interest nowadays for biosensors and the optoelectronics industry. Thanks to the pi-conjugation, they are characterized by high polarisabilities, large oscillator strengths, and low electronic transitions, and hence show much potential for efficient organic solar cells. These properties depend sensitively on particular structural components, such as the functional groups, the chain length, and the type of central metal atom and molecular bridge, which determine the localization and formation of excitons.

In this presentation we describe ab-initio calculations of electronic and optical properties of conjugated porphyin systems. Excited states are computed using the GW approximation for the self-energy, and optical spectra are computed by solving the Bethe-Salpeter equation. Physical and technical problems encountered during the calculations of the isolated molecules are addressed. Finally, we discuss the observed spectral dependence and excitonic properties on the chain length.

Molecular Foundry

J. B. Neaton

Molecular Foundry, Lawrence Berkeley National Laboratory

Abstract

Description of the Molecular Foundry.

Efficient all-electron GW calculations of complex semiconductors and metals

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Abstract

The *GW* approximation for the electronic self-energy yields quasiparticle band structures in very good agreement with experiment, but almost all implementations so far are based on the pseudopotential approach, which limits their range of applicability. We have developed an implementation (SPEX, http://www.flapw.de/spex/) within the full-potential linearized augmented-plane-wave (FLAPW) method, which treats core and valence electrons on an equal footing. Within this method a large variety of materials can be treated, including d- and f-electron systems, oxides and magnetic systems. Our implementation employs a mixed product basis for the representation of basis-function products in the interstitial and muffin-tin regions. The self-energy can be evaluated either by an analytic continuation from the imaginary frequency axis or directly by a contour integration, which makes a comparison of the two approaches possible. A basis transformation to the eigenfunctions of the Coulomb potential allows a reduction of the basis-set size without compromising the accuracy, thus leading to a considerable speed-up in computation time. To demonstrate the wide applicability and accuracy of the implementation we present an overview of results for a variety of semiconductors. We then discuss results for two prototype ABO_3 semiconductors, paraelectric $SrTiO_3$ and ferroelectric $BaTiO_3$, which are widely used in electronic applications. Their band gaps are difficult to measure experimentally and have been under debate for a long time. Ab initio theoretical studies of their electronic structure have mostly been based on density-functional theory so far , which showed a strong underestimation of the band gaps. Here we present *GW* calculations that overcome this problem and yield band gaps very close to the best experimental estimates. Finally, we also show a first application of our code to metallic systems. Financial support from the Deutsche Forschungsgemeinschaft through the Priority Programme 1145 is gratefully acknowledged.

Spin excitations in solids from first principles

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Abstract

The long-range order of the electron spins in magnetic solids gives rise to new types of excitations that preserve the particle number but change the total spin of the electron system by $\pm 1\hbar$. While Stoner excitations, which correspond to spin-flip transitions between the majority and minority spin channels, can be well described within a single-particle picture, spin waves are collective modes that result from the Coulomb attraction between excited electrons and holes. The latter are of particular interest, because they dominate the thermodynamic properties of magnetic solids at low temperature due to their small excitation energies. In my talk I discuss different approaches that we have explored to calculate material-specific spin-wave spectra from first principles. All are based on the dynamic transverse spin susceptibility, whose spectral function can be directly related to experimental spectroscopies, but employ either time-dependent density-functional theory or many-body perturbation theory to treat electronic exchange and correlation. The implementation uses the full-potential linearized augmented-plane-wave method, which is well suited to describe the localized d and f orbitals in which the magnetism originates. For ferromagnetic transition metals like Fe, Co or Ni our results are in good agreement with experimental measurements and reproduce all important features.

Non-equilibrium dynamics in strongly correlated clusters

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Abstract

We study the effect of a time dependent external field on small, strongly correlated clusters, described by a Hubbard hamiltonian, by propagating the Kadanoff-Baym equations within the GW and the T-matrix approximations. To obtain the initial conditions for the time propagation, we calculate the self consistent GW and T-matrix groundstates. From the time dependent densities we obtain the corresponding exchange correlation potentials through reverse engineering. The GW and T-matrix based results are then compared to those of the exact solution.

Band offsets prediction within many-body perturbation theory

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Abstract

Standard DFT calculations fails to give an estimation of band offsets in a quantitative agreement with existing experimental data. This makes it important to go beyond framework of DFT toward more accurate methods for band gap calculations. We used the many-body perturbation theory, to examine the capability of the quasiparticle corrections to predict the band offsets for silicon interfaces with oxides and silicates . Our analysis for a well known system, namely SiO_2/Si, has shown that (i) the separate treatment of band-structure and potential lineup contributions, the latter being evaluated within density-functional theory, is justified, (ii) most plasmon-pole models lead to inaccuracies in the absolute quasiparticle corrections, (iii) vertex corrections can be neglected, (iv) eigenenergy self-consistency is adequate. Our theoretical offsets agree with the experimental ones within 0.3 eV.

Binding energy of adsorbates on a noble-metal surface: exchange and correlation effects

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Abstract

The binding of atoms and molecules to noble-metal surfaces is often controlled by weak physisorption, mainly of van der Waals type. Prototypical systems include both rare-gas atoms (e.g. xenon) and aromatic molecules like PTCDA (relevant for coating and contacting) on silver. A theory of physisorption must account for non-local correlation effects. Present implementations of density-functional theory fail here since the common functionals focus on short-range exchange-correlation effects. Here we demonstrate that physisorption can be correctly described with a combination of exact exchange and correlation energy, which we obtain from the adiabatic-connection fluctuation-dissipation theorem. At large distance the correlation energy causes van der Waals attraction. At closer distance the attraction, in combination with the repulsive exchange energy, yields an equilibrium of xenon and of PTCDA on the Ag(111) surface in close agreement with experiment.

Posters

Electron-phonon coupling in graphite within the GW-approximation

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Abstract

A seemingly trivial thing such as the phonon dispersion of graphite continues to present surprises. For most materials (both metals and insulators), density-functional perturbation theory using LDA or GGA gives phonon dispersions in close agreement with experimental results. For graphene and graphite, however, there is a pronounced failure of LDA and GGA for the highest optical phonon branch (HOB) around the Kohn-anomaly at the high-symmetry point K. We evaluate the electron-phonon coupling (EPC) between the pi-bands and the HOB in the GW-approximation. Non-local exchange-correlation effects renormalize the square EPC by almost 80 %. This almost doubles the slope of the HOB around K and explains recent experimental results on graphite-phonons using inelastic x-ray scattering and double-resonance Raman spectroscopy. The short-coming of LDA and GGA may be partially fixed by the use of hybrid-functionals such as B3LYP (which partially contains exact Hartree-Fock exchange). The use of pure Hartree-Fock, however, leads to an extremely strong EPC and consequently to an instability of graphene under distortion along the HOB phonon.

Structural and optical transitions of Biliverdin

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Abstract

Phytochromes constitute a widespread family of photoreceptors found in plants and bacteria, where they act as photomorphogenesis regulators. It is now well-established that they exists in two forms: the P_r, physiologically inactive and absorbing red light, and the P_{fr}, absorbing in the far-red domain. It has also been shown that the P_r-P_{fr} transition mechanism involves an isomerisation process. Yet the actual transition path is still an open question, since it has only been possible to directly observe the P_r form so far, and several models are still being debated.

Biliverdin is the phytochrome found in bacteria, and has been recently observed as a crystal in its P_r form within the surrounding protein. Its P_r-P_{fr} transition has been studied both experimentally and theoretically. The most involved region of the molecule around the chromophore has been identified, though no definitive answer has been given for the final geometry. The protonation state of the P_{fr} form is not clear either, and the mechanisms of the possible proton transfers occuring are still unknown. Adding to the confusion, recent studies suggest that the transition might involve a rotation around a single bond in addition to isomerisation. Due to steric clashes, the rotation would in turn cause further conformational changes in the protein environment. Until now this process has been eluded because of its complexity.

All these considerations clearly call for a finer-grained theoretical insight, that we aim at providing the experimentalists with. The chromophore itself being composed of a few dozens of atoms, we are able to study it by means of *ab initio* DFT calculations. When adding the protein environment, the number of atoms immediately goes above 1500, requiring a different level of approximation, and we are exploring several available frameworks. In all cases we are evaluating and comparing the possible protonation states, in addition to systematically study the aforementioned single-bond rotation. In order to compare our results with experiments, we calculate the infrared, Raman, and optical spectra of all the structures we investigate.

Not only will the knowledge of the P_{fr} form give the correct mechanism of the photochemical transition, as it will open the way to the description of the reverse transition (called *dark reversion*), which is orders of magnitude slower and follows obviously a different path. It will then become possible to understand how the metabolic state of the cell, in particular oxygen levels, influence both transitions.

This work is carried out in contact with the experimentalists at the Freie Universität Berlin, Pr. R. Bittl, Dr. M. Brecht, and J. Nieder. We gratefully acknowledge support from Nanoquanta/ETSF, UPV/EHU (Arina & Urano) and BSC (Mare Nostrum).

First principle calculation of optical rotation and electronic circular dichroism spectra: a real-time real-space implementation.

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Abstract

Electronic circular dichroism (CD) is a powerful optical technique to determine the absolute configuration of chiral molecules. Moreover, it can be used to help determine the structure of macromolecules (including the secondary structure of proteins and the handedness of DNA). CD is routinely employed as a post-synthesis characterization technique and several measured data still call for interpretation. We describe a new implementation that permits calculations of cicular dichroism spectra in the TDDFT framework, as implemented in the real-time real-space code Octopus [1]. The method automatically satisfies sum rules, as well as the Kramers-Kronig relations between ciruclar dichroism and optical rotatory power. The problems of the gauge invariance in pseudopotential calculations and of the effect of the egg-box error are discussed, and we show that the actual implementation can provide gauge-free results. An application to bio-molecules for which experimental results are available is presented. Although quantitative agreement between measured and computed spectra is beyond the current level of theory, the theoretical spectra can shed light on some open problems.

[1]Castro, A.: Appel, H; Oliveira, M; Rozzi, C.A.; Andrade, X; Lorenzen, F.; Marques, M.A.L.; Gross, E.K.U.; and Rubio, A ; Phys. Stat. Sol. B, 2006, **243**, 2465 available at www.tddft.org/programs/octopus.

Optical Excitation of the F center in Calcium Fluoride within Many-body Perturbation Theory

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Abstract

As a prototype for defects in insulators, we discuss the optical properties of the F center in calcium fluoride (CaF_2), which constitutes a prominent defect of the material. The F center of CaF_2 exhibits a defect state deep in the band gap. Its excitation by light (at 3.3 eV excitation energy) is described by *ab-initio* many-body perturbation theory (GW approximation and Bethe-Salpeter equation), including electronic exchange, correlation, and electron-hole interaction effects. The excitation can be regarded as a $1s \rightarrow 2p$ transition. The exciton of the absorption band is strongly localized around the vacancy (within about 2 Å for the hole and 4 Å for the electron), but they induce prominent lattice distrotion around the defect. Constrained density-functional theory can predict reliable geometric relaxation for the localized excitons in ionic insulators. The excitation causes strong relaxation of the defect geometry, leading to significant broadening of the optical spectrum by 0.5 eV and to an large Stokes shift of 1.5 eV.

Excited state properties of water in its various phases: vapour, liquid and solid

Viviana Garbuio, Michele Cascella, Rodolfo Del Sole, Olivia Pulci

ETSF and Universitá di Tor Vergata and CNR-INFM, Roma

Abstract

In the last years, great effort has been devoted to the study of the excited state properties of water in its different phases. Here we present ab-initio calculations, both within DFT and TDDFT and in the framework of many body Green's function formalism, of the water monomer, of liquid water and of cubic ice. For the liquid system, we use snapshots taken from classical molecular dynamics as input geometries for the study of the electronic and optical spectra and we carry out an accurate analysis of the system size effects. The cubic ice excited state properties are calculated by exploiting a model cell without considering the proton disorder. The resulting spectra are strongly modified by passing from the gas phase to the condensed ones and differences are also shown between liquid and solid water.

The calculation of vibrational states of triatomic molecules using an algebraic approach

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Abstract

An accurate and efficient algebraic method for the calculation of vibrational spectra of three-atomic molecules is presented. The method is based on the factorization approach and a known potential energy. The Hamiltonian is expressed as a function of two bond lengths (r_1 and r_2) and the internal angle (θ). The two stretching parts of potential depended on r_1 and r_2 are fitted on a sum of powers of the Morse function, which is different for each oscillator. The bending part depending on θ is fitted on a sum of powers of the *cos* function. The basis functions are products of generalized quasi number states basis set (gQNSB) [A. Bordoni, N. Manini, J. Phys. Chem. A **111** 12564 (2007)], (for the stretching modes which is based on Morse oscillator wavefunctions), and spherical harmonics (for the bending vibrations). Due to the properties of spherical harmonics the singularity in the kinetic term of the Hamiltonian for linear molecules is canceled. The matrix element of the Hamiltonian in this basis can be computed exactly by algebraic techniques [A. Bordoni, N. Manini, I. J. Quant. Chem. **107** 782 (2007). , G. Arfken, "Legendre Functions." Mathematical Methods for Physicists, Academic Press, 1985.]. As a testing system is taken the D.Q. Xie potential energy function [D.Q. Xie, Y.H. Lu, D.G. Xu, *et al.*, Chem. Phys. **270** 415 (2001).] of the linear triatomic molecule carbonyl sulfide. The calculated energy levels are in very good agreement with experimental data (difference $< 0.5\text{cm}^{-1}$).

Ab-initio study of the optical properties of flavin mononucleotide (FMN) in gas phase and in protein environment.

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Abstract

Flavins play a key role in metabolism, and in DNA repair mechanisms. In these processes the ground states and the excited states are involved, respectively. The optical properties of flavin mononucleotide (FMN) in gas phase, in the oxidized, reduced and anionic form, are here studied by ab-initio methods. The optical spectra were obtained within the density functional theory (DFT) and within the time dependent density functional theory (TDDFT). At the DFT level we verified that only the cromophore is optically active, at least within the range of energy of our interest. At TDDFT level the optical spectra show a qualitative good agreement with the experimental ones. An analysis of states, involved in the low energy structure of the spectra, was carried out by Casida linear response. We also performed a QM/MM study on oxidized FMN as cofactor of the flavodoxin, a bacterial protein, in order to explore the effect of the biological protein environment on FMN.

Theoretical studies of quasiparticle, excitonic, and dielectric properties of InN

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Abstract

For a long time not only experiment but also theory was unable to provide the correct value of the fundamental gap of InN and to describe related properties as e.g. the dielectric function. For most standard semiconductors, e.g. silicon, standard tools like density functional theory (DFT) in the local density approximation (LDA) or the generalized gradient approximation (GGA) for the exchange and correlation (XC) interaction work quite well, apart from the well-known gap problem of DFT. Major improvements can be achieved by adding up on top of a DFT calculation many-body perturbation theory, e.g. using Hedin's GW approximation for improved single-particle band structures or solving the Bethe-Salpeter equation (BSE) to describe excitonic effects. However, for InN the situation is extremely complicated. The anomalously small gap of InN, which is to a large extent due to an exceptionally large p-d repulsion pushing the p-like valence band maximum (VBM) towards the s-like conduction band minimum (CBM), paired with the DFT gap problem results in a complete disaster. One obtains a completely wrong band ordering and a vanishing gap for both InN polytypes, zinc-blende and wurtzite. Since standard many-body perturbation theory tools rely on a reasonable starting point (correct band ordering, reasonable dielectric function in order to obtain a reasonable screened Coulomb interaction W and a reasonable Green's function G entering GW and BSE calculations) the wrong InN band structure in standard DFT has the serious consequence that the commonly used non-self-consistent GW approach and solutions of the BSE also suffer serious inaccuracies and can in fact not be applied to InN as usual (starting from DFT). Hence one needs to improve the theoretical framework. One possibility would be tricks like first freezing the In 4d into the core (removing the p-d repulsion and opening a gap), then putting GW (or BSE) on top and finally restoring the p-d repulsion ("three-step-procedure"). However such tricks, although working surprisingly well, cannot be the final answer. Fortunately, there exist approaches beyond LDA and GGA that were mainly inspired by quantum chemistry and have in common that they employ in a certain way a fraction or a screened version of exact exchange to describe the XC interaction. Such "generalized Kohn-Sham schemes" employing these hybrid functionals give highly improved Kohn-Sham band structures from which one can start reliable GW and BSE calculations. The main reason why these schemes perform so well is that they resemble already in a crude way the structure of the GW quasiparticle equation. Among the many proposed hybrid functionals we have finally used the HES03 approach (after demonstrating that the different starting points arising from different functionals give almost identical GW results). Based on HSE03+GW we will provide a detailed discussion of the band structure of InN (band gap, position of the In 4d, effective masses, densities of states, crystal-field splitting in the wurtzite case, spin-orbit splitting). Within the framework of the BSE (although not based on HSE03 but the simpler three-step procedure mentioned above) we discuss excitonic optical spectra and the exciton binding energy. We discuss the overall optical spectra as well as the onset behaviour. We will demonstrate that a simple model Hamiltonian as proposed by Kane can provide valuable insights and that full ab initio results are not only consistent with simple models but also with available recent experimental data achieved for high-quality samples.

Electrons in the conduction bands of TCOs

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Abstract

Recently, devices like liquid crystal displays, OLEDs (organic light emitting diodes), or solar cells have become more and more important especially for mobile applications. One important aspect in this context is the availability and controllability of transparent and conductive materials. Being possible candidates with the desired properties TCOs (transparent conducting oxides) are subject to intensive experimental and theoretical research. In this ab initio study of three semiconducting oxides – ZnO, TiO2, and In2O3 – we adress the influence of electrons in the conduction bands. While they are responsible for the conductivity they ideally should not change any of the other properties like the transparency and the optical properties. To study this, we perform DFT-GGA calculations to obtain Kohn-Sham wavefunctions and energy eigenvalues. The all-electron PAW wavefunctions allow the computation of optical transition matrix elements. The Pauli blockade and the occurence of inter-conduction band transitions are studied on the RPA level and within the independent quasiparticle approximation. We explicitly show that occupied conduction bands lead to a shift of the absorption onset to higher energies (Burstein-Moss shifts). A second effect are additional contributions to the absorption caused by conduction-conduction band transitions. They can be found at energies lower than the lowest valence-conduction band transitions.

Towards a full understanding of "CO adsorption puzzle": many-body electronic structure of CO/Cu(111)

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Abstract

Density functional theory (DFT), within present-day local/semi-local approximations (LDA/GGA), has well-known problems for describing certain surface chemical processes. For instance, when a CO molecule is adsorbed at close-packed Cu(111) and Pt(111) surfaces, both LDA and GGA predict the wrong adsorption site, favoring the threefold-coordinated hollow site rather than the onefold-coordinated top one [1]. Moreover, recent investigations indicated that even the performance of hybrid functionals is not satisfactory [2]. This status calls for more accurate many-body treatment of the problem. In this work, we focus on the CO adsorption at Cu(111) surface, and model the surface with properly selected Cu clusters. Electronic structures for the clusters are obtained by performing PBE0+G_0W_0 calculations, and compared with those obtained from GGA and PBE0 hybrid functional calculations. The adsorption energies are also calculated at the level of random phase approximation (RPA) within the formalism based on the adiabatic connection fluctuation-dissipation theorem (ACFDT). This is done in particular along the line of a 'local XC correction scheme' proposed recently [3] such that the converging behavior can be achieved at relatively small cluster size. In this way new aspects of the physics are revealed that are missing in DFT studies.

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Second order harmonic generation in crystalline semiconductors

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Abstract

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. In principle the optical response of a material depends strongly on local field effects and electron correlations and important advances have been made recently to account for these in the linear response using Time Dependent Density Functional Theory (TDDFT) [1]. We formulate a derivation for the calculation of the second-order susceptibility tensor for crystals of any symmetry within TDDFT and show how this approach can be simplified and expressed in terms of the second order response function and of the dielectric functions. Numerical results will be presented for several materials with zincblende and hexagonal structure as well as preliminary results on $KTiOPO_4$, which is a crystal frequently used for frequency doubling in optical laboratories.

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Nickel oxide: spin and exchange effects

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Abstract

The issue at the origin of this work is the nature underlying the considerable gap (4.3 eV) of nickel oxide. The question is whether the spin or the exchange interaction could contribute to its formation, or if maybe both could explain it. NiO is historically the prototype of the so-called "strongly correlated materials". The problem of its correct theoretical description was raised by N.F. Mott, since the paramagnetic phase of NiO was badly described by "band theories". He stated that strong correlation (strong d-electrons localization and repulsion) causes gap opening (Hubbard insulator) and prevents a band structure description (independent electrons approximation). The DFT-LDA calculations show us that spin leads to an insulating Kohn-Sham structure, whereas without it the system is in a metallic configuration. This is a well known result in literature. When a HF calculation is performed without accounting for the spin, NiO is still predicted to be a metal, as the DOS is non-zero at the Fermi level. Nevertheless, the density of states is heavily modified and loses most of its weight around the Fermi level. Introducing the spin in LSDA produces an insulating structure with an energy gap of about 0.5 eV, and the subsequent Hartree-Fock calculation predicts an insulator as well, with a high energy gap of about 14 eV (Hartree-Fock is known to highly overestimate the gap).

Quantum Transport Through Gold Wires: Ab Initio Studies Using Plane Waves and Supercells

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Abstract

We present a numerical scheme for an *ab initio* implementation of the Landauer-Büttiker theory for quantum transport by means of supercells and plane-wave basis sets. The suggested method works entirely in **k** space which allows to circumvent the complicated projections onto tight-binding Hamiltonians necessary in the more common real-space approaches to quantum transport. Here we apply this method to DFT calculations of transmission functions for quasi-1D Au nanowires of various lengths and widths. The results are discussed with respect to the possible replacement of semi-infinite electrodes by short Au nanowires in future calculations of metal-molecule-metal junctions.

Optical Properties of Ge-Si Alloy Nanocrystals

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Abstract

The optical properties of Si and Ge nanocrystals are important in view of potential opto-electronic applications. The strong differences in the intrinsic radiative lifetimes [1] of nanocrystals in the two materials have been explained with reference to the bulk band structures of the two materials [2]. While Ge nanocrystals have been found to show very short lifetimes, Si nanocrystals remains essentially indirect. The differences in the radiative lifetimes have been confirmed by calculations in TDLDA [3]. On the other hand, the two materials are miscible at any composition. This raised the question as to how the mixture of the two materials behaves. In the independent-particle approximation, the effects of alloying and confinement have been found to be independent [4]. In the present work we present calculations in TDDFT-ALDA of the optical properties of GeSi alloy nanocrystals. It is found that the change of the first absorption peak with changing composition is very similar to the pair excitation energies calculated within the Delta-SCF method [1]. The full inclusion of the Coulomb term which is responsible for the (crystal) local-field effects is found to change the spectra strongly. However, the low-energy spectra behave very similar with changing composition. An analysis of the spectra using the Casida formalism shows that in the ALDA, the transitions at the Ge side of the spectra are well represented by single transitions between Kohn-Sham states, while starting from about 30% of Si, a strong mixing of transitions is found.

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Quasiparticle-calculations of TCOs

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Abstract

The band structure of transparent conducting oxides is at the heart of fundamental electro-optic and transport properties whose systematic prediction would open new avenues in different areas of the materials industry, from electronics to the manufacture of coatings. The perfect crystalline basis oxides ZnO (wurtzite), CdO (rock-salt), In2O3 (bixbyite), SnO2 (rutile), SnO (litharge) are degenerate wide-gap polar semiconductors with challenging electronic properties as well as transport properties which are characterised by important electron-phonon interaction effects (indirect gap conduction as in In2O3; importance of polarons in ZnO) and strongly bound Wannier excitons (ie, 60 meV in ZnO). The combination of transparency and conduction in TCOs requires the formation of point defects in the crystal lattices, either intrinsic under thermal equilibrium conditions or extrinsic after doping. Most TCOs are n-type, and the conventional mechanism responsible for the combination of material properties is usually described by the existence of (a) a large fundamental band-gap which ensures transparency, (b) shallow donor defect-gap states very close in energy to the bottom of the conduction band, (c) a valence band whose states at the top have a large effective mass, (d) a conduction band whose states at the bottom have a small effective mass and are centred at Gamma. Furthermore, a second band-gap above the conduction band seems to be an important ingredient common to all TCOs. A comprehensive band-bap engineering research project for TCOs has been devised in collaboration with several experimental groups and industrial organisations in order to make theoretical predictions dicrectly measureable, and regarding the interplay of structure, electronic properties and conductivity/optical properties of thin-film TCOs. With a view on contributing towards new material properties, for instance for the development of new p-type TCOs, we have a long-term interest in structural work using a combination of ab-initio, global optimisation (genetic algorithm) and cluster expansion techniques. The workhorse of our current theoretical work is LDA and the GW approximation for studying the electronic band structure of TCOs. Those are usually described as weakly correlated, and hence, predictions of the fundamental band gap, effective masses of valence and conduction bands, quasiparticle energy of the defect-gap localised states, as well as the second band-gap should be accessible from Many-Body Perturbation Theory. ABINIT is the main first-principles package which is being applied for our computations. At a later stage, the possibility of strong dynamic correlations [1] involved in the conductivity of TCOs are to be investigated with LDA+Dynamic-Mean Field Theory. LDA+GWA has serious limitations as evidenced by important cases like the fundamental band-gap of ZnO [2,3], ie, the underestimate of the band gap and the presence of underbound Zn d-bands found in the LDA [4] seem to be present in GWA. Since a pre-requisite for accurate band-structure prediction of the point defect systems is the accurate computation of fundamental band- gaps in the perfect crystal lattice of the parent oxide, our calculations are being complemented by other methods. On the one hand, LDA+U previous to GWA quasiparticle-calculations using ABINIT is being applied to the basis oxides. On the other, the B3LYP hybrid functional with the CPMD implementation is being tried on those.

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The Antiferromagnetic Transition Metal Oxides MnO, FeO, CoO, and NiO: Electronic and Optical Properties

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Abstract

Although the electronic properties of MnO, FeO, CoO, and NiO have been studied for decades, the treatment of the antiferromagnetic transition metal oxides within many-body perturbation theory (MBPT) remains a challenge. It is well-known for years that density-functional theory (DFT) in the local-density (LDA) and generalized-gradient approximation (GGA) yields by far too small or even negative gaps for this class of materials. Consequently, quasiparticle calculations using Hedin's GW approximation in the commonly utilized one-shot approach G_0W_0 on top of a GGA calculation underestimate the fundamental gap significantly or cannot even be applied. For this reason we use the non-local HSE03 exchange-correlation functional to obtain a reasonable starting point for a quasiparticle calculation within first-order perturbation theory and compare to results obtained within the computationally less expensive GGA+U scheme. The respective value for the on-site interaction U is determined by a fit to the more sophisticated HSE03+G_0W_0 band structure. Furthermore, we examine the optical properties of these materials. Accounting for their antiferromagnetic ordering, we solve the spin-polarized Bethe-Salpeter equation (BSE) and calculate optical absorption spectra including excitonic and local-field effects. In order to keep the computational demand treatable, we utilize the GGA+U approach for the subjacent electronic structure calculation.

ABINIT + Wannier90: a powerful tool for a wide variety of problems

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Abstract

Motivated by the wide range of applications of maximally-localized Wannier functions (MLWFs), we have implemented an interface to Wannier90 [1] in ABINIT [2]. The general methodology is to use ABINIT to obtain a set of Bloch orbitals $|\psi_{n\mathbf{k}}\rangle$ after a ground state calculation and then to use Wannier90 to get a set of Maximally-Localized Wannier functions (MLWFs). Due to the fact that MLWFs are localized in real space and that they represent a complete basis set, they are useful in large scale calculations and in dielectric response calculations among other applications. Here we show two specific applications, in particular, quantum transport calculations using a Landauer approach and GW band-structure interpolation.

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Influence of the structure on electronic properties of V_2O_3

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Abstract

Aim of our work is to study through ab initio theoretical approach the paramagnetic insulting phase of V_2O_3 compound, in order to understand better the metal-insulator phase transition subdues by V_2O_3 [1,3]. It has been shown that at DFT level it's not possible to describe properly the insulating paramagnetic phase because of a poor treatment of the strong electronic correlation for such systems with d- or f-incomplete shell. Moreover the comparison of DFT density of state with the experimental one is not possible until including many body effect such the ones arising from a direct photoemission spectroscopy. Within a traditional ab initio density functional scheme, the Kohn-Sham eigenvalues can' t be used for an appropriate description of the electronic bands structures, leading in particulaer for such a system (VO_2, V_2O_3) [2] to an underestimation or even to a closure of the photoemission band gaps [4].

Moreover, from the experimental point of view it has bee shown that adding small amount of Cr into the V_2O_3 a sharp metal-insulator transition arises as a function of both Cr concentration and pression at room temperature. Since to the best of our knowledge all the theoretical calculation present in the literature concern only V_2O_3 without really taking into account the Cr dopant atom cite1, we decide to perform ab initio calculation by considering Cr-doped V2O3 supercell system in order to point out which role the presence of the dopant plays on the electronic structure during the metal-insulator paramagnetic transition of the V_2O_3 .

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Ab initio energy loss spectra of Si and Ge nanowires

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Abstract

We report on an *ab initio* investigation of the energy-loss probability of fast electrons in silicon and germanium nanowires. In agreement with recent experiments and as suggested by classical theoretical models, the energy loss spectra are characterized by a strong enhancement of the direct interband transitions peak at low energy. Our calculations predict, for very thin wires, an important diameter dependence of the bulk volume plasmon peak, which is consistent with the blue shift observed experimentally in bigger wires.

Electronic excitations at finite momentum transfer and short-range order changes in covalent materials - a joint theoretical and experimental approach

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Abstract

Time-dependent-density-functional theory (TDDFT) within the adiabatic local density approximation (ALDA) is successfully employed to calculate the dynamical structure factor $S(\mathbf{q}, \omega)$ nowadays. Good agreement with experimental data obtained from inelastic x-ray scattering have been reported for different materials such as silicon , aluminum or transition metal oxides.

In this work we explore the potential of TDLDA calculations of $S(\mathbf{q}, \omega)$ to detect changes in the local geometry of a material. For this purpose we performed TDLDA calculations for amorphous and crystalline GeTe. This covalently bonded material has been reported to exhibit a significant change in the local geometry upon amorphisation. For $\mathbf{q} = 0$ it has been shown that the change in local geometry results in a decrease of the matrix elements of the optical transitions upon amorphisation. However, our calculations show that for finite momentum transfer $\mathbf{q}$ the differences in the spectra of $\epsilon_2(\mathbf{q}, \omega)$ as well as of $S(\mathbf{q}, \omega)$ vanish. Inelastic x-ray scattering experiments confirm these calculations. Here we present an explanation for these unexpected results.

Attempt to Spatially Resolved Electron Energy-Loss Spectroscopy from First Principles

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Abstract

The recent progress in electron microscopy to achieve atomic resolution in electron energy-loss experiments (EELS) opens a new way to investigate nanostructures. Within the framework of time-dependent density functional theory (TDDFT), first principles calculations[1] have been successfully applied to study momentum resolved electron energy-loss spectra (EELS) of solids and molecules. We try to extend the current approach to spatially resolved EELS in the low loss region (less than 40 eV) and face several questions: Can we neglect relativistic effects ? To which extend is the longitudinal dielectric function appropriate to describe the energy-loss ? What about the non-local response (depolarization effects) ? And which quantum effects will be included ? [1]

[1] www.dp-code.org

Pressure effects on the superconducting transition in *nH*-CaAlSi

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Abstract

We present a theoretical study of the effects of pressure on T_c of the hexagonal layered superconductors nH-CaAlSi (n = 1, 5, 6), where nH labels the different stacking variants that have been recently discovered. Experimentally, the pressure dependence of T_c has been investigated by measuring the magnetic susceptibility of single crystals up to 10 kbar. In contrast to previous results on polycrystalline samples, single crystals with different stacking sequences display different pressure dependences of T_c. $1H$-CaAlSi shows a decrease of T_c with pressure, whereas $5H$ and $6H$-CaAlSi exhibit an increase of T_c with pressure. *Ab-initio* calculations for $1H$, $5H$ and $6H$ -CaAlSi reveal that an ultrasoft phonon branch associated to out-of-plane vibrations of the Al-Si layers softens with pressure, leading to a structural instability at high pressures. For $1H$-CaAlSi the softening is not sufficient to cause an increase of T_c, which is consistent with the present experiments, but adverse to previous reports. For $5H$ and $6H$ the softening provides the mechanism to understand the observed increase of T_c with pressure. Calculations for hypothetical $2H$ and $3H$ stacking variants reveal qualitative and quantitative differences.

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

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Abstract

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 STM, LEED and RHEED experimental studies of the surface [1], the structure remains a matter of some controversy, with several c(2x6), (4x3), and c(2x10) reconstructions being proposed for the various observed phases. Total energy calculations favour the (4x3) structures [2], and unlike most other III-V materials the c(4x4) and (2x4) reconstructions are not calculated to be the stable phases.

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]. We present ab initio calculations [4] of RA spectra for various GaSb(001) reconstructions and compare with experiment. We find that agreement is obtained only by mixing contributions of certain structures, and in particular we propose a new structure for the c(2x6) phase. Furthermore, the importance of including spin-orbit coupling in the calculations is investigated, following recent suggestions [5] that they can have considerable effects on RA spectra for compounds containing antimony.

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Optical response of CdSe nanowires

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Abstract

Some experiments and theoretical calculations carried out on CdSe nanowires pointed out some interesting optical properties (giant optic anisotropy, for example) with promising technological applications. These works motivated us to do a more complete theoretical study of the optical response of CdSe wires, in the framework of time-dependent DFT. We analyze the effect of local fields (big anisotropy of this system strongly suggests that local fields have a relevant role in the optical response). Furthermore, we test the effect of more accurate exchange and correlation kernels, such as adiabatic time-dependent local density approximation and long-range exchange-correlation kernels.

Electronic structure and effective masses in strained silicon

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Abstract

Metal-oxide-semiconductor field-effect transistors (MOSFETS) based on strained silicon hold considerable interest for modern microelectronics, because they are compatible with existing manufacturing technology but promise higher carrier mobility and hence faster switching times than conventional devices based on unstrained silicon. Here we quantitatively examine the effect of strain on the electronic structure of silicon, combining density-functional theory within the local-density approximation and the GW approximation for the electronic self-energy. Quasiparticle band structures, deformation potentials and effective masses are obtained for tetragonal, orthorhombic and trigonal distortions of the unit cell, corresponding to biaxial strain in the (100), (110) and (111) planes with full relaxation, respectively. The tetragonal and orthorhombic distortions induce a valley splitting that lifts the sixfold degeneracy of the conduction-band minimum. Furthermore, strain in any direction causes the band structure to warp , and a splitting between light and heavy holes occurs at the top of the valence band. Even for small strain values, these changes in the electronic structure significantly affect the mobility of the charge carriers.

Ab initio many-body calculation of magnetic excitations in 3d transition metals

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Abstract

To study excitation spectra of magnetic materials from first principles we have developed a computational scheme based on many-body perturbation theory. The main quantity of interest is the dynamical transverse spin susceptibility, from which magnetic excitations, including single-particle spin-flip Stoner excitations and collective spin-wave modes as well as their lifetimes, can be obtained. In order to describe spin waves we include appropriate vertex corrections in the form of a multiple-scattering T-matrix, which describes the coupling of electrons and holes with different spin. To reduce the numerical cost for the calculation of the four-point T-matrix we exploit a transformation to maximally localized Wannier functions that takes advantage of the short spatial range of electronic correlation in the partially filled d or f orbitals of magnetic materials. Our implementation is based on the full-potential linearized augmented plane-wave (FLAPW) method. As an illustration, we show spin-wave spectra and dispersions for the elementary ferromagnets Fe, Co and Ni calculated with our scheme. The results are in good agreement with available experimental data.

Excess electron solvation on crystalline ice films

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Abstract

Water, water clusters and ice possess the fascinating ability to bind and solvate excess electrons. In water the excess electron is enclosed during the solvation in a cavity. On small water-clusters the excess electron occupies a diffuse surface state, whereas for larger clusters also interior excess electrons states were reported. Both amorphous and crystalline ice films absorbed on metals bind excess electron transferred by photo-excitation from the metal to the ice surface. On crystalline ice films on Ru(0001) a long-living excess electron states was observed[1], which is trapped at pre-existing sites on the ice surface. Pump-probe experiments showed a continuous energy relaxation of 0.6eV as the trap evolves through conformational substates, on going on an unusually long time scales from femto seconds to minutes. From the experiments alone the nature of the initial trap site and its evolution during the energy relaxation could not be deduced. Our theoretical investigations focus on these issues and the spectroscopical fingerprint of identified excess electron traps. In the first stage we address prototypical trapping sites for the excess electron on (0001) surface of hexagonal ice within the frame work of density functional theory. As the primary traps we identify admolecules, orientational defects and reconstructed orientational defects that have sufficiently low formation energy to explain the experimentally deduced abundance of trapping sites. At the latter the excess electron binds to the dipole moment resulting from the orientation of an H-O towards the vacuum. The excess electron is entirely localized in front of the surface and its tunneling back into the metal is sufficiently suppressed to allow for further enhancement of the screening by thermally activated reorganization of water molecules around the trap. [1] U. Bovensiepen *et al.* unpublished

Optical properties of graphene nanoribbons

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Abstract

The recent discovery of stable, single-layer graphene has focused the attention on novel carbon-based systems, graphene nanoribbons (GNRs) [1,2,3]. The possibility of producing graphene nanostructures with desired shape and dimensions opens exciting opportunities also in view of novel phenomena occurring under light excitation as well as nanoscale optoelectronics applications. We investigate from first principles the optoelectronic properties of nanometer-sized armchair graphene nanoribbons. Our theoretical approach includes both self-energy corrections and excitonic effects through the state-of-the-art GW-BSE formalism, providing full understanding of excited-state properties. We show that many-body effects are essential to correctly describe both energy gaps and optical response. Electron-hole interaction is found to suppress the van Hove singularities -as known for other 1D systems- and introduces strongly bound excitonic peaks. We analyse different geometries and show that strong exciton binding is accompanied by relevant effects of the ribbon family and passivation [4]. Based on simple prototype structures, we discuss the effect of width modulation on the optical response [5] and highlight new strategies to control local excitations in graphene nanostructures.

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Coupling of nonlocal potentials to electromagnetic fields: influence on the induced current

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Abstract

Most of today's ab-initio calculations are based on Density-Functional Theory using nonlocal pseudopotentials. Due to the spatial nonlocality of the hamiltonians, different results can be obtained in the calculation of the response to an external electromagnetic field, when using either the length or the velocity gauge. To solve this problem, one has to properly define the velocity operator **v**, see for instance [1] and the coupling hamiltonian describing the electrons moving in the nonlocal potential and the elecctromagnetic field, [2]. However, in order to fulfill the continuity equation, the current density operator has to be defined in terms of the velocity operator but should also take into account the correct coupling hamiltonian. The expression of the current density will be given in the framework of perturbation theory, to first and second order. Our result will be applied to the case of linear optical response (sumrules) and for second harmonic generation.

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Structural, magnetic and optical properties of Fe, FeS2 and CoS2

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Abstract

The aim of the present work is the study of magnetic and optical properties of iron and cobalt sulfur compounds, which are important for spintronic applications. Here we first report the results of ab-initio calculations of the complex conductivity of BCC iron, in good agreement with experimental data already at the IP-RPA and RPA-LF level [1]. Spin resolved density of states [2] and joint density of states are also presented. Our bandstructure calculation, for the two spin channels [3], has also been extended to include semicore states (3s and 3p), using an optimal pseudopotential generated within the scalar relativistic approximation [3,4]. The 3s level splitting appears to compare well with experimental results [5]. Structural, magnetic and electronic properties of iron and cobalt sulfur are also calculated. We obtain the spin density distribution and the total magnetization of such compounds, the latter being in agreement with experiments. Moreover, the analysis of the density of states allows us to study the very different electronic behaviour of FeS_2 and CoS_2: the former shows a gap for both spin channels (and behaves hence as a semiconductor), while the latter is an half metal [6]. These calculations open the way to explore also more complex compounds based on Fe, Co and S, such as those which are actually used in spintronic applications [7].

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Applications of Molecular Electronics

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Abstract

Plasmon excitations of small single-walled Carbon Nanotubes

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Abstract

The collective electronic excitations in small-diameter single-walled carbon nanotubes are studied for different non-zero momentum-transfer vectors q. For this purpose, we performed ab initio calculations in the framework of TDDFT within the random-phase-approximation (RPA) including the local field effects (LFE). In particular, the evolution of the loss-functions, the importance of the inclusion of the LFE and the plasmons dispersion are discussed. We found some signatures on the optical spectra which can be associated with a specific tube orientation. Our theoretical results suggest a method which could be useful for experimentalist to discern, among the mixed optical response, a particular tube and its orientation from electron energy loss spectra (EELS).

Multiple scattering approach to valence band and resonant photoemission

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Abstract

Valence band photoemission and resonant photoemission are widely used spectroscopic techniques. In particular, energy and angle resolved ultraviolet photoemission is a powerful technique for mapping energy bands of solids, while resonant photoemission has often been exploited to study the local electronic structure and the retainment or not of some atomic features. We have extended the real space multiple scattering approach with the aim of interpreting experimental diffraction patterns and energy spectra obtained from transition metals both in the UV and X-ray regime. The LMTO method has been used for the calculation of potentials in the atomic sphere approximation and the exchange correlation part was calculated in the LDA approximation. For valence band photoemission we show that a dual behaviour between itinerant and well localized electronic states is observed when analyzing the anisotropy of UV diffraction patterns, respectively in the cases the analysis is performed in energy-resolved or energy integrated mode. Resonant photoemission has been studied with applications to Fe and TiO2 and several preliminary encouraging results on the CIS spectra (showing the typical Fano profile), diffraction patterns and angle integrated energy spectra will be presented.

Electronic Properties of Graphene Sheets with Adsorbed Atoms

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Abstract

Thanks to its recent synthesis [1], graphene has attracted a great deal of attention as an interesting prototype to investigate bidimensional physics and nanoelectronics. Two possibilities for confining electrons using graphitic-like structures which have been intensely studied are carbon nanotubes and graphene nanoribbons, both of which are effectively 1D systems. In the former, the mobility of electrons is confined to the direction parallel to its axis, while in the latter, confinement arises both from intrinsic 2D structure and finite width. The adsorption of atoms (or molecules) on top of graphene sheets offers yet another way to confine the electronic gas through patterning of the adsorbed atoms into lines, which would give the sheet electronic properties similar to those of nanoribbons. Using first-principles calculations, Chernozatonskii *et al* [2] have recently proposed that lines of H atoms adsorbed on top of graphene sheets could provide confinement of carriers to a narrow strip centered on these lines. These adsorbed atoms are found to induce a band gap whose value depends on the spacing between the H lines. The adsorption of species other than H would provide yet another way of tailoring the conductivity of graphene, through the different chemical dopings that different atoms would entail. The goal of the present study is to investigate the effect of different chemical species acting as donors or acceptors on the electronic properties of the carbon sheet. Results for lines of H, O, F, Na and Li adsorbed on graphene, in the DFT-LSDA framework as implemented in the SIESTA code, will be presented. From a structural point of view, H (a donor), O and F (acceptors) are found to form straight lines of atoms covalently bonded to the C atoms, slightly tilted toward the centers of the hexagons of the sheets. One particular configuration of these lines, not considered by the authors of Ref. [2], has shown to be energetically more stable and turns out to have a net spin moment, suggesting that these systems could eventually be magnetic. On the other hand, Na and Li (donors) tend to form a zigzag line parallel to the sheet. Both densities of states (DOS) and Bader analysis of the charges on the atoms of the system suggest that charge carrier densities can be quantitatively different depending on the nature of the adsorbed atomic species. Such a new degree of freedom based on the electronic charge transfer will certainly expand the potential applications of graphene in nanoelectronics.

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Notes