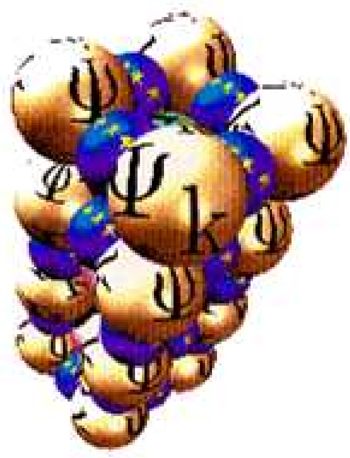
Ψ_k Newsletter

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

Number 88 August 2008



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Sponsored by: UK's CCP9
and ESF Psi-k Programme

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1 Editorial

In this 88th issue of the Psi-k Newsletter, of the 8th month of 2008, we have a number of interesting contributions. One of them is a report on a recent Workshop on "Recent Advances in the KKR Method", which took place on 4-6 July, in the beautiful city of Canterbury, UK. We also have a reminder of the forthcoming UK's CCP9 Conference, covering all aspects of ab-initio and materials specific calculations of the electronic properties of condensed matter systems, which will take place in Cambridge, UK, in the early September. The set of plenary and invited speakers is very impressive, promising a very successful conference. Another announcement is that of the "Meeting on Optical Response in Extended Systems 2008" which will take place in Austria in November. Please check the specific announcements for further details.

As always, this Psi-k Newsletter contains also a few position announcements and abstracts of newly submitted or recent papers.

The scientific highlight of this issue is by Jan M. Tomczak (AIST, Tsukuba) and Silke Biermann (Ecole Polytechnique, Palaiseau) on "Optical Properties of Correlated Materials – or Why Intelligent Windows may look Dirty". It is based on their recent LDA+DMFT results and presents a very interesting reading.

The Psi-k's web page is

http://www.psi-k.org.uk/

From this page you can also access the Psi-k Portal and all the information stored there.

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

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

Dzidka Szotek, Martin Lüders and Walter Temmerman
e-mail: psik-coord@dl.ac.uk

2 General News

2.1 Psi-k Portal

We are pleased to announce that more than 900 members of the old Psi-k mailing list have already joined the Psi-k Portal. Thus, it seems that many people have accepted the transition from the moderated, requiring approvals, mailing list to the self-service provided by the Psi-k Portal. We would like to thank all of you for helping us to make this transition as smooth as possible and encourage you to use it to distribute and receive all the Psi-k related information to the advantage of all involved.

We would also like to ask all the members of the Psi-k Portal to encourage all their co-workers and colleagues to join the Portal if they have not done it yet.

Note that on the main Psi-k web page (http://www.psi-k.org) we have a detailed information on how to join and use the Psi-k Portal. Also, on the Psi-k Portal itself there is available some helpful information. However, in case of any problems we shall be happy to provide further help, if or when we are around.

We welcome all your input and criticism in trying to improve the Psi-k Portal and make it as easy to use as possible.

Thank you for your cooperation.

Psi-k Coordinator

3 Psi-k Activities

”Towards Atomistic Materials Design”

3.1 Reports on Psi-k Workshops

3.1.1 Report on the Canterbury Workshop: ”Recent Advances in the KKR Method”

4-6 July 2008, Canterbury Cathedral Lodge, UK

[The above photograph was taken during the Workshop by Igor Maznichenko (Halle) and shows Malcolm Stocks and Balazs Gyorffy discussing burning issues of the full potential KKR at the entrance to the Conference venue at the Cathedral precinct.]

Sponsored by Psi-k and CCP9

On July 4-6, 2008, an international group of 29 researchers and students met at a weekend workshop to discuss the most recent advances in the KKR method for the electronic structure of complex electron materials. The meeting was held under the auspices of the University of Kent in the picturesque surroundings of the International Study Centre of Canterbury Cathedral which is within the precincts of the Cathedral itself. The workshop was organised within the framework of the Psi-k Programme and UK's CCP9 by Hubert Ebert, Martin Lueders, Paul Strange, Z. (Dzidka) Szotek and Walter Temmerman.

The main goal of the workshop was to bring together all people, from different institutions in Europe, Japan and USA, actively developing, working with or using the KKR method as the main tool for electronic structure calculations. The aim was to discuss and share experiences regarding various implementations of the method (real space, k-space), approximations used (muffin-tin, ASA, ASA-MT corrections and most importantly full-potential), as well as burning issues and problems with various modes of execution. One important point was to establish a set of benchmark systems for all the different KKR codes to fullfil at various modes of implementation, but also review new advances and applications, in particular for complex electron systems.

This two-day workshop was organized around a number of talks, given by the representatives of all the different 'KKR centres', a poster session and round table discussions. The poster session took place on the Saturday afternoon and was started with five-minute presentations by all the contributing authors in which they outlined the main points of their posters. This useful introduction to the posters led to very lively discussions during the session. The whole list of posters is given below.

The first talk of the Workshop was by Martin Lueders (Daresbury), representing the Daresbury-Halle KKR collaboration around the so-called HUTSEPOT code. Martin outlined a number of issues, in particular angular momentum convergence, importance of the Lloyd's formula, stationarity of the CPA, importance of ASA-MT correction in non-full potential implementations, etc. In another talk, Pawel Buczek (Halle) gave more detailed description of the HUTSEPOT code, and its most recent implementation, namely calculating the response functions and spin dynamics. Hubert Ebert (Munich) presented the Munich spin-polarized KKR (SPR-KKR) package, stressing recent developments and applications, and in particular the calculation of the exchange coupling tensor J, using a new calculational scheme which does not require various magnetic reference configurations of the investigated system to determine all its components. Hubert also described a new method of solving the coupled radial Dirac equations in the full potential mode, by making use of the Born series. Rudi Zeller (Juelich) concentrated on the issues underlying the iterative solution of the TBKKR Dyson equation for large systems, while more technical points, like massively parallel implementation, were addressed in a poster by his student Alexander Thiess. Peter Weinberger (Vienna) talked about applications including domain walls and resistivities, considering alloys of Co/Fe, Fe/Ni (permalloy), and Co/Ni. Laszlo Szunyogh (Budapest) also talked about magnetic pattern and domain wall formation in thin Mn and Fe films on W(110) and W(001) as well as on the magnetic configurations of Co and Cr clusters deposited on Au(111), based on the theoretical approach formulated within fully relativistic KKR method and Monte Carlo simulations. Yang Wang (Pittsburgh) and Malcolm Stocks (Oak Ridge) gave a duet presentation in which they explained their innovative technique for solving electrostatic potential in solids, and demonstrated that this technique can be easily implemented within the framework of full-potential multiple scattering theory based electronic structure methods (such as KKR and LSMS). They also discussed some numerical pitfalls they had experienced in self-consistent full-potential LSMS calculations, which were mainly caused by the divergent behavior of the single site irregular solutions near the atomic origins. Julie Staunton (Warwick) talked about how ab-initio modelling of magnetic materials can be accomplished with a theory which includes thermally excited 'local moment' fluctuations alongside relativistic effects such as spin-orbit coupling. This also allows to treat disorder of various types, including short-range order effects, and effect of strong correlations. Julie discussed how key magnetic anisotropies are calculated via a formalism for magnetic torque. Among many things, she discussed the onset of magnetic order in both the heavy rare earths and the 3d transition metal oxides where strong electron correlation effects are important. Hisazumi Akai (Osaka) presented the first-principles calculations of transport properties using the Kubo-Greenwood formula in the framework of the local density approximation of the density functional theory, implemented within KKR. He focussed on the spin injection associated with a DC current in a GMR geometry composed of magnetic materials. Peter Zahn (Halle) discussed spin relaxation as an important issue in spintronics, using KKR as the main theoretical framework. He considered the Elliott-Yafet mechanism of spin relaxation caused by the scattering at point defects. Robert Hammerling (Vienna) talked about the electrostatic problem within a DFT-KKR description, concentrating on solution of the so-called near field correction, which goes beyond a multipole description of the intercell potential. Balazs Gyorffy (Bristol) compared and contrasted two approches to determining the thermodynamic phases of condensed matter from first-principles. One of them in the spirit of 'first-principles' Landau Theory, and the other based on performing ground state calculations for a set of selected symmetries and mapping on to a simple model. In the final talk, Walter Temmerman (Daresbury) discussed some problems of the LMTO that affect also the KKR implementations, most notably the division of space in more complex applications. Walter reviewed some examples of how the results depend on that and finished with stressing a number of points for the future developments in this area. This was in a way a summarizing talk of the Workshop, but also an introduction to a very lively round table discussions. In summary, it was a very successful and useful workshop. One of its outcomes was agreeing to set up a KKR forum for the developers and users of the KKR codes to share their problems and experiences, and to formulate a set of benchmark examples for developers to follow. The KKR formum can be accessed via

http://ccpforge.cse.rl.ac.uk/projects/KKR-forum/

and we encourage all the interested parties to join in to maintain discussions on all aspects of the KKR method, its developments and implementations.

Two future workshops were scheduled for the next year, one more general in Budapest and a technical one, meant mostly for the developers, in Oak Ridge.

Workshop Programme

Friday

14:00 - 14:30	Registration and Welcome
14:30 - 15:00	Martin Lueders *”Overview of the current KKR issues”*
15:00 - 15:30	Hubert Ebert *”The Munich SPR-KKR package - recent developments and applications”*
15:30 - 16:00	Coffee

16:00 - 16:30	Rudi Zeller *”Iterative solution of the TBKKR Dyson equation for large systems”*
16:30 - 17:00	Pawel Buczek *”HUTSEPOT environment & recent developments in multiple scattering theory”*
17:00 - 17:30	P. Weinberger *”Domain wall widths & resistivities & race track memories”*

Saturday

9:30 - 10:00	Laszlo Szunyogh *”Magnetic Interactions from KKR: Spin-Hamiltonian and beyond”*
10:00 - 10:30	Yang Wang *”Full-potential self-consistent KKR and LSMS calculations”*
1S0:30 - 11:00	Malcolm Stocks *”Full-potential self-consistent KKR and LSMS calculations: technical details ”*
11:00 - 11:30	Coffee
11:30 - 12:00	Julie Staunton: *”Ab-initio modelling of the temperature dependence of magnetic properties of crystals, films and nanoclusters”*
12:00 - 12:30	Hisazumi Akai *”Calculation of spin transport using the KKR Kubo-Greenwood formula”*
12:30 - 14:00	Lunch
14:00 - 15:15	Five-minute Poster Presentations
15:15 - 17:00	Posters
17:00 - 17:30	Peter Zahn *”SPIN-FLIP SCATTERING IN METALS: IMPURITIES IN CU”*
17:30 - 18:00	Robert Hammerling *”Near field corrections to electrostatic potential in KKR”*
19:30 - 21:00	Workshop Dinner

Sunday

9:30 - 10:00	Balazs Gyorffy *”First-principles Landau Theory of Phase Transitions: The Disordered Local Fluctuation (DLF) Picture”*

10:00 - 10:30	Walter Temmerman *KKR vs. LMTO: A comparison”*
10:30 - 12:00	Round Table Discussion and Closing

List of Posters

M. Czerner, *”Electronic Structure and Transport under Finite Bias”*

M. Daene, *”Self-Interaction Corrections - Applications to Transition Metal Oxides”*

D. Koedderitzsch, *”The Relativistic Optimized Potential Method for Spin-polarized Atoms and Solids”*

S. Lowitzer, *”Implementation and Application of the Kubo-Greenwood Formalism within the NL-CPA”*

S. Mankovskyy, *”A New Scheme to Calculate the Force Tensor”*

I. Maznichenko, *”Electronic and Magnetic Properties of* $Zr(Mn)O_2$*”*

J. Minar, *”DMFT-KKR Implementation: Total Energies”*

M. Ogura, *”Application of the Full-Potential Screened KKR Method to Metal/Semiconductor Interfaces”*

A. Thiess, *”Massive Parallelization of the KKR Green Function Method”*

B. Ujfalussy, *”Numerical Investigations of Interactions between Magnetic Impurities”*

List of Participants

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The full report of the workshop, including abstracts of the presented talks, will be available on the Psi-k Portal as soon as all abstracts are available.

4 News from UK's CCP9 Programme

UK's Collaborative Computational Project 9 (CCP9) on "Computational Studies of the Electronic Structure of Solids"

4.1 CCP9 Conference Announcement

Extended Deadlines for Registration and Abstract Submission!

Cambridge, 4-5 September 2008

General Information

The Conference will be held at Robinson College in Cambridge on 4th and 5th September 2008. The scope of the Conference is all ab-initio and materials specific calculations of the electronic properties of condensed matter systems such as metals, semiconductors, magnets, superconductors, biological systems, minerals, surfaces etc. Topical areas incorporated in the Conference are magno-electronics, catalysis, nanotechnology, high temperature superconductors, novel wide band gap semiconductors etc. Techniques covered by the Conference include QMC, DFT, GW, TD-DFT, DMFT, SIC-LSDA, LSDA+U. We welcome all contributions dealing with the solution of the Schrodinger/Dirac equation in condensed matter systems.

Plenary Speakers

Hardy Gross (FU Berlin, Germany)
Nicola Marzari (MIT, USA)
David Ceperley (University of Illinois, USA)
Gustavo Scuseria (Rice University, USA)
Vladimir Anisimov (Ekaterinberg, Russia)

Invited Speakers

Hubert Ebert (LMU Muenchen, Germany)
Leon Petit (University of Aarhus, Denmark)
Dario Alfe (UCL, UK)
Neil Drummond (University of Cambridge, UK)
Jonathan Yates (University of Cambridge, UK)

Massimiliano Stengel (Santa Barbara, USA)
Marcus Neumann (AVMATSIM)
Robert Laskowski (TU Wien, Austria)

Important Dates

Submission of Abstracts: by 5th of August 2008

Registration and Fees: paid by 15th of August 2008

Selection of submitted Talks: 15th of August 2008

Conference Dates: 4th and 5th of September 2008

Talks and Posters

We welcome your contributions. Please submit an abstract by 5th of August 2008

Registration & Fees

The online registration for this event is now open, and extended until 15th of August.

Registration Fees are £200 and include two nights accommodation at Robinson College (Wednesday 3rd and Thursday 4th September), all meals and refreshments. Extra accommodation can be booked at the delegates' own expense - please contact Damian Jones (D.R.Jones@dl.ac.uk) to extend your accommodation booking.

A substantial number of Scholarships will be made available from CCP9/Psi-k. These scholarships are especially for UK based young researchers. The Scholarship will take the form of full reimbursement of fees - no support will be provided for travel. If you would like to apply for a Scholarship please tick the relevant box on the online registration form. Successful applicants will be notified by email by 15th of August.

Further up-to-date information can be obtained from

`http://www.ccp9.ac.uk/ccp9workshops_2008_cambridge_sept.shtml`

5 General Workshop/Conference Announcements

5.1 Meeting on Optical Response in Extended Systems 2008

Vienna, Austria

November 19-21, 2008

supported by WIEN KULTUR and the BMWF

organized by the association
Physicae et Chimicae Solidorum Amici

http://www.physics.at; contact: amici@physics.at

”MORE of LESS”. MORE 2008 follows the spirit of the highly successful Low Energy Spectrometry Symposium (LESS) that took place in Vienna in 2007.

Optical absorption spectroscopy, resonant inelastic X-ray scattering, reflection EELS, ellipsometry, and reflectance difference spectroscopy are just a few of a large variety of experimental probes to explore the optical and higher-energy response of materials, including elemental solids, compounds, molecules on surfaces or complex nanostructures.

On the theoretical side many body perturbation theory (MBPT) and time-dependent density functional theory (TDDFT) represent state-of-the-art developments to describe excitation processes in materials. Both, theory and experiment being complementary, ideally should go hand in hand to promote development and leading edge research.

The main intention of MORE 2008 is bringing together experimentalists and theoreticians of these different fields and form a platform for leading experts to discuss and exchange their recent results and scientific concepts. At the same time, tutorial-like lectures given by key experts of these fields will provide high-quality introductions for young researchers. To ensure lively and fruitful discussions and a personal atmosphere, we limit the number of participants including speakers to 50. Contributions in the form of posters are readily welcome.

Invited Speakers (confirmed):

- **C. Ambrosch-Draxl** (MU Leoben, Austria)
- **F. J. Garcia de Abajo** (CSIC Madrid, Spain)
- **R. Gomez-Abal** (FHI Berlin, Germany)
- **R. Hambach** (Ecole Polytechnique, France)

- **M. Kociak** (CNRS-Orsay, France)
- **G. Kresse** (CMS Vienna, Austria)
- **A. Marini** (Univ. of Rome, Italy)
- **L. Reining** (Ecole Polytechnique, France)
- **A. Rubio** (Univ. del Pays Vasco, Spain)
- **P. Schattschneider** (Vienna Univ. of Technology, Austria)
- **W. Sigle** (MPI f. Metallforschung, Stuttgart, Germany)
- **J. Verbeeck** (Univ. of Antwerp, Belgium)
- **W. Werner** (Vienna Univ. of Technology, Austria)
- **P. Zeppenfeld** (JKU Linz, Austria)

Venue:

The MORE 2008 is located at the Erwin Schrödinger International Institute for Mathematical Physics (ESI), Boltzmanngasse 9, 1090 Vienna (www.esi.ac.at).

Registration:

The fee for attending this symposium including tutorial is EUR 195,–.
This fee covers the coffee breaks and the conference dinner.
Registration is now available at http://www.physics.at/.

The final deadline for registration is October 19, 2008.

The payment of the registration fee should be done by **Bank Transfer, without charges for the beneficiary**, to

”Freunde der Festkörperphysik”.
Account Number 304.149.150.06, Volksbank Wien, Bank Code 43.000
IBAN: AT11 4300 0304 1491 5006
SWIFT/BIC: VBWIATW1

Please do not forget to indicate the name of the participant!

Organizers:

C. Ambrosch-Draxl (MU Leoben, Austria)

K. Hummer (Univ. Vienna, Austria)

J. Luitz (Lausanne, Switzerland)

P. Schattschneider (TU Vienna, Austria)

Programme:

**Wednesday,
November 19**

13:30 – 14:00	Registration
14:00 – 14:10	Welcome
14:10 – 16:10	Tutorial 1 (incl. 10min break) **Lucia Reining** *Electronic response - theoretical approaches and link to experiment*
16:10 – 16:30	Coffee break
16:30 – 18:30	Tutorial 2 (incl. 10min break) **Peter Schattschneider** *EELS in the TEM*
18:30 – 19:30	Aperitif

**Thursday,
November 20**

08:30 – 09:00	Registration
09:00 – 09:15	Opening
09:15 – 10:00	**Claudia Ambrosch-Draxl** *The role of electron-phonon interaction in photoemission spectra: an ab-intio study*
10:00 – 10:45	**Andrea Marini** *Breakdown of the electronic picture of the excitonic states: evidence of phonon mediated bright to dark (and vice-versa) transitions?*
10:45 – 11:15	Coffee break
11:15 – 12:00	**Mathieu Kociak** *Nanometer-scale mapping of electromagnetic fields in the visible range*
12:00 – 12:45	**Georg Kresse** *TD-DFT using hybrid functionals: an alternative to GW & BSE?*
12:45 – 14:15	Lunch
14:15 – 15:00	**Ricardo Gomez-Abal** *All-electron GW method based on LAPW: Implementation and applications*
15:00 – 15:45	**Peter Zeppenfeld** *Reflectance Difference Spectroscopy for quantitative surface and thin film analysis*

15:45 – 16:15	Coffee break
16:15 – 17:00	**Wilfried Sigle** *Valence electron spectroscopy using energy-filtered TEM*
17:00 – 17:45	**Wolfgang Werner** *to be announced*
19:00 –	Dinner

Friday,
November 21

09:00 – 09:45	**Angel Rubio** *Modelling excited state dynamics in low dimensional systems*
09:45 – 10:30	**Maria A. Loi** *to be confirmed*
10:30 – 11:00	Coffee break
11:00 – 11:45	**Ralf Hambach** *Electron Energy Loss Spectra of Graphite, Graphene and Carbon Nanotubes: Plasmon Dispersion and Crystal Local Field Effects.*
11:45 – 13:30	Lunch
13:30 – 14:15	**Johan Verbeeck** *Recovering the dielectric function from nanoscale regions with low loss EELS: dream or reality?*
14:15 – 15:00	**Javier Garcia de Abajo** *Plasmons in metallic nanoparticles*
15:00 – 15:30	Closing

6 General Job Announcements

Two Post-doctoral Positions in the Field of Ab Initio Spintronics

Institute of Physics and Chemistry of Materials of Strasbourg (IPCMS), France

Two postdoctoral positions are available immediately at the institute of physics and chemistry of materials of Strasbourg (IPCMS) in the field of ab initio electronic transport in molecular tunnel junctions between ferromagnetic electrodes and molecular magnetism.

We are looking for candidates who have strong experience in the field of ab initio electronic structure and quantum transport. The successful candidates will have to develop an existing numerical code to calculate the tunneling magnetoresistance through molecular junctions using non- equilibrium Green's function formalism. A good knowledge and experience with Fortran programming is therefore essential for the success of this project. The successful candidates will work in the group of Mebarek Alouani (Professor), Olivier Bengone (Assistant Professor), Sébastien Lebégue (CNRS research staff), Carlo Massobrio (CNRS research staff), and are expected also to collaborate with our strong experimental group in the field of spintronics at the nanoscale. Those positions are funded by a grant of the French research council ANR and can be renewed for up to two years. The IPCMS is a joint research center affiliated with the Centre National de Recherche Scientifique (CNRS) and with the University of Strasbourg. It currently employs 77 faculties and research staff.

An application letter together with a résumé of recent research, a list of publications, and the names and emails of at least two referees should be sent by email to Prof. Mebarek Alouani (email: mea@ipcms.u-strasbg.fr).

Research Associate

The Center of Physics of Fundamental Interactions (CFIF) Instituto Superior Tecnico of the Technical University of Lisbon, Portugal

The Center of Physics of Fundamental Interactions (CFIF) of Instituto Superior Tecnico of the Technical University of Lisbon, has one opening for a Research Associate in the area of Theoretical Condensed Matter Physics.

Candidates should have a high quality research record and at least 3 years of post-doctoral research experience in Condensed Matter Physics with emphasis in the field of strongly correlated electronic systems. The candidate is expected to conduct independent research within the interests of the Condensed Matter Group: superconductivity; heavy fermion physics; magnetism and spintronics; non Fermi liquids and quantum phase transitions. The applicant should have a good knowledge of quantum field theory methods and techniques.

The contract offered will have duration of up to 5 years, renewed yearly based on mutual agreement. The application deadline is September 17, 2008.

For more information consult the web page:

`http://www.eracareers.pt/opportunities/index.aspx?task=global&jobId=9830`

under Job reference:C2008-IST/CFIF.2

Instituto Superior Técnico-C2008 Lisboa
Research Field: Physics

For more information on CFIF and IST:

`http://cfif.ist.utl.pt/`
`http://www.ist.utl.pt/en/?language=en`

We would appreciate if you could advertise this position to suitable candidates within your group.

Pedro D. Sacramento
CFIF-IST, TU Lisbon

Post-doctoral Positions

”Electronic simulations of high-temperature superconductors by quantum Monte Carlo”

DEMOCRITOS National Simulation Center, SISSA Trieste, Italy

Post-doctoral positions in ”Electronic simulations of high-temperature superconductors by quantum Monte Carlo”, available at the DEMOCRITOS National Simulation Center, SISSA Trieste, group headed by Prof. Sandro Sorella. Search for outstanding candidates, motivated by the following challenging project: Study of two important high temperature (HTc) superconducting materials, La_2CuO_4 that is the prototype of copper oxide superconductors discovered in 1988, and LaFeAsO, representative of the novel iron based rare-earth oxypnictide superconductors. By means of state-of-the-art quantum Monte Carlo (QMC) techniques, our main target is to understand whether the peculiar magnetic properties of the two materials are related to the pairing mechanism that leads to superconductivity. The superconducting regime in the iron-based and copper-oxide families occurs in close proximity to a long-range ordered antiferromagnetic state. This suggests a strong interplay between magnetism and HTc superconductivity, which is probably one of the most challenging problems in condensed matter physics.

Applications should be sent by e-mail to sorella@sissa.it, with complete CV in attachment.

The deadline for applications is September 24.

7 Abstracts

The magnetization reversal process in spin spring magnets

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Departamento de Física Teórica, Atómica y Óptica,
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Abstract

The behavior of soft magnetic films in spring magnets, as a function of the intensity and orientation of an external magnetic field, is described in the framework of spin-polarized non-collinear electronic structure calculations. As experimentally observed the critical intensity of the external field required for the onset of the non-collinear spiral formation depends on both the thickness of the soft magnetic phase and the orientation of the field. The spin spiral structure undergoes a change of chirality in rotating fields. Our theoretical approach opens new prospects for investigating the response of other nanostructures to external magnetic fields beyond usual phenomenological models.

(Nanotechnology **19**, 315401 (2008))
Contact person: A. Vega (vega@phenix.fam.cie.uva.es)

Possibility of collinear magnetic order in frustrated free-standing Fe_2-Cr_4 clusters

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Abstract

Using the fully unconstrained version of the density-functional method SIESTA with the generalised gradient approximation to exchange and correlation, we compute the structures and magnetic configurations of the lowest-energy isomers of the free-standing cluster Fe_2Cr_4. The two most stable isomers, of distorted octahedral geometry, exhibit magnetic frustration but collinear magnetic order, in contrast to the behaviour of extended Cr/Fe frustrated systems. Confinement effects and structural relaxation are shown to be the origin of the different behavior of Fe-Cr clusters, which illustrates the importance of finite size effects on magnetism.

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Density-functional studies on spin, charge, and orbital ordering in $\mathrm{YBa}T_2\mathrm{O}_5$ (T = Mn, Fe, Co)

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Abstract

Spin, charge, and orbital orderings are influenced by electron/hole doping, cation radii, oxygen stoichiometry, temperature, magnetic field etc. In order to understand the role of electron/hole doping, we have studied the variations in the spin, charge, and orbital ordering in terms of the d-band filling for $\mathrm{YBa}T_2\mathrm{O}_5$ (T = Mn, Fe, Co). The calculations are performed with density-functional theory as implemented in the full-potential linearized-augmented plane-wave method. We have carried out calculations for nonmagnetic, ferromagnetic, and antiferromagnetic configurations. A ferrimagnetic ground state has been established for $\mathrm{YBaMn_2O_5}$ whereas $\mathrm{YBaFe_2O_5}$ and $\mathrm{YBaCo_2O_5}$ have antiferromagnetic ground states in agreement with experimental findings. The effects of spin-orbit coupling, Hubbard U parameter, and orbital polarization on magnetic properties are also analyzed. The electronic band characteristics are analyzed using total as well as site- and orbital-projected density of states. Inclusion of spin-orbit coupling and Coulomb correlation effects in the calculations are found to be important to reproduce the experimentally established semiconducting behavior for $\mathrm{YBaFe_2O_5}$ and $\mathrm{YBaCo_2O_5}$. In order to quantify the charges at each atomic site, we made use of the Bader "atom in molecule" concept and Born-effective-charge (BEC) analyses. The structural optimization and BEC tensor calculations were performed using the VASP-PAW method. The different types of charge and orbital orderings in these compounds are visualized by making use of the energy-projected density matrices of the d electrons. Substantial differences in ordering patterns emerge with respect to the d-band filling. The ordering of the d_{z^2} orbital on Mn atoms in $\mathrm{YBaMn_2O_5}$ gives rise to G-type ferrimagnetic spin ordering along the c direction and a checker-board-type charge ordering whereas an ordering of the $d_{x^2-y^2}$ orbital at Fe in $\mathrm{YBaFe_2O_5}$ causes Wollan-Koehler G-type antiferromagnetic spin order along the b direction and a stripe-type charge order. Similarly, a complex orbital ordering pattern in $\mathrm{YBaCo_2O_5}$ activates spin and charge orderings like in the case of $\mathrm{YBaFe_2O_5}$.

In press on Inorganic Chemistry

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Magnetic-Instability-Induced Giant Magnetoelectric Coupling

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Abstract

Understanding the coupling between magnetism and ferroelectricity is of high fundamental and technological importance. For example, a sufficiently strong coupling between magnetic and dielectric properties may enable switching of the magnetization by means of an electric field, thereby drastically reducing the thermal power needed for a magnetic memory. If electric writing and magnetic reading were to be used, one of the layers in spin valves composed of ferromagnetic and magnetoelectric layers would no longer have to be pinned. Giant magnetoelectric materials facilitate highly efficient elastic interactions and a strong magnetic response to an electric field (or vice versa) via magnetostriction and piezoelectric effects. Several materials possess both ferroelectricity and a co-operative magnetism; however, the coupling between these properties is not necessarily large. The structural and electronic circumstances that would favor a large coupling remain unexplored, partly due to the lack of materials with a large coupling. Here we report that such a large coupling occurs in BiCoO3, for which an external electric field can induce a strong magnetic response by changing the spin-state of cobalt from a magnetic high spin state to a nonmagnetic low spin state.

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Spin-orbital superexchange physics emerging from interacting oxygen molecules in KO_2

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Abstract

We propose that the spin-orbital-lattice coupled phenomena, widely known for the transition-metal oxides, can be realized in molecular solids, comprising of orbitally degenerate magnetic O_2^- ions. KO_2 is one of such systems. Using first-principles electronic structure calculations, we set up an effective spin-orbital superexchange model for the low-energy *molecular* bands and argue that many anomalous properties of KO_2 indeed replicate the status of its orbital system in different temperature regimes.

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Combining DFT and Many-Body Methods to Understand Correlated Materials

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Abstract

The electronic and magnetic properties of many strongly-correlated systems are controlled by a limited number of states, located near the Fermi level and well isolated from the rest of the spectrum. This opens a formal way for combining the methods of first-principles electronic structure calculations, based on the density-functional theory (DFT), with many-body models, formulated in the restricted Hilbert space of states close to the Fermi level. The core of this project is the so-called "realistic modeling" or the construction of the model many-body Hamiltonians entirely from the first principles. Such a construction should be able to go beyond the conventional local-density approximation (LDA), which typically supplements the density-functional theory, and incorporate the physics of Coulomb correlations. It should also provide a transparent physical picture for the low-energy properties of strongly correlated materials. In this review article, we will outline the basic ideas of such a realistic modeling, which consists of the following steps: (i) The construction of the complete Wannier basis set for the low-energy LDA band; (ii) The construction of the one-electron part of the model Hamiltonian in this Wannier basis; (iii) The calculation of the screened Coulomb interactions for the low-energy bands by means of the constrained DFT. The most difficult part of this project is the evaluation of the screening caused by outer bands, which may have the same (e.g., the transition-metal $3d$) character as the low-energy bands. The latter part can be efficiently done by combining the constrained DFT with the random-phase approximation for the screened Coulomb interaction. The entire procedure will be illustrated on the series of examples, including the distorted transition-metal perovskite oxides, the compounds with the inversion symmetry breaking caused by the defects, and the alkali hyperoxide KO_2, which can be regarded as an analog of strongly-correlated systems where the localized electrons reside on the *molecular orbitals* of the O_2^- dimer. In order to illustrate abilities of the realistic modeling, we will also consider solutions of the obtained low-energy models for a number of systems, and argue that it can be used as a powerful tool for the exploration and understanding of properties of strongly correlated materials.

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Orbital Ordering and Magnetic Interactions in BiMnO$_3$

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Abstract

This work is devoted to the analysis of the orbital ordering patterns and associated with them interatomic magnetic interactions in the centrosymmetric monoclinic structures of BiMnO$_3$, which have been recently determined experimentally. First, we set up an effective lattice fermion model for the manganese $3d$ bands and derive parameters of this model from the first-principles electronic structure calculations. Then, we solve this model in terms of the mean-field Hartree-Fock method and derive parameters of interatomic magnetic interactions between Mn ions. We argue that although the nearest-neighbor interactions favors the ferromagnetism, they compete with the longer-range antiferromagnetic (AFM) interactions, the existence of which is directly related to the peculiar geometry of the orbital ordering pattern realized in BiMnO$_3$ below 474 K. These AFM interactions favor an AFM alignment, which breaks the inversion symmetry. The formation of the AFM phase is additionally assisted by the orbital degrees of freedom, which tend to adjust the nearest-neighbor magnetic interactions in the direction, which further stabilizes this phase. We propose that the multiferroelectric behavior, which was observed in BiMnO$_3$, may be related to the emergence of the AFM phase under certain conditions.

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Spin injection from Fe into Si(001): ab initio calculations and role of the Si complex band structure

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Abstract

We study the possibility of spin injection from Fe into Si(001), using the Schottky barrier at the Fe/Si contact as tunneling barrier. Our calculations are based on density-functional theory for the description of the electronic structure and on a Landauer-Buttiker approach for the current. The current-carrying states correspond to the six conduction band minima of Si, which, when projected on the (001) surface Brillouin zone (SBZ), form five conductance hot spots: one at the SBZ center and four symmetric satellites. The satellites yield a current polarization of about 50%, while the SBZ center can, under very low gate voltage, yield up to almost 100%, showing a zero-gate anomaly. This extremely high polarization is traced back to the symmetry mismatch of the minority-spin Fe wavefunctions to the conduction band wavefunctions of Si at the SBZ center. The tunneling current is determined by the complex band structure of Si in the [001] direction, which shows qualitative differences compared to that of direct-gap semiconductors. Depending on the Fermi level position and Schottky barrier thickness, the complex band structure can cause the contribution of the satellites to be orders of magnitude higher or lower than the central contribution. Thus, by appropriate tuning of the interface properties, there is a possibility to cut off the satellite contribution and to reach high injection efficiency. Also, we find that a moderate strain of 0.5% along the [001] direction is sufficient to lift the degeneracy of the pockets so that only states at the zone center can carry current.

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Spin-Orbit Coupling and Ion Displacements in Multiferroic $TbMnO_3$

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Abstract

The magnetic and ferroelectric (FE) properties of $TbMnO_3$ were investigated on the basis of relativistic density functional theory (DFT) calculations. We show that, due to spin-orbit coupling, the spin-spiral plane of $TbMnO_3$ can be either the *bc*- or *ab*-plane, but not the *ac*-plane. As for the mechanism of FE polarization, our work reveals that the "pure electronic" model by Katsura, Nagaosa and Balatsky is inadequate in predicting the absolute direction of FE polarization. Our work indicates that to determine the magnitude and the absolute direction of FE polarization in spin-spiral states, it is crucial to consider the displacements of the ions from their centrosymmetric positions.

(Phys. Rev. Lett. **101**, 037209 (2008))

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Small polarons and magnetic anti-phase boundaries in $Ca_{2-x}Na_xCuO_2Cl_2$ (x=0.06, 0.12): origin of striped phases in cuprates

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Abstract

We report hybrid density functional theory calculations on hole doped $Ca_{2-x}Na_xCuO_2Cl_2$ performed in 4x4, $4\sqrt{2}$x$4\sqrt{2}$ and 8x2 supercells with hole concentrations x = 0.0625 and x = 0.125. Holes at the lower concentration form small polarons in which the hole is mainly localized on four oxygen ions surrounding one copper ion. The polaron is a spin one-half ferromagnetic polaron (Cu_5O_4) in which the moment on the central copper ion is parallel to those on the four neighboring copper ions and the moment on the oxygen ions is opposed to that on the copper ions. This is therefore an Emery-Reiter spin polaron rather than a Zhang-Rice singlet. At the higher hole concentration (x = 0.125) many cuprates form stripes. Hybrid density functional theory calculations on linear chains of spin polarons separated by 4 a_o show a group of bands localized mainly on the stripe. Spins on neighboring copper ions in the stripe are parallel and so the stripe forms a magnetic anti-phase boundary between antiferromagnetically ordered blocks of copper spins. Stripes of this kind, which run in one direction only, may explain recent STM data from $Ca_{2-x}Na_xCuO_2Cl_2$ by Kohsaka *et al.* [Science **315**, 1380 (2007)]. We also consider an ordered spin polaron phase where magnetic anti-phase boundaries intersect at right angles. In this case, sets of four copper ions in squares at stripe intersections have parallel spins. This phase may be the 4x4 checkerboard pattern reported by Hanaguri *et al.* [Nature (London) **430**, 1001 (2004)].

(Phys Rev B **77**, 94523 (2008))
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Self-interaction correction in multiple scattering theory: application to transition metal oxides

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Abstract

We apply to transition metal monoxides the self-interaction corrected (SIC) local spin density (LSD) approximation, implemented locally in the multiple scattering theory within the Korringa-Kohn-Rostoker (KKR) band structure method. The calculated electronic structure and in particular magnetic moments and energy gaps are discussed in reference to the earlier SIC results obtained within the LMTO-ASA band structure method, involving transformations between Bloch and Wannier representations to solve the eigenvalue problem and calculate the SIC charge and potential. Since the KKR can be easily extended to treat disordered alloys, by invoking the coherent potential approximation (CPA), in this paper we compare the CPA approach and supercell calculations to study the electronic structure of NiO with cation vacancies.

(Submitted to Journal of Physics: Condensed Matter)

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Identification of Silica-Supported Vanadia by X-ray Absorption Spectroscopy: combined theoretical and experimental studies

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Abstract

In this study we combine density-functional theory (DFT) calculations on oxygen core excitations in vanadia-silica model clusters with experimental in situ X-ray absorption fine structure (NEXAFS) near the oxygen K edge of vanadia model catalysts supported by SBA-15 silica in order to identify structural details of the vanadia species. The silica support is found to contribute to the NEXAFS spectrum in an energy range well above that of the vanadium oxide units allowing a clear separation between the corresponding contributions. Further, differently coordinated oxygen which is characteristic for particular vanadia species, monomeric or polymeric, can be clearly identified in the theoretical spectra in agreement with the oxygen K-edge NEXAFS measurements. The comparison of the theoretical and experimental NEXAFS spectra provides clear evidence that under in situ conditions different molecular vanadia species, in particular polymeric VOx, exist at the catalyst surface and the exclusive presence of monomeric vanadia groups can be clearly ruled out. The present analysis goes beyond earlier work applying vibrational spectroscopy to the present systems where, as a result of extended vibrational coupling, a separation between vanadia, silica, and interface contributions is less successful.

(Submitted to: J. Catal.)

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Influence of Core-Valence Interaction and Pseudoization on the Electron Self-Energy

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Abstract

State-of-the-art theory addresses single-electron excitations in condensed matter by linking density-functional theory (DFT) with many-body perturbation theory. In actual calculations it is common to employ the pseudo-potential (PP) approach, where the pseudoized wave functions enter the calculation of the self-energy, and the core-valence interaction is treated at the DFT level. In this Letter we present accurate all-electron calculations of the self-energy and systematically compare the results to those of PP calculations. The analysis for a range of different materials reveals that both above mentioned approximations are indeed problematic.

(Accepted for publication in: Phys. Rev. Lett.)

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Examination of the concept of degree of rate control by first-principles kinetic Monte Carlo simulations

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Abstract

The conceptual idea of degree of rate control (DRC) approaches is to identify the "rate limiting step" in a complex reaction network by evaluating how the overall rate of product formation changes when a small change is made in one of the kinetic parameters. We examine two definitions of this concept by applying it to first-principles kinetic Monte Carlo simulations of the CO oxidation at RuO2(110). Instead of studying experimental data we examine simulations, because in them we know the surface structure, reaction mechanism, the rate constants, the coverage of the surface and the turn-over frequency at steady state. We can test whether the insights provided by the DRC are in agreement with the results of the simulations thus avoiding the uncertainties inherent in a comparison with experiment. We find that the information provided by using the DRC is non-trivial: It could not have been obtained from the knowledge of the reaction mechanism and of the magnitude of the rate constants alone. For the simulations the DRC provides furthermore guidance as to which aspects of the reaction mechanism should be treated accurately and which can be studied by less accurate and more efficient methods. We therefore conclude that a sensitivity analysis based on the DRC is a useful tool for understanding the propagation of errors from the electronic structure calculations to the statistical simulations in first-principles kinetic Monte Carlo simulations.

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(Submitted to: Surf. Sci.: Special Ertl issue)

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Ab Initio Molecular Simulations with Numeric Atom-Centered Orbitals: FHI-aims

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Abstract

We describe the Fritz Haber Institute "*ab initio* molecular simulations" (`FHI-aims`) computer program package, designed to describe a wide range of molecular and materials properties from quantum-mechanical first principles: density-functional theory (DFT) including hybrid functionals, Hartree-Fock theory, and MP2 / GW electron self-energies for total energies and excited states. An all-electron / full-potential treatment that is both computationally efficient and accurate is achieved through the use of flexible, numerically tabulated atom-centered orbitals. Preconstructed hierarchical basis sets allow to move from qualitative tight-binding like accuracy to meV-level total energy convergence with the basis set. Since all basis functions are strictly localized, the otherwise computationally dominant grid-based operations scale as $\mathrm{O}(N)$ with system size N. For local and semilocal DFT, periodic and cluster-type geometries are supported on equal footing, including relaxation and *ab initio* molecular dynamics. Together with a scalar-relativistic treatment, the basis sets allow to access all elements from light to heavy. 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. Tested architectures include single- and multicore desktop PCs, PC clusters, shared-memory IBM p-series servers, and the massively parallel BlueGene/P series.

(Submitted to: Computer Physics Communications)
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Defect Formation Energies without the Band-Gap Problem: Combining DFT and GW for the Silicon Self-Interstitial

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Abstract

We present an improved method to calculate defect formation energies that overcomes the band-gap problem of Kohn-Sham density-functional theory (DFT) and reduces the self-interaction error of the local-density approximation (LDA) to DFT. We demonstrate for the silicon self-interstitial that combining LDA with quasiparticle energy calculations in the G_0W_0 approach increases the defect formation energy of the neutral charge state by ∼1.1 eV, which is in good agreement with diffusion Monte Carlo calculations (E. R. Batista *et al.* Phys. Rev. B **74**, 121102(R) (2006), W.-K. Leung *et al.* Phys. Rev. Lett. **83**, 2351 (1999)). Moreover, the G_0W_0-corrected charge transition levels agree well with recent measurements.

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8 SCIENTIFIC HIGHLIGHT OF THE MONTH: "Optical Properties of Correlated Materials – or Why Intelligent Windows may look Dirty"

Optical Properties of Correlated Materials – or Why Intelligent Windows may look Dirty

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Abstract

Materials with strong electronic Coulomb correlations play an increasing role in modern materials applications. "Thermochromic" systems, which exhibit thermally induced changes in their optical response, provide a particularly interesting case. The optical switching associated with the metal-insulator transition of vanadium dioxide (VO_2), for example, has been proposed for use in numerous applications, ranging from anti-laser shields to "intelligent" windows, which selectively filter radiative heat in hot weather conditions.

Are present-day electronic structure techniques able to describe, or – eventually even predict – such a kind of behavior? How far are we from materials design using *correlated oxides*? These are the central questions we try to address in this Highlight.

We review recent attempts of calculating optical properties of correlated materials within dynamical mean field theory, and present results for vanadium dioxide obtained within a novel scheme aiming at particularly simple and efficient calculations of optical matrix elements within localized basis sets.

Finally, by optimizing the geometry of "intelligent windows", we demonstrate that this kind of technique can in principle be used to provide guidance for experiments, thus giving a rather optimistic answer to the above questions.

1 Towards Materials Design for Correlated Materials ?

August is the month when (at least in southern European countries) air-conditioning bills are peaking. The reader will thus forgive us if we start the August Ψ_k Scientific Highlight by describing a proposal made by materials scientists geared at saving air-conditioning costs [1–3].

As a function of temperature, vanadium dioxide (VO_2) undergoes a metal-insulator transition [4]. The changes through this transition are such that, coating your window with a thin layer of VO_2

may – under certain conditions (see below) – create a fenestration that is "intelligent" in the following sense : At low temperatures, when VO_2 is in its insulating regime, the transmission properties change only remotely with respect to the bare glass window, whereas infrared radiation is filtered when the oxide layer switches to its metallic phase in hot weather conditions. Then, heat radiation is substantially prevented from entering the building and there is less need for energy-intensive cooling by air-conditioning.

In geographical regions, where seasonal temperature changes are important (and this encompasses in particular the industrialized world), these "thermochromic" windows are clearly superior to "static" window coatings [5], that filter certain wavelengths irrespective of external conditions and which thus would increase the need for heating buildings during winter.

Even if in vanadium dioxide the changes of its properties with temperature are particularly abrupt, VO_2 is by no means the only material where a tiny change in external parameters can radically modify the physical properties. In fact, such behavior can nearly be considered a hallmark of materials with strong electronic Coulomb interactions[1]. The concerted behavior of electrons in correlated materials causes indeed quite in general an extreme sensitivity to external stimuli, such as temperature, pressure or external fields. Heating insulating $SmNiO_3$ beyond 400 K or applying a pressure of just a few kbar to the Mott insulator $(V_{1-x}Cr_x)_2O_3$ (x=0.01) [6], for example, makes the materials undergo transitions to metallic states. This tuneability of even fundamental properties is both, a harbinger for diverse technological applications and a challenge for a theoretical description.

An increasing role is nowadays played by artificial structures, ranging from multilayers that display the giant magnetoresistance effect (widely used in storage devices) [7, 8] to functional surfaces, where appropriate coatings e.g. provide a self-cleaning mechanism [9]. The huge freedom in the design, which concerns not only parameters such as the chemical composition, the doping and the growth conditions, but also the geometry of the device (e.g. the layer thicknesses), however makes the search for devices with specific electronic properties a tedious task. This leads us to the central question of the present Highlight : Can modern first principles calculations help in the quest for promising materials and setups for particular devices? The aim can of course not be to replace experiments, but rather to provide some guidance in order to minimize expensive experimental surveys and prototypings.

The Achilles heel of electronic structure theory is the description of electronic many-body interactions. Indeed, a great majority of materials used in modern applications fall in the class of so-called "strongly correlated materials" where electron-electron interactions profoundly modify (if not invalidate) a pure band picture [6]. State-of-the-art first principles methods, such as density functional theory, are then no longer sufficient to predict the physical properties of these materials. Despite these difficulties, in this Highlight we give a quite optimistic view on the above question.

In fact, important steps to bridge the gap between band structure methods and many-body physics have been made in recent years. For materials with moderately strong correlations, Hedin's GW approximation [10] – which has seen increasingly sophisticated implementations

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[1] Vanadium dioxide could in some sense be considered an exception due to the fact that it may be less correlated than many of the other examples.

in the electronic structure context [11–14] – has established itself as a method of choice. For strongly correlated materials, progress was for instance brought about by combining density functional theory within the local density approximation [15] with dynamical mean field theory (DMFT) [16]. The resulting approach, dubbed LDA+DMFT [17, 18] (for reviews see [19–21]), joins the accurate description of strong local Coulomb correlations in a many-body framework with the material-specific information provided by state-of-the-art band theory. It has – over the last years – helped to elucidate physical mechanisms at work in systems such as transition metals [22–25], their oxides [26–34] or sulphides [35, 36], as well as f-electron compounds [37–40]. The number of applications is nowadays too large to give a complete list in this Highlight, but recent reviews provide an extensive picture [19–21, 41].

While tremendous progress has been achieved, there remains a chasm between what state-of-the-art electronic structure methods can calculate and what experimentalists are actually measuring. On the one hand, the chemical complexity of many systems does simply not (yet) allow for being tackled by costly many-body techniques. On the other hand, the variety of experimental observables that are being computed for the sake of comparison is rather unsatisfactory. It is therefore an important task to make more experimentally measurable quantities accessible from theoretical calculations.

Within dynamical mean field theory, emphasis is commonly put on *spectral* properties, and the evaluation of observables other than spectral functions is a rather new advancement in the realistic context. Yet, it is rather the *response* behavior of correlated materials that is promising for applications.

In this Highlight, we review some recent attempts of calculating optical properties of correlated materials, and explore the implications for technological applications on the specific example of VO_2-based intelligent windows.

2 Optical Spectroscopy

2.1 General formula and some physical implications

Numerous experimental techniques have been devised for and applied to the study of correlated materials of ever growing complexity. Optical spectroscopy, which is the subject of this work, is, in a way, the most natural among them : Optical detectors are sampling the response to incident light, as do our eyes, albeit accessing frequencies, and thus phenomena, that are beyond our vision. The technique is particularly suited to track the evolution of a system under changes of external parameters like temperature or pressure. This is owing to a generally high precision, and the fact that, contrary to e.g. photoemission spectroscopy or x-ray experiments, results are obtained in absolute values. Especially, the existence of sum-rules (see e.g. [42, 43]) allows for a quantitative assessment of transfers of spectral weight upon the progression of the system properties. Moreover, while in photoemission the electron escape depth and thus surface effects are often an issue, the larger skin penetration depth assures that optical spectroscopy is a true bulk probe. One might add that the transition matrix elements are also better understood in optics (this is an important part in this work) than in photoemission (see e.g. [44]). On the other hand, response functions are two-particle quantities that are less obvious in their interpretation than a one-particle spectrum. An important simplification is achieved when neglecting vertex corrections. In the framework of linear response theory, the optical conductivity can then be expressed as (for reviews see [34, 42])

$$Re\,\sigma^{\alpha\beta}(\omega) \quad = \quad \frac{2\pi e^2\hbar}{V}\sum_{\mathbf{k}}\int d\omega' \,\frac{\mathrm{f}(\omega')-\mathrm{f}(\omega'+\omega)}{\omega}\;\mathrm{tr}\Big\{A_{\mathbf{k}}(\omega'+\omega)v_{\mathbf{k},\alpha}A_{\mathbf{k}}(\omega')v_{\mathbf{k},\beta}\Big\} \tag{1}$$

Here, $A_{\mathbf{k}}(\omega)$ is the momentum-resolved many-body spectral function, and different optical transitions are weighted by the Fermi velocities $v_{\mathbf{k},\alpha} = \frac{1}{m}\langle \mathbf{k}L'|\mathcal{P}_\alpha|\mathbf{k}L\rangle$, matrix elements of the momentum operator $\mathcal{P}$. Both, spectral functions and velocities are matrices in orbital space L, which we will specify later on. The Fermi functions $\mathrm{f}(\omega)$ select the range of occupied and empty energies, respectively, V is the unit-cell volume, α,β denote cartesian coordinates, and $Re\,\sigma^{\alpha\beta}$ is the response in $\alpha-$direction for a light polarization E along β.

To get an idea about the physical content of this formula, we first consider some archetypical cases. At zero temperature, and for a system that is well described by its band structure as given by density-functional theory (DFT) based methods, the (Kohn-Sham) spectrum is the defining quantity, since vertex correction are absent[2]. Moreover, the spectral functions $A(\mathbf{k},\omega)$ of the system become Dirac distributions and the trace in Eq. (1) reads[3]

$$\sum_{m,n}\delta(\omega'+\omega+\mu-\epsilon^m_{\mathbf{k}})v^{mn}_{\mathbf{k}}\delta(\omega'+\mu-\epsilon^n_{\mathbf{k}})v^{nm}_{\mathbf{k}} \tag{2}$$

While at finite frequencies, $\omega > 0$, only inter-band transitions, $m \neq n$, can give a contribution, we see that at zero frequency, $\omega = 0$, and provided a band $\epsilon^n_{\mathbf{k}}$ is crossing the Fermi level, the response is a delta function deriving from intra-band transitions[4]. The latter is just the result of the fact that without interactions (electron-electron correlations or a coupling to a bosonic mode) or disorder, the lattice momentum $\mathbf{k}$ is a constant of motion and the current thus does not decay.

In the (effective) non-interacting case the response of a metal to an electric field is thus infinite. It was P. Drude in 1900 who derived an expression that takes into account the finite lifetime of the electron excitations, by introducing a relaxation time τ for the charge current $j(t) = j(0)e^{-t/\tau}$. His expression for the conductivity can be recovered from our linear response result, Eq. (1), by assuming free particles, $\epsilon_{\mathbf{k}} = \frac{\hbar^2k^2}{2m}$, and a constant self-energy $\Sigma = -\frac{\imath}{2\tau}$. The resulting optical conductivity is then given by

$$Re\,\sigma(\omega) = \frac{ne}{m}\frac{\tau}{1+\omega^2\tau^2} \tag{3}$$

with n being the average charge carrier density. Hence, the response at zero frequency is finite and non-zero at finite energies.

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[2] There are thus two effects that DFT does not account for : (a) In general, it lacks the correct spectrum. This is mended by the formalism described above, since it uses spectral functions stemming from a many-body calculation. (b) The particle-hole interaction (vertex corrections). This is not addressed in the above formalism and remains a challenge for future work. We note that vertex corrections vanish within DMFT (infinite dimensions) in the one-band case [45]. For the current multi-orbital case, this is however an approximation. Within GW [11], vertex corrections are included on the RPA level. However, GW *spectra* may not be sufficient for strongly correlated systems.

[3] for simplicity we assume to work in the Kohn-Sham basis, i.e. the spectral function $A_{\mathbf{k}}(\omega)$ is diagonal.

[4] or transitions within degenerate bands, $\epsilon^n_{\mathbf{k}} = \epsilon^m_{\mathbf{k}}$, while $m \neq n$.

While the physical picture about the origin of the relaxation time had to be revised with the advent of quantum mechanics and Bloch's theory of electronic states in periodic potentials, Drude's theory still accounts well for the response of simple metals. However, it becomes insufficient, even in the one-band case, in the presence of substantial correlation effects. Indeed, the influence of electronic interactions is beyond a mere broadening of bands. For the case of the one-band Hubbard model within DMFT, the paramount characteristic is the appearance of Hubbard satellites in the spectral function. Therewith, not only transitions within a broadened quasi-particle peak are possible (à la Drude), but also transitions from and to these Hubbard bands arise [46, 47]. Thus, in such a metal two additional contributions occur, stemming from transitions between the quasi-particle peak and the individual Hubbard bands. This feature is often referred to as the mid-infrared peak, due to its location in energy in some compounds. At higher energy, transitions between the two Hubbard bands appear. In the Mott insulating phase, only the latter survive. Therewith the complexity of optical spectra is considerably enhanced [46, 47] with respect to the independent particle picture.

Within DMFT, calculations of the optical conductivity were first performed by Pruschke and Jarrell *et al.* [48, 49] for the case of the Hubbard model. Rozenberg *et al.* [46, 47] studied the phenomenology of the different optical responses of the Hubbard model throughout its phase diagram, discussing the above described phenomenology and comparing it to experiments on V_2O_3. With the advent of LDA+DMFT, optical conductivity calculations gained in realism: Blümer *et al.* [50, 51] and later on Pavarini *et al.* [52] and Baldasarre *et al.* [53] used the LDA+DMFT spectral functions for the calculation of titanate and vanadate optical spectra. A more general approach, was developed by Pálsson [54] for the study of thermo-electricity. Our work [34, 55–57] goes along the lines of these approaches. We will however employ a formulation using the full valence Hamiltonian therewith allowing for the general case including interband transitions and we extend the intervening Fermi velocities to multi-atomic unit cells, which becomes crucial in calculations for realistic compounds.

Alternative techniques were proposed by Perlov *et al.* [58, 59] and by Oudovenko *et al.* [60]. The former explicitly calculated the matrix elements, albeit using a different basis representation than in the DMFT part, while the latter diagonalized the interacting system, which allows for the analytical performing of some occurring integrals due to the "non-interacting" form of the Green's function. Owing to the frequency-dependence of the self-energy, however, the diagonalization has to be performed for each momentum and frequency separately so that the procedure may become numerically expensive. This technique has in particular been applied to elucidate the $\alpha - \gamma$ transition in Ce [61], and, more recently, to the heavy fermion compound CeIrIn_5 [62].

The calculation of accurate absolute values of the optical response has however turned out to be a challenge which mainly stems from the way the Fermi velocities are treated. In view of predictive materials design accurate absolute values in the optical conductivity are a condition *sine qua non*.

2.2 A scheme for the optical conductivity using localized basis sets

Correlation effects enter the calculation of the optical conductivity, Eq. (1), only via the spectral functions $A_{\mathbf{k}}(\omega)$, while the Fermi velocities $v_{\mathbf{k},\alpha} = \frac{1}{m}\langle \mathbf{k}L'|\mathcal{P}_\alpha|\mathbf{k}L\rangle$ are determined by the one-particle part of the system[5].

While the computation of the latter is straightforward in a plane-wave setup, the application of many-body techniques such as LDA+DMFT, necessitates the use of localized basis sets (see e.g. [63]). It is thus convenient to express also the Fermi velocities in terms of this basis. Therewith, however, the evaluation of matrix elements of the momentum operator (see above) becomes rather tedious. The aim of the current approach is to employ a controlled approximation to the full dipole matrix elements in a Wannier like basis set that is accurate enough to yield absolute values of the conductivity to allow for a quantitative comparison with experiment.

We choose our orbital space by specifying $L = (n, l, m, \gamma)$, with the usual quantum numbers (n, l, m), while γ denotes the atoms in the unit cell : $|\mathbf{k}L\rangle$ then is the Fourier transform of the Wannier function $\chi_{\mathbf{R}L}(\mathbf{r})$ localized at atom γ in the unit cell $\mathbf{R}$. Extending the well-known Peierls substitution approach for lattice models (see the review [42]) to the realistic case of multi-atomic unit cells, we find that

$$v^{L'L}_{\mathbf{k},\alpha} = \frac{1}{\hbar}\Big(\partial_{k_\alpha}\mathbf{H}^{L'L}_{\mathbf{k}} - \imath(\rho^\alpha_{L'} - \rho^\alpha_L)\mathbf{H}^{L'L}_{\mathbf{k}}\Big) + \mathcal{F}_{\mathbf{H}}\left[\{\chi_{\mathbf{R}L}\}\right] \tag{4}$$

Here, ρ_L denotes the position of an individual atom within the unit-cell. The term in brackets, which is used in the actual calculations, is in the following referred to as the "generalized Peierls" term : While the derivative term is the common Fermi velocity, the term proportional to the Hamiltonian originates from the generalization to realistic multi-atomic unit-cells and accounts for the fact that while the periodicity of the lattice is determined by the unit-cell, the Peierls phases couple to the real-space positions of the individual atoms. The correction term that recovers the full matrix element is denoted $\mathcal{F}$ (for its explicit form see [34]). The latter reduces to purely atomic transitions, $(\mathbf{R}, \gamma) = (\mathbf{R}', \gamma')$, in the limit of strongly localized orbitals. In other words, the accuracy of the approach is controlled by the localization of the basis functions. This generalized Peierls approach has in particular been shown to yield a good approximation for systems with localized orbitals, such as the 3d or 4f orbitals in transition metal or lanthanide/actinide compounds [34].

3 Application :
From Bulk Vanadium Dioxide to Intelligent Windows

3.1 A brief reminder about the electronic structure of VO_2

The electronic structure of VO_2 and its metal-insulator transition has been the subject of numerous theoretical studies. We give here only a brief list of prior work, for reviews see [34, 64].

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[5] While for lattice models this is only true for local density-density interactions, in the continuum formulation of a solid this requires the interaction only to be local and of two-body kind. Then the interaction part of the Hamiltonian $\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_{int}$ commutes with the position operator $\mathcal{R}_\alpha$ and thus $\mathcal{P}_\alpha = -\imath m/\hbar\,[\mathcal{R}_\alpha, \mathbf{H}_0]$. For details see [34].

In particular, we here do not enter at all in the decade-long discussion on whether the metal-insulator transition in VO_2 should be considered as a Peierls- or a Mott transition. Our point of view on this question is summarized in [33].

While capturing structural properties surprisingly well [65], band-structure methods do not reproduce experimental spectra: in the metal, incoherent weight at higher binding energies is absent, and in the insulator the gap is not opened in the corresponding Kohn-Sham spectra. Nevertheless, thorough LDA studies [64, 65] gave useful indications and paved the road to the application of more sophisticated many-body approaches.

LDA+DMFT results for the spectral properties of VO_2 agree well with experimental findings in both, the metallic [32, 66, 67] and the insulating phase [32], as can be seen from Fig. 1 which compares photoemission and LDA+DMFT spectra.

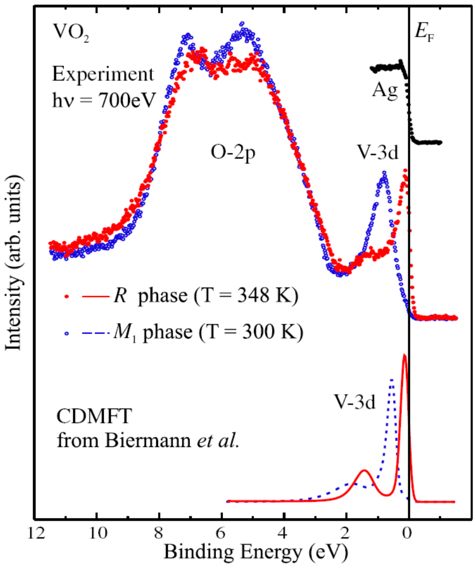
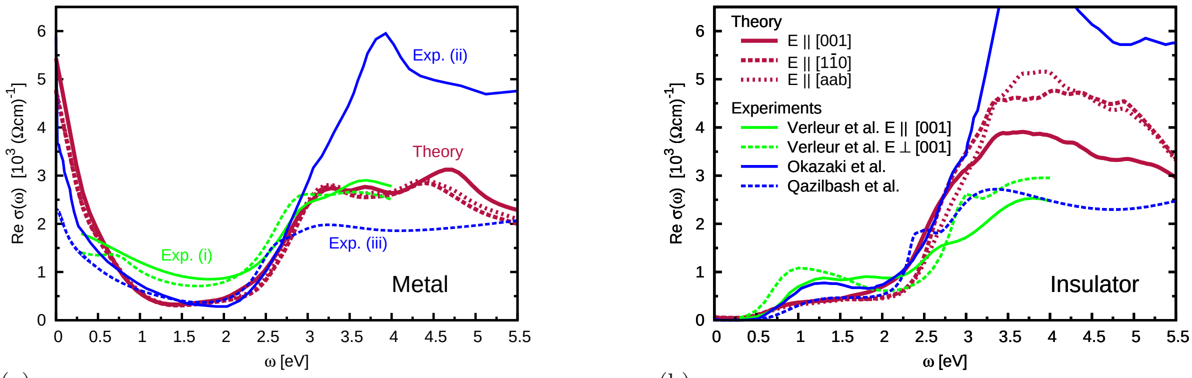


Figure 1: Comparison of valence band photoemission spectra of Koethe *et al.* [68] (top) and LDA+DMFT results from [32] (bottom). Picture from [68].

Since at least the insulating phase of VO_2 is in fact relatively band-like [33], an interesting alternative approach is provided by the GW approximation [11]. While pioneering early work [69] had to resort to a simplified scheme, recently it became possible to perform fully *ab initio* GW calculations for VO_2 [34, 70, 71]. Full GW calculations for the optical conductivity and the spectral function thus seem to come into reach, opening the way to systematic comparisons with DMFT results.

Our calculation of optical properties [34, 55] is footing on the LDA+DMFT electronic structure of Ref. [32] and our recent extension thereof [33, 72]. Since this many-body calculation used a downfolded Hamiltonian, we use an upfolding scheme to include optical transition from, to and between higher energy orbitals (For details see [57]).



(a) (b)

Figure 2: Optical conductivity of (a) metallic, (b) insulating VO_2 for polarizations E. Theory (red) ([aab]=[0.85 0.85 0.53]), experimental data (i) single crystals [73] (green), (ii) thin film [75] (solid blue), (iii) polycrystalline film [74] (dashed blue).

3.2 The bulk conductivity

In Fig. 2(a) we display the LDA+DMFT theoretical optical conductivity of the high temperature phase of VO_2 as a function of frequency and in comparison with several experimental data [73–75], see also [76]. As can be inferred from the crystal structure [64], the optical response depends only weakly on the polarization of the incident light. The Drude-like metallic response (see also above) is caused by transitions between narrow vanadium 3d orbitals near the Fermi level. As a consequence, it only influences the low infra-red regime – a crucial observation as we shall see in the following. The shoulder at 1.75 eV yet stems from intra-vanadium 3d contributions, while transitions involving oxygen 2p orbitals set in at 2 eV, and henceforth constitute the major spectral weight up to the highest energies of the calculation.

In Fig. 2(b) we show the conductivity for insulating VO_2. As was the case before, the theoretical results are in good agreement with the different experiments. This time, a slight polarization dependence is seen in both, experiment and theory, owing to the change in crystal symmetry. Indeed, vanadium atoms pair up in the insulator to form dimers along the c-axis, leading to the formation of bonding/anti-bonding states for 3d orbitals[6] [77]. Optical transitions between these orbitals result in an amplitude of the conductivity that, in the corresponding energy range ($\omega = 1.5 - 2.5$ eV), is higher for a light polarization parallel to the c-axis than for other directions (For a detailed discussion see [34, 56]).

3.3 Intelligent windows

Having established the theoretical optical response of *bulk* VO_2, and thus verified that our scheme can *quantitatively* reproduce optical properties of correlated materials, we now investigate the possibilities of VO_2-based intelligent window coatings [2, 3, 78, 79]. The effect to be

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[6] We stress that the bonding/anti-bonding splitting is not sufficient to open a gap within band-structure approaches. Indeed correlations enhance this splitting (after having transfered the e_g^π charge into the a_{1g} band), and we refer to this scenario as a “many-body Peierls insulator” [33].

exploited here can already be seen in the above responses of the bulk : The respective conductivities of both phases (Fig. 2(a), (b)) exhibit a close similarity in the range of visible light ($\omega = 1.7 - 3.0$ eV), whereas in the infra-red regime ($\omega < 1.7$ eV) a pronounced switching occurs across the metal-insulator transition. As a result, heat radiation will be let through at low external temperatures, while its transmission will be hindered above the transition, which reduces the need for air conditioning in e.g. office buildings. The insensitivity to temperature for visible light, in conjunction with the selectivity of the response to infrared radiation, is an essential feature of an intelligent window setup. Yet, for an applicable realization, other important requirements have to be met. First of all, the switching of the window has to occur at a relevant, i.e. ambient, temperature. Also, the total transmittance of VO_2-films needs improvement in the visible range [2,3], and the visible transmission should be rather independent of the wavelength such as to provide a colorless vision. Experimentalists have addressed these issues and have proposed potential solutions [80] : Diverse dopings, $M_xV_{1-x}O_2$, were proven to influence the transition temperature, with Tungsten ($M = W$) being the most efficient : A doping of only 6% resulted in $T_c \approx 20°C$ [81]. However, this causes a deterioration of the infrared switching. Fluorine doping, on the other hand, improves on the switching properties, while also reducing T_c [82,83]. An increase in the overall visible transmittance can also be achieved without modifying the intrinsic properties of the material itself, but by adding antireflexion coatings with for example TiO_2 [84].

Here, we address the optical properties of window coatings from the theoretical perspective. In doing so, we assume that the specular response of VO_2-layers is sufficiently well-described by the optical properties of the bulk, and we use geometrical optics to deduce the properties of layered structures, as shown e.g. in Fig. 3(a), Fig. 4(a). We employ a technique equivalent to the transfer matrix method (see e.g. [85]), which accounts for the multiple reflexions within the layers. The reflectivity of the setup is then given by a recursion formula. Defining for the j^{th} layer the phase factors $\alpha_j(\omega) = \exp(\imath\omega/cn_j\delta_j)$, with δ_j and $n_j(\omega)$ being the layer thickness and the complex refractive index, respectively, and denoting the "bare" reflection and transmission coefficients of an isolated interface between semi-infinite layers $j-1$, j by $r_{j-1,j}$, $t_{j-1,j}$ one obtains a recursion formula for the "dressed" quantities $\tilde{r}_{0,j}$, $\tilde{t}_{0,j}$ that account for the resulting properties of the stack of the layers 0, 1, $\cdots$, j :

$$\tilde{r}_{0,j} = \tilde{r}_{0,j-1} + \tilde{t}_{0,j-1}\tilde{t}_{j-1,0}\frac{r_{j-1,j}\tilde{\alpha}^2_{j-1}}{1 - r_{j-1,j}\tilde{r}_{j-1,0}\tilde{\alpha}^2_{j-1}} \quad (5)$$

$$\tilde{r}_{j,0} = r_{j,j-1} + t_{j,j-1}t_{j-1,j}\frac{r_{j-1,0}\tilde{\alpha}^2_{j-1}}{1 - r_{j-1,j}\tilde{r}_{j-1,0}\tilde{\alpha}^2_{j-1}} \quad (6)$$

where $\tilde{\alpha}_j = \prod_{k \leq j} \alpha_k$ is an effective phase, $\tilde{t}_{j,k} = 1 - \tilde{r}_{j,k}$ the complex transmission, and $\tilde{r}_{j,0}$ the dressed reflectivity for light that reaches the setup from the opposite side.

3.3.1 A VO_2-coated window

First, we consider the most simple possible setup, which consists of a single VO_2-layer (of thickness d_{VO_2}) on a glass substrate[7]. Such a window has been experimentally investigated by

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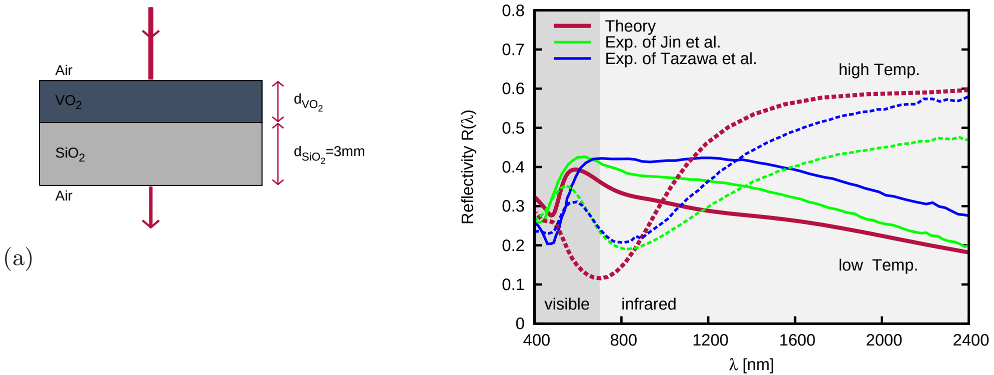
[7] We suppose quartz glass, SiO_2. All auxiliary refractive indices (other than the one of VO_2) are taken from *Handbook of Optical Constants of Solids* by Edward D. Palik, Academic Press, 1985.

Tazawa *et al.* [86] and Jin *et al.* [84]. In Fig. 3 we show their measured reflectivity data as a function of wavelength, in comparison with our theoretical results.



(a)

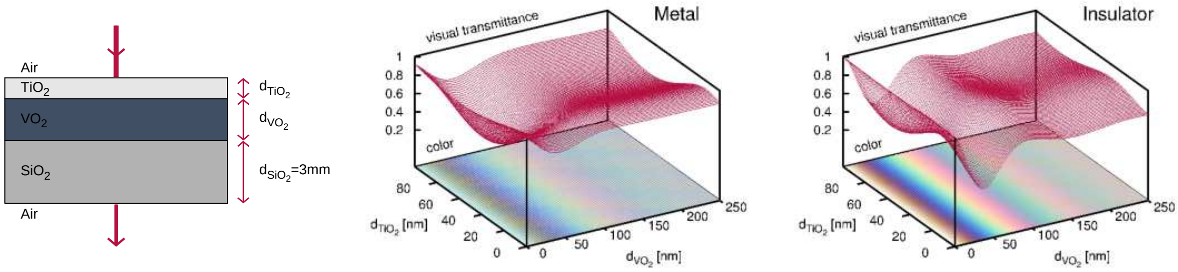
(b)

Figure 3: Setup of an intelligent window. (a) geometry of the setup : VO_2 on SiO_2. (b) Reflectivity at high (dashed) and low temperatures (solid). Theory (red) : 60 nm VO_2 on SiO_2. Experiments : 50 nm VO_2 on SiO_2 [84] (green), 50 nm VO_2 on Pyrex glass [86] (blue). All theoretical data for $E \parallel [1\bar{1}0]$ polarization, and a 3mm SiO_2 substrate.

At low temperatures, i.e. insulating VO_2, the calculated reflectivity is in quantitative agreement with the experimental data. In the visible range ($\lambda \approx 400 - 700$ nm), the reflectivity depends strongly on the wavelength. Therefore, the current window will filter certain wavelengths more than others, resulting in an illumination of a certain color – an obvious drawback. Moreover, the reflectivity in this region is rather elevated, causing poor global transmission. In the infrared regime ($\lambda > 700$ nm) – and beyond – the reflectivity decreases, and heat radiation can pass through the setup. At high temperatures, the infrared reflectivity switches to a rather elevated value, thus filtering heat radiation. The changes in the visible region are less pronounced, but still perceptible, and both, the degree of transparency and the color change through the transition. As a result, though presenting qualitative features of an intelligent window, the current setup is not yet suited for applications.

3.3.2 Improving the window with an anti-reflexion layer

Having established the accuracy of our approach also for the case of layered structures, we now investigate a more complicated setup. As depicted in Fig. 4(a), an additional (rutile type) TiO_2-coating is added on the VO_2-layer, with the objective of serving as an antireflexion filter [84]. With the thicknesses d_{VO_2} and d_{TiO_2}, the geometry of the current setup thus has already two parameters that can be used to optimize the desired optical properties. Since, however, each variation of them requires the production of a new individual sample under comparable deposition conditions, along with a careful structural characterization in order to guarantee that differences in the optical behavior are genuine and not related to variations of the sample quality, the experimental expenditure is tremendous. This led Jin *et al.* [84] to first estimate a highly transmitting setup by using tabulated refractive indices and to produce and measure only *one* such sample. Here, we shall use our theoretical results on VO_2 to not only optimize the geometry (d_{TiO_2}, d_{VO_2}) with respect to the total visible transmittance, but we shall be concerned with yet another important property of the window that needs to be controlled : its color.



(a)

(b)

(c)

Figure 4: Setup of an intelligent window. (a) geometry of the setup : TiO_2 on VO_2 on SiO_2. (b), (c) Setup with antireflexion coating : TiO_2 / VO_2 on SiO_2. Shown is the normalized visible transmittance (see text) an the corresponding color of the transmitted light for (b) high and (c) low temperature as a function of the layer thicknesses $d_{\mathrm{VO_2}}$ and $d_{\mathrm{TiO_2}}$. All theoretical data for $E \parallel [1\bar{1}0]$ polarization, and a 3mm SiO_2 substrate.

Fig. 4 displays the normalized visible specular transmittance[8] for our window in its high (b) and low (c) temperature state, as a function of both film thicknesses. On the same graph, we moreover show the corresponding transmission color. The evolution of the light interferences within the layers results in pronounced changes in both, the overall transmittance and the color. The coating of VO_2 globally degrades the transparency of the bare glass window. An increase of the TiO_2-coating, on the other hand, has the potential to improve on the total transmittance. This can be understood from the mechanism of commonly used quarter-wave filters. The wavelength-dependence of the real-part of the TiO_2 refractive index, $n_{\mathrm{TiO_2}}(\lambda)$, results in an optimal quarter-wave thickness, $\delta_{\mathrm{TiO_2}}(\lambda) = \lambda/(4n_{\mathrm{TiO_2}}(\lambda))$, which varies from blue to red light only slightly from $\delta_{\mathrm{TiO_2}}(\lambda) = 40$ to 60 nm. This and the fact that the imaginary part of the refractive index, $k_{\mathrm{TiO_2}}(\lambda)$, is negligible for visible light also explains why the color does not change significantly with $d_{\mathrm{TiO_2}}$. While, as for TiO_2, the variation of the real-part of the VO_2 refractive index yields a rather uniform ideal thickness $\delta_{\mathrm{VO_2}}(\lambda)$, its imaginary part changes significantly (by a factor of 4) within the range of visible light. As a consequence, the color is very sensitive to VO_2-deposition. At higher thickness $d_{\mathrm{VO_2}}$, however, this dependence becomes smaller and the color lighter, as seen in Fig. 4(b) and (c). Our theoretical transmittance profiles suggest relatively thick windows to yield good visual properties. Indeed, at low temperatures (Fig. 4(c)) the local maximum that gives the thinnest window (Fig. 4(b)) is located at $(d_{\mathrm{TiO_2}}, d_{\mathrm{VO_2}}) \approx (40\ \mathrm{nm}, 85\ \mathrm{nm})$ within our calculation. However, this setup is still in the regime of important color oscillations. Given the uncertainties in industrial deposition techniques, it seems rather cumbersome to consistently stabilize colorless samples. From this point of view, a thicker VO_2-film would be desirable. Indeed, while almost preserving the overall transmittance, a colorless window at low temperatures is realized in our calculation for $(d_{\mathrm{TiO_2}}, d_{\mathrm{VO_2}}) \approx (50\ \mathrm{nm}, 220\ \mathrm{nm})$, or for $(d_{\mathrm{TiO_2}}, d_{\mathrm{VO_2}}) = (\geq 100\ \mathrm{nm}, 220\ \mathrm{nm})$. In the high temperature state, Fig. 4(b), the

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[8] $\int_{400nm}^{700nm} d\lambda S(\lambda)T(\lambda) / \int_{400nm}^{700nm} d\lambda S(\lambda)$, with the spectrum of the light source $S(\lambda)$, and the transmittance $T = 1 - R$, R being the specular reflexion given by Fresnel's formulae (see e.g. [43]). We thus neglect absorption due to inhomogeneities that lead to diffuse reflexion. This is justified for our applications to windows. Also, VO_2 has a glossy appearance and hence a preponderant specular response.

transmittance is globally lower than at low temperatures. Moreover, only the $(d_{\mathrm{TiO_2}}, d_{\mathrm{VO_2}}) = (\geq 100\ \mathrm{nm}, 220\ \mathrm{nm})$ setup exhibits a simultaneously high transmittance in *both* states of the window.

4 Conclusions and Perspectives

In this Highlight, we have reviewed a scheme for optical properties of correlated materials that is geared at fast and accurate calculations within localized basis sets.

As an example, we have addressed – for the first time from a theoretical perspective – the fascinating application of the seemingly simple composition of vanadium dioxide in so-called intelligent windows.

Even if, from a technical point of view, possible ways of improvement in view of a fully general optics scheme are obvious – ideally, one may want to include e.g. vertex corrections, replace the LDA+DMFT starting point by a GW+DMFT calculation [87] or calculate at least the ligand-orbital energies by many-body techniques such as GW – our simple and efficient optics scheme has proven to reach quantitative agreement with experiments, at least for materials where excitonic effects are negligible. We have shown that it is not only useful to address questions of fundamental physics, but that one can indeed think of exploiting present day techniques for applied purposes.

One may argue, of course, that a crucial ingredient entering LDA+DMFT calculations is a reliable estimate for the local Coulomb interactions, the Hubbard U. In general, it is probably a fair statement to say that – despite a number of techniques that have by now been proposed for the calculation of U (constrained LDA [88], constrained RPA [89,90], or GW+DMFT [87,91,92]) – its determination still presents one of the bottlenecks for *ab initio* materials design. The recently much discussed new family of iron pnictide superconductors provides an interesting test case for calculations of U [93, 94], where we dispose of little *a priori* knowledge.

For vanadium dioxide, however, this issue seems to be more a conceptual than a practical one, since experimental and theoretical estimates finally converge towards a common answer.

In conclusion, we thus give an optimistic answer to the central question of this Highlight : electronic structure techniques can – if not design – at least help guiding the search for functional materials and their devices, and this even for the particularly challenging class of correlated materials.

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