Ψ_k Newsletter

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

Number 101 October 2010

Editor: Z (Dzidka) Szotek
E-mail: psik-coord@stfc.ac.uk

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Contents

1 Editorial

With this issue, No 101, we enter the next, 2nd, hundred "pack" of the Psi-k Newsletters. Seeing the enthusiasm at the Psi-k2010 Conference in Berlin, the future of the Psi-k family, and thus also the newsletter, seems promising.

We dedicate the first section of this issue to the Psi-k2010 Conference and encourage everybody to at least glance through it. In particular the views of a small sample of participants printed in one of its subsections make a very interesting reading. We are extremely grateful to all who agreed to share with us their personal views on the Conference. There are also a few conference photos to be found here and there to make the whole thing an 'archive document'-like.

There are also some workshop and job announcements in this newsletter, and quite a few abstracts of recent or newly submitted papers. Just before the scientific highlight article, we have a short information on a method/code USPEX (for crystal structure prediction) by A Oganov and collaborators, which is free to download.

The scientific highlight article of this issue is by Jan Minar (LMU Munich, Germany) on "Correlation effects in transition metals and their alloys studied by the fully self-consistent KKR+DMFT method".

Please check the table of contents for details on this Psi-k Newsletter.

The *Uniform Resource Locator* (URL) for the Psi-k webpage is:

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

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

The email address for contacting us and for submitting contributions to the Psi-k newsletters is

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

Z (Dzidka) Szotek, Martin Lüders and Walter Temmerman
E-mail: psik-coord@stfc.ac.uk

2 Psi-k2010 Conference

`http://www.fhi-berlin.mpg.de/th/Meetings/psik_2010/`

13-16 September

Henry Ford Building (HFB), Berlin, Germany

Since this is the first newsletter after the recent Psi-k2010 Conference, it seems almost natural to dedicate a prominent part of it, specifically the three following subsections, to reminescing on this event.

In the first subsection we provide only a brief, likely subjective, editorial documentation and selected photographs of the event. In the next subsection we present views, comments and impressions of other participants, because, as Volker Heine said in his closing remarks, " ... these are the participants that make the conference". The ones who have contributed their comments to the newsletter represent different ages, sexes, countries/continents, etc., and their brief reports make a very interesting reading. We are truly grateful to them for taking the time to record their thoughts, comments and impressions for the newsletter.

The final subsection is by Peter Dederichs and is dedicated to the Volker Heine Young Investigator Award and the associated session at the Psi-k2010 Conference.

2.1 Brief editorial documentation of the event

The Conference was organized by Matthias Scheffler (chair), Hardy Gross (co-chair), and Volker Heine (honorary chair), with the help of the whole group of Matthias at the FHI Berlin. The organization seemed perfect from start to finish. The whole team, in black T-shirts with the Psi-k2010 Conference yellow logo, was on stand-by from the registration on the Sunday early evening, 12th of September. Soon the whole foyer of HFB and the outside filled with participants carrying the characteristic black bags with the yellow logo, containing a very imaginative set of useful conference stuff, and in particular black umbrellas, again marked by the yellow Conference logo. Congratulations to the organizing team for being so insightful and predictive regarding the weather in Berlin.

The Conference was officially opened by Matthias on the Monday morning, who extended a heartfelt welcome to everybody, expressed vividly through the little jelly teddy bears carrying a big red heart, seen in the photo below, welcoming all to Berlin.

Based on the facts, this was the largest of the four Psi-k conferences, with over 1000 participants, five parallel sessions and the same number of plenary talks, about 120 invited talks, 22 symposia and about 700 poster presentations. The latter were distributed over two poster sessions; about 350 posters on a day, with even and odd numbers receiving 1,5 hours slots in succession. This was a very good idea, allowing an easier access to the posters, as well as, giving everyone a chance to see the posters. Of course, a longer time for discussions at any poster would have been useful, although likely difficult to realize, given a very tight programme, and the constantly full and buzzing foyer (see photos below).

It definitely felt very dynamic and full of energy.

Plenary sessions were usually very well attended with the largest, Audimax, hall rather full on all the ocassions. The five talks covered a broad range of topics, starting from computational spectroscopy, involving a number of theoretical methods and implemented in Quantum ESPRESSO code (S. Baroni), through fuels from sunlight (J. Norskov), Berry phase/potential and curvature, and how they impact on the theories of orbital magnetization and electric polarization (D. Vanderbilt), materials genome project (G. Ceder) and finishing with molecular transport junctions (M. Ratner).

Of course, with five symposia running in parallel it was impossible for an individual to visit a substantial number of invited and contributed talks or to get an idea which symposia were most popular. However, it seemed that talks presenting new methodological and computational developments were, as usual, most highly appreciated.

The oral and poster presentations and scientific discussions in the foyer were providing plenty of food for thought, but access to real food was also easy and well managed. The whole food distribution process ran very smoothly. Fruits, snacks and drinks were nearly always available in the foyer, while the lunches and evening meals were served in another building, in close proximity to HFB. The efficient access to food and sitting areas is another thing for which the organizers deserve full marks (see photos below).

The conference dinner was quite unusual and happend on the Wednesday evening/night under the banner "The Conference outing" in an interesting "Rodeo" museum and club (see photos below).

It was certainly a conference dinner with the difference and probably highly regarded by young participants. The waiting for food was a bit long and tiring, but that was presumably to be considered as an integral part of the unusual set. All in all, with three bands, two DJs and at 1 am several hundred theoreticians still in motion, it was an interesting experience even for the older folks. As a result, the fifth plenary talk was perhaps less attended than the other four, but the numbers were still quite impressive.

A new element of this Psi-k Conference was a Young Investigator Award, set up in the name of Volker Heine, the grand father of the Psi-k family. It was associated with an interesting and well attended session, in which the five finalists gave 30 minutes' presentations. In one of the following sections Peter Dederichs, the Psi-k Network chairman, reports on this event and the award itself in more detail.

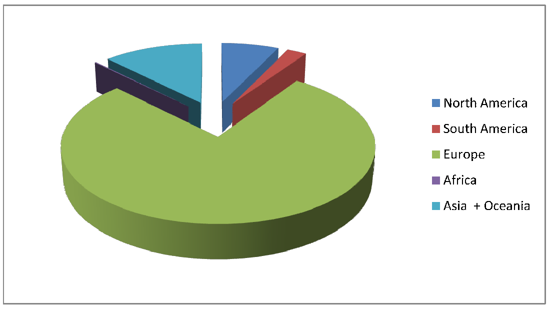
At the closing words, both Volker Heine and Peter Dederichs praised very highly Matthias, Hardy and the rest of the organizing team for a fantastic and smoothly run conference. Not only Volker, seen in the photo below, was aplauding them very warmly, but all the remaining participants joined in a long and loud clapping. Below we see Matthias and Hardy, sitting behind each other, having very solemn and concerned faces. Surely, they must have been pleased with the outcome and really happy for being so highly appreciated by everybody. Nevertheless, they must have also been relieved that it was finally over.

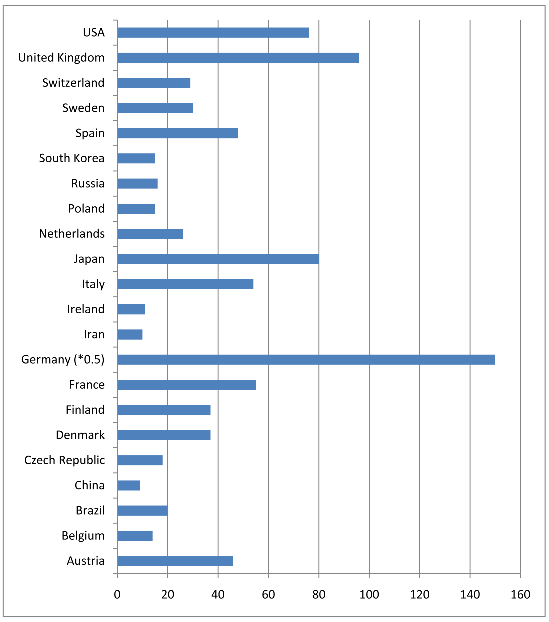
The Conference was being very accurately documented in photographs and the album can be accessed from the webpage

`http://www.fhi-berlin.mpg.de/th/Meetings/psik_2010/album/index.html`

In the above write-up we have only selected a very few of them to give it a bit of an 'archive document'-like feel.

To finish this documentation of the event it seems interesting to look at the distribution of the participants over countries and then continents. In the graphs below, one can see that Europe leads the field, with Germany well ahead with impressive 299 participants, followed by UK (96), France (55), Italy (54), Spain (48), Austria (46), and so on. North America falls behind Asia+Oceania, where Japan leads with 80 participants out of the total of 118. It is perhaps surprising that there were only two participants from the whole continent of Africa, despite the fact that there are a few groups there, in particular in the North African countries, that are active in the field. Maybe even more surprising is the fact that there were only 9 participants representing China, whilst a small country like Czech Republic had 18 participants.





2.2 Brief Reports on the Psi-k2010 Conference by a Sample of Participants

Comment by John Dobson (Brisbane, Australia)

This was the biggest physics conference I have attended, except for the APS March meeting, which is of course much less focussed. The size of the meeting reflects the very pleasing success of the Psi-k network, and is a tribute to the hard work of many people from Volker Heine onwards. Despite its size and complexity, the meeting ran amazingly smoothly. The programming, the building, rooms and food service were all first-class, and at any sign of a problem the army of helpers in black T-shirts would be there to sort things out. Many thanks to Matthias Scheffler, Hardy Gross, Peter Dederichs, Walter Temmerman and their T-shirt brigade. As a non-EU attendee, I was also impressed by the welcoming and inclusive treatment that I received. In general, the Psi-k network seems to have succeeded in fostering cooperation rather than the competitive approach that is often evident elsewhere in the physics community.

The scientific content and presentation were both of a very high standard, and the subject matter was quite diverse, given the focus area of Psi-k. I will comment on the technical content from my personal viewpoint as a theorist interested in van der Waals physics and related many-body formalisms. For me the highlights from the talks and posters were the following.

(i) The emerging ability to model periodic systems in detail via microscopic many body methods, starting from the RPA/TDDFT correlation energy. Microscopic beyond-RPA corrections (e.g. SOSEX) are also now becoming available, and they seem to improve the theory where tested to date. One of the important achievements of this class of microscopic approach is the reliable ab initio description of dispersion forces in "difficult" systems such as graphitics. There is still a need for much work here, however, since the computational effort of RPA-class theory still precludes full description of nanostructures with ”interesting” geometries and correspondingly large repeat cells. At least two groups also reported good progress with the tricky ”OEP” problem of providing selfconsistent orbitals for the RPA class of theory. It seems that this can make major improvements in the description of small systems where ”direct” RPA has some serious diseases.

(ii) The progress evident in the modelling of more complex dispersion-bound systems via more approximate and/or empirical theories. One example was an efficient transcription procedure to obtain polarisabilities of ”compressed” atoms in molecules and solids starting from accurate free-atom polarisabilities. There was also the discovery (surprising to me at least) that dispersion forces are important in the stability of beta helices in biomolecules, i.e. in determining secondary structures rather than just tertiary structure where such effects were already considered to be important.

(iii) The successful application of GW and Bethe-Salpeter equation technologies to predict quasi-particle and excitonic properties in a variety of solids.

John Dobson
Griffith University
Australia
John F. Dobson

Comment by Mojmir Šob (Brno, Czech Republic)

Impressions from the 4th Psi-k Conference

I have been lucky enough to have a chance to attend all Psi-k Conferences, starting from the first one in 1996 to the fourth one, taking place in Berlin in the middle of September 2010. From the very beginning, these conferences have constituted topical and highly specialized meetings devoted exclusively to ab initio electronic structure calculations (AIESC) and their applications in solid-state physics and chemistry, and in materials science.

I have been working in the field since 1974 and, obviously, have greatly welcomed the first Psi-k meeting in 1996. With the exception of the first Psi-k Conference, I have always taken my whole group with me. At this Conference, we’ve had a chance to assess the present status of our beloved ab initio approach and have observed a continually extending range of its applications. It has been a fantastic achievement of the Psi-k Network and the efforts of the Conference organizers that the 4th Psi-k Conference, now over 3 times larger than the first one (!), has happened.

That large increase in the number of participants indicates unambiguously a steadily increasing interest in AIESC and their applications. I can also confirm this trend from the field of calculating phase diagrams, associated with the CALPHAD Journal. When I attended my first CALPHAD Conference in 2005 in Maastricht (it was the XXXIVth CALPHAD Conference), and had a talk on application of AIESC, the CALPHAD community was largely reluctant to adopt ab initio results in their simulations; the CALPHAD scientists were rather sceptical. Five years later, at the XXXIXth CALPHAD Conference at the Jeju Island in Korea in 2010, about 1/3 of contributions included or touched upon ab initio input.

Of course, such scepticism is not our problem, we do know that AIESC have their great value and provide results at the most fundamental level. Rapid increase of numbers of scientists adopting our belief is a proof that applications of AIESC constitute a correct and promising direction, although one could perhaps do more. As Gerbrand Ceder said in his talk, "$100Ms are poured into materials research and innovation, without fully using the benefits that large-scale computational modelling could offer". Maybe we should intensify our propaganda?

The 4th Psi-k Conference brought together more than 1000 scientists of our ab initio belief. The presentations were distributed among 22 symposia which were running in 5 parallel sessions. Plenary and invited speakers were carefully selected to provide topical reviews of most interesting developments in the field and/or their own results illustrating that development. I really enjoyed all plenary talks which had a very high level indeed. Many oral contributed talks also presented very interesting results. Regrettably, it was not possible for one person to attend all five parallel sessions; all of us had to make our choices.

It was a very good initiative to establish the Volker Heine Award for young investigators. The level of the finalists was high indeed and it was a pleasure to meet them at the closing ceremony, as they were awarded their prizes from Peter Dederichs and Volker Heine himself.

Due to limited time at the conference, most of the contributions had to be presented in poster sessions. At this conference we had two evenings to see in total about 750 posters, and that was really impossible. I wish I had more time for posters. Of course, I do realize that it was technically quite hard to organize poster sessions with such a large amount of contributions. Nevertheless, it was a pity, in my opinion, that the posters could not have been exhibited at least during the first three days of the conference in order to give one more time to inspect them in more detail.

Conference dinner was in a very interesting place, however, we experienced somewhat close-packed arrangement (I was not able to establish whether it was the hcp or fcc or some other type of packing).

This, however, does not diminish the huge amount of work exerted by the Organizing Committee chaired by Matthias Scheffler. They have made a very good job indeed and deserve big applauds from all participants of the 4th Psi-k Conference!

Now, that ESF has endorsed the Psi-k Network as one of its possible programmes for the next five years, subject to securing funding from the National Research Councils of the ESF member countries, we can only wish that this financial support will be granted to fund our future activities, including the next big conference. Let us hope that in 5 years, we shall meet again at the 5th Psi-k Conference. Well, extrapolating from the observed trends, it seems that more than 2000 participants can be expected. It will definitely be a challenge for the future Organizing Committee, and we should wish them good luck!

However, such a large number of participants would bring us to the format comparable with the DFG, APS, MRS or TMS Meetings. Thus, maybe we should also think about establishing an organization like APS, MRS or TMS, called e. g. the Psi-k Society, to bring all the scientists from our community closer together, shouldn't we?

I look forward to attending the 5th Psi-k Conference in 2015!

Sincerely,

Mojmir (Mirek) Šob
Masaryk University
Brno, Czech Republic

Comment by Anton Kozhevnikov (Zürich, Switzerland)

More than a hundred invited talks, hundreds of posters, over a thousand of participants and very intensive personal communication - now we know how the largest conference on electronic structure calculations looks like. Organized by the Psi-k network, the conference took place in Henry Ford Building of the Free University of Berlin during September 12-16. With the absolute certainty we can say that the Psi-k 2010 conference was excellently arranged and conducted. Indeed, five conference halls seated all the interested listeners, there was enough space for the poster session, it was always possible to find a free seat or a table down at the ground floor, internet connection was fast and stable, snacks and drinks were always available and the lunch and dinner meals were really good. The conference brought together many scientists from the electronic structure community, giving everybody a chance to meet new people and charge oneself with new ideas. What else do you need for the productive discussions and intense work? Special thanks must go to the authors of the "participant's bag" with an abstract book, notepad, pen, city map of Berlin and umbrella, which proved to be the most useful item during this four days. The only thing that I would like to wish to the Psi-k network is "Continue in the same spirit!"

With best regards,
Dr. Anton Kozhevnikov
ETH Zürich
Institute for Theoretical Physics
Wolfgang-Pauli-Str. 27
8093 Zürich
SWITZERLAND

Comment by Paweł Buczek (Halle, Germany)

A short account of Psi-k 2010 Conference, Berlin

Paweł Buczek
Max Planck Institute of Microstructure Physics, Weinberg 2, Halle/S., Germany

I must admit that my expectations regarding Psi-k 2010, the first conference of the series I attended, were completely different from reality. I had naively imagined Psi-k as a platform for computational physics craftsmen trying to figure out together the best possible coherent directions for the future numerical electronic theory. Instead I found myself amidst enormous particulate field with everyone following firmly his/her own path. But not-met expectations do not mean disappointment and let me state I enjoyed Psi-k 2010 enormously.

Not one single plenary or semi-plenary presentation disappointed me. The speakers were fully aware they do not have solely specialists in the audience and often managed to throw bridges between sub-fields or even way beyond the computational condensed matter. I have more mixed feelings about the idea of 12 minute long scientific talks, earlier safely constrained to events like German Physical Society spring meetings and recently gloriously making its way to the most of the modern conferences. While I am strongly convinced that it is not the best practice, I admit that most of these short talks were very well prepared and presented. This kind of presentations offers an excellent overview and often gives a rare chance to meet the speaker in the real life.

And this is exactly what Psi-k 2010 was all about for me. On one hand an opportunity for deepening knowledge and polishing numerical approaches, on the other a chance for seeing what and how it is done and how it is sold and presented; a possibility to exchange gossips, to network and to look for collaborators and allies.

I would like to add only few comments to this general praise. The conference could have easily taken one or two more days and ended up with a reduced number of sessions. It was almost impossible to construct any personal program including even most of the relevant presentations. This severe compression was one of the reasons for the low impact of the poster sessions. Many of the posters were of low quality and even more apparently completely unmanned. The lenght of the meeting might be the only serious flaw (together with the drinks one had to pay for after 6pm) in the well organized event. I loved the night in "Rodeo" - it is a great idea to give up official and stiff social dinners; I was relieved to hear there would be no "fun-scientific" talks that night.

I am sure I will not miss the next Psi-k meeting if I am still in the business.

Comment by Marilia J. Caldas (Sao Paulo, Brazil)

Dear Matthias,

I imagine you have already received several messages in the same spirit, so ... one more.

We have all participated in far too many conferences at this time of life, and some of them were really good. I must say yours was the best organized I have ever been to. It made life easy for the students, for which I thank you all very, very much.

Apart from the organization per se, and counting also the quality of the talks and the opportunities for interaction, this last Psi-K was really, for me, one of the best conferences I had the pleasure to attend.

`[ ]s`
Marilia J. Caldas
Grupo Nanomol
Depto de Física dos Materiais e Mecânica Instituto de Física University Sao Paulo, Brazil

Comment by Julie B. Staunton (Warwick, UK)

Ψ_k Conference 2010

September 12 - 16, 2010, Berlin

Julie Staunton, Department of Physics, University of Warwick

Diary of a Conferee.

Monday September 13:
Departmental commitments meant I had to miss first day of conference which is a pity because the programme looks excellent. Eventually arrived in Berlin in the evening - really impressed by Berlin's public transport. Raining..uh oh, travelling light with no waterproofs. Fortunately Hotel Ravenna close to U-Bahn.

Tuesday September 14:
Up bright and early -bumped into several familiar faces in Hotel before catching bus and train to Henry Ford Building. Still raining...Registered and obtained impressively thick abstracts book and programme. (Also acquired large T-shirt and .. fantastic, an umbrella... now that's what I call good organisation). It is striking how big the Conference has grown since its 2000 and 2005 predecessors. There are now over 1000 participants. Must be largest international electronic structure conference - the stats demonstrate this, 5 plenary speakers, 22 symposia with 120+ invited talks covering the whole discipline and its impact on condensed matter and materials science.

David Vanderbilt gave an excellent, thought-provoking plenary talk on the role of Berry phase ideas in theories of magnetoelectric effects in multiferroic insulators. There was then a tough choice as to which of the five symposia to attend. I opted for 'Crystalline, Amorphous and Glassy Alloys' and enjoyed a series of talks on topics on phase stability and disordered systems. Also gave a talk myself. All talks stimulated good discussion. There was some amusement in a couple of the talks when a disembodied voice floated around the lecture theatre. This turned out to be that of someone in another lecture theatre asking a question which our AV system picked up. After the talks and dinner took the opportunity to discuss some work with colleagues and then looked around the posters.

Wednesday September 15:
Another superb plenary talk by Gerbrand Ceder on hot topical high-throughput ab-initio calculations. Decided then to attend parts of 2 very different symposia to appreciate the scope of our conference. To that end chose the 'Earth and Planetary Materials and Matter at Extreme Conditions' on the one hand and 'Organic Electronics' on the other. Later attended the V Heine Young Investigator Award session where all the speakers were very impressive. Throughout the day and the day before I found plenty of opportunities for discussions with other conference participants.

Caught the conference bus to Postfuhramt passing by many famous Berlin landmarks. Good evening where I witnessed an interesting experiment on the collective behaviour of condensed matter physicists at high density where there are competing short- and long-range interactions.

Thursday September 16:
Flight times home meant I had to leave in the morning and miss part of the day. Home in evening - reflecting on a very enjoyable and stimulating conference. Many thanks to all the organisers.

Comment by Warren Pickett (Davis, USA)

Selected Impressions from Psi-k 2010

Warren Pickett

University of California Davis

The brain teeters toward overload when considering how to comment in some useful way on the scientific spectacle of Psi-k 2010. Such a condition is just what one should expect from a four-day period in which 1000 computational electronic structure theorists (”10^3-CEST”) convocate for a bidecadal download, from speaker to audience, of the recent progress of knowing the unknowable. Eighty years ago Dirac pronounced the quantum mechanical study of condensed matter (”chemistry”) too demanding to fathom, i.e. unknowable. The community is, arguably, at a point where it can be said that this statement is Dirac's ”cosmological constant” - a position not withstanding the test of time. Just what should be expected from a meeting of 10^3-CEST would have been guesswork except to the organizers, since it had never happened before in our current universe. I provide just 2-3 selective observations.

Comment from a subjective viewpoint. I attended Psi-k 2010 partially with the American viewpoint in mind, so it was with interest that I noted that Dzidka mentioned I might addresss the American viewpoint, after which I might segue into more general comments. My specific ”American viewpoint” in this context in fact arose from a recent Psi-k Newsletter in which Peter Dederichs pointed out that, in terms of DFT-based publications in journals followed by the Web of Science, the U.S. is in the process of being left in the dust. The U.S. is behind Europe in terms of publications per year and gap is increasing with time. In addition, the U.S. will be overtaken by Asia within a couple of years. Growth in China is at least quadratic (Europe and U.S. growth is linear), so China may overtake the U.S. in such publications before it overtakes it in economic terms. These trends are the kind of messages that may stimulate action in some of the right places (funding offices) if one can attract their attention.

There is widespread belief, probably in Europe but certainly in the U.S., that the origin lies in some basic sociological differences in the funding, indeed in the activity, in the two geographical and geopolitical areas. A central feature – a very evident distinction – is the development and support of robust, user-friendly, open source, supported DFT codes in Europe. Austria and Germany have contributed several. At the risk of upsetting some by being (necessarily) incomplete, I can mention also as active contributors groups in Italy, Belgium, the U.K., Spain, Sweden. I am not totally unaware of the efforts in Japan, where they have codes as well, though I am much less familiar with them.

The modus operandi in the U.S. has been, for some decades, to distribute a large fraction of research funding in large part through teams: from MRSECS to EFRCs (what these acronyms stand for is not so important here). A given professor or national lab leader typically gets research funding from a few directions and works in a few different (albeit related) areas, for three years at a time. Funding from different sources should basically fund distinguishable projects. In Europe the infrastructure culture is different. There are (to overstate the case, but only mildly I hope) professors or single-institution tightly knit groups with long-term funding who can think and act with the longer timescale in mind. The result has been (many) of the aforementioned DFT codes. The U.S. has some quantum chemistry codes but no DFT codes whose acronym would be identifiable to most Europeans (unlike the other way around).

The availability of these DFT codes, and the scientific environment that surrounds them, seem to many of us in the U.S. to be a substantial part of the stimulus that has propelled Europe into their current leadership role in our area of research, with 10^3-CEST at Psi-k 2010 held in Berlin and not in (say) Baltimore. The U.S. does have its "Electronic Structure Methods" annual conference, more than 20 years in the running now, but it has still the challenge (if it aspires to do so) to attract the wider electronic structure community attendance in the U.S. that Psi-k does in Europe. Commendably, East Asian countries also hold a regular electronic structure conference.

Broad observations. I have digressed. The subject of this comment is Psi-k 2010, and the impression it leaves and, possibly, its impact. The foment (which is ferment, without the alcohol) in our field is palpable; new ideas and the implementation to test them continues at a scary rate. Scary, if you have delicate disposition, but in such a vigorous research environment one should think in terms of boldness. We refer to our approach as 'first principles' but principles often give way to practicalities with the justification to be tightened up later, with finding 'what works' being a profitable way forward. We are, after all, working to know the unknowable.

Some of our Bloch functions have been concealing their intimate private behavior (phase twists) until recently. The bulk electronic structure of topological insulators hosts an entanglement that must be straightened out at the surface, so such insulators must have surface bands through the gap that disentangle the phase warp. (Is it possible there is an analogous phase twist in some insulating surfaces that may lead to yet richer behavior? Sounds only fair.) The excellent plenary exposition was followed by a number of computational investigations, in this field which is only in its infancy. The entanglement may be more subtle than the proofs of its occurrence and of its consequences. The effect requires both spin=orbit coupling and multiple bands. Band structure practitioners understand that spin-orbit coupling introduces phases (complex numbers) into the treatment, arising rudely due to its order of magnitude increase in computational cost (complex arithmetic versus scalar; doubling of the size of the secular equation, if the Kramers degeneracy is not taken into account to counteract the loss of symmetry [distinct spins]). A more intuitive understanding of the (gauge-invariant) Berry curvature is desirable, and the early investigations to do so that were presented provide confidence that this understanding will proceed forthwith.

Comment on part of the future of "psi-k". Note: I use psi-k here to refer not to the bidecadal conference which should have 1600 attendees in 2015, not to the organization, but to the computational materials theory community that drives the advances, applies them, and furthers the understanding of materials behavior from the first principles viewpoint. One of the notable advances that is evident from this conference is the impressive extension in the treatment of dynamic electronic behavior.

The continued progress, and several extensions and new applications, in the description of electron dynamics was discussed several symposia (Electronic Excitations; Dynamical Mean Field Theory; Strong Correlations from First Principles; Solar Energy Conversion and Harvesting; Organic Electronics). In rough historical order, the primary topics were GW, tdDFT, and DMFT. To one who has not kept up with many of the applications of tdDFT (me), the progress seems impressive indeed.

The GW approach has been around for some time and continues to advance. At the newer end, dynamical mean field theory is showing increasing promise. The impressive advances at this meeting from my viewpoint were from the very significant inroads that time-dependent DFT (tdDFT) is achieving. This progress illustrates a crucial aspect of DFT: the ansatz is an energy functional with clear formal foundation. LDA (and its immediate offspring) has been enormously successful in spite of a simplistic approximation (extreme locality, whereas exchange and correlation are not really so). Such success can be possible only because the formulation is correct: a (rigorously justified) density functional (theory) is formulated, which is then approximated (necessarily so), and minimized (via the Kohn-Sham approach) to provide the many results that are obtained. The GW approach follows the (many body) Hamiltonian formalism, with energetics evaluated (via the Galitskii-Migdal expression) only extremely rarely, and not yet approaching any application of linear response methods.

The tdDFT approach is (from my personal view) unexpectedly successful, apparently due to its formal basis and application in terms of a well-formulated energy functional. A time-dependent Hamiltonian alters the emphasis of the theory (away from non-conserved energetics), but the successes build on the formulation. Extending predictive computational theory to strongly correlated materials will surely require treatment of dynamical processes, with the dynamical mean field theory (DMFT) approach being the most widely used a present. Significant further development of DMFT will, in my view, necessitate focus on the functional (thermodynamic grand potential).

A breakthrough in this area was made around the turn of the 21st century when Andy McMahan, Richard Scaletter, and Karsten Held devised a DMFT energy functional in an all-electron LDA+DMFT implementation to model the volume collapse transtion in Ce under pressure. Their .Kondo volume collapse. result remains the most convincing microscopic explanation of this classic problem in correlated electron physics. Their basic functional form has been used occasionally in the subsequent decade with some success. Further progress, almost certainly, relies on the formulation of the functional, necessarily approximated, then minimized, generating the single particle Green's function and related results (free energy hence thermodynamics; linear response quantities). Obviously full charge self-consistency is essential; advances in accomplishing this is underway in a few groups. This are of research is well positioned to form one of the more exciting symposia at Psi-k 2015.

This highly successful meeting has been a great reminder of how our community has converted the great promise of the 1970s and 1980s into the tremendous success story that it now is. All signs point toward a continuing bright future as more and more materials properties are understood from a first principles viewpoint.

Comment by Samir Lounis (Irvine, USA)

Report on the Ψ_k-2010 Conference

Samir Lounis

Department of Physics and Astronomy
University of California Irvine, California, 92697 USA

The Ψ_k conference is a traditional meeting that covers theoretical and computational research on electronic structure and properties of matter. It takes place every 5 years. This time it has been organized in Berlin where sufficient space is available to host the huge attending community (more than 1000 participants).

The overall organization of the conference was well handled: from the initial stage including registration and submission of scientific contributions to the final stage of talks and posters. Five plenary talks were given and several symposia were organized covering a large number of topics where certainly every participant, even if not expert, could benefit from. It occurred that sometimes, interesting sessions were scheduled at the same time making difficult my personal choices. As an example, I was personally interested in magnetism and strongly correlated systems on Monday while during the last day there were two interesting sessions on the same topic: Excitations (Quantum Dynamics and Electronic Excitations). There was definitely a plenty of choice for the different audiences.

Most of the talks were invited accompanied by contributed presentations and four poster sessions. The latter ones were organized during the evenings of the first two days, giving exposure to interesting and original works. These evening sessions were, in my opinion, very convivial and allowed me, probably other participants as well, great opportunities to exchange and discuss new and old ideas.

I could not attend all talks and cannot report on all presentations I have seen. Thus, I will mention briefly the plenary talks which were very educational. The first plenary speaker, Stefano Baroni, discussed the basic concepts of density functional perturbation theory, the Liouville-Lanczos approach to time-dependent density functional theory and several applications obtained with the quantum-espresso code. David Vanderbilt reviewed the Berry phase and Berry curvature and their crucial role in the theories of electric polarization, orbital magnetization and anomalous Hall effect. To efficiently synthesize fuels from sunlight, new catalysts are required. Jens Norskov introduced and discussed some of the challenges to catalyst discovery and how first-principles calculations can help in designing them. Gerbrand Ceder presented the Material Genome Project that has the objective to perform large scale ab-initio property prediction of all known inorganic materials. Several examples of new compounds were investigated and different short-coming of the usual approximations in the exchange and correlation functionals were mentioned. Mark Ratner discussed transport in molecular junctions and demonstrated by different examples and limiting cases different approaches to tackle this non-trivial problem.

To conclude, I believe that the Ψ_k-2010 conference was a success and I am looking forward to the next one.

Comment by Manuel dos Santos Dias (Warwick, UK)

Psi–k conference report

My name is Manuel dos Santos Dias, and I'm a PhD student under Prof. Julie Staunton at the University of Warwick, working in collaboration with Prof. László Szunyogh of the Budapest University of Technology and Economics. My research and main interests are on nanoscale magnetism. I present my impressions on the Psi–k 2010 Conference in a few paragraphs below.

The scientific program was outstanding. With often three sessions in parallel drawing my attention, it was hard to decide which talks to attend. Moving from one session to a different one was hindered by a somewhat fluid enforcement of the alloted times for each talk. In the first two days the program was quite long, and the poster sessions took place afterwards, in the evening, taking a toll on the people presenting a poster and on those wanting to discuss the posters with their authors — I was part of both groups. The breadth and depth of the research presented was truly exceptional, and of great interest and relevance. To me this was very fruitful.

I wish to highlight a few talks that I found particularly interesting. Prof. Stefan Blügel's talk, 'Topologically protected spin textures at metal surfaces', was an excellent summary of the physical mechanisms giving rise to complex magnetic states in ultrathin films, as unveiled experimentally and theoretically.

On the topic of strong correlations, Prof Zhong Fang's talk, 'LDA+Gutzwiller method for strongly correlated systems', was an interesting addition to the existing manifold of methods and techniques created to address this problem.

As I'm a computational physicist, Prof. Thomas Schultess talk, 'Petascale computing in condensed matter physics', was a very compelling review of the development and usage of large scale simulations, even if peppered with an amusing technical incident.

The Volker Heine session was an insightful addition to the conference program. The talk by Dr. Samir Lounis, 'Investigations of the intriguing magnetic and electronic behaviour at the nanoscale', was a vivid illustration of how distinct systems with a finite number of atoms are.

The last talk I will mention was given by Dr. Pawel Buczek, 'Spin dynamics of complex metallic magnets'. He presented a theory of the transverse magnetic susceptibility which incorporates both local moment and itinerant electron magnetism on the same footing, thus reconciling the Stoner and Heisenberg pictures, with very interesting applications.

The Henry Ford building was an excellent choice for the conference venue. Its ample interior and abundant natural lighting made sharing the same space with 1000 people an almost pleasant experience. The lecture halls are comfortable and well equiped, and it was easy to change from one room to another. The conference staff was very helpful. There was some inevitable strife during the coffee breaks, and some slices of cake became mythological foodstuffs, as I seldom got hold of them. The main meals were also adequate.

I close by commending all the speakers, participants and organisers on making such a good conference possible.

Comment by Hisazumi Akai (Osaka, Japan)

Short report on the Psi-k 2010 Conference in Berlin

Hisazumi Akai

Osaka University, Japan

The number of submitted papers as well as the number of participants at the Psi-k 2010 conference is clear proof of the tremendous vitality of the field of first principles calculations. The plenary and invited talks were extremely stimulating. Moreover, the contributed oral presentations were also uniformly excellent. Indeed, after attending the poster sessions, I came to the conclusion that the program committee faced an almost impossible task in selecting contributions for oral presentation, such was the high standard.

First-principles calculation now seems to be a field that is dominated by techniques such as NMR, photo-emission, STM, and so on. Many authors reported the results of analyses using a reliable and easy-to-use program package (in many cases, VASP). While there is no question that this is a scientifically sound approach, I hope in the future to see more contributions reporting efforts to further develop the fundamental theories and methodologies that are the foundation of first-principles electronic structure calculations. This, of course, probably reflects a general tendency in the field of first-principles electronic structure calculations.

The Psi-k 2010 conference was a great success and I would like to congratulate the organizers, the contributors and all those who attended, for making it so.

Comment by Peter Kluepfel (Reyjavik, Iceland)

Extraordinary events with global impact are kind of daily business here in Iceland. So when I marked the PsiK2010, the "biggest electronic structure conference in the world" in my calender I thought it was funny appearing right next to volcanic eruptions and the collapse of a nation's financial system and the superlative "biggest" lost some of its magic in this context. But I was about to be taught differently:

The Organizers:

Everyone knows that Germans are well-organized. Thus I was not expecting anything else than a conference running smoothly from welcome reception to closing session. But I was impressed by the way the organizers managed to keep things as simple and informal as possible. Already the short waiting time and the relaxed atmosphere at the welcome reception made it easy to find time and space to get connected to old friends and collaborators. Short distances between the lecture halls allowed for getting the most of the contributions without the typical "I will stay here in room X, just because I anyway won't make it in time to the talk in room Y"-feeling. I don't want to forget about expressing my appreciation and thanks to the numerous helpers from the FHI and elsewhere. It felt like there was one of them for each participant.

The Contributions:

PsiK2010 offered one of the finest selections of key-note speakers. This already became clear from the first announcements and the conference program. But the plenary talks even got eclipsed by the numerous invited and contributed presentations. It was impressive to see that there is a new generation of highly motivated, intelligent and charismatic scientists propagating electronic structure theory into the 21st century. One of the few negative aspects of the conference is related to the poster sessions which was in my opinion too short. It was impossible to get through all the interesting presentations or to have deeper discussions with all visitors to ones own poster.

Coffee, Cocktails and Cuisine:

An often underrated descriptor for a good conference is given by the quality of food, the permanent availability of coffee and the amount of free alcoholic beverages at the conference outing. But probably nobody had reason to complain about any of those aspects at the PsiK2010. Enjoying the delicious meals and the excellent catering service on site gave enough energy for the afternoon sessions. I was amazed what seems to be considered as "finger-food" in Berlin, but still also the food at the outing was very tasty.

I'm personally not a friend of big conferences and usually prefer the atmosphere and efficiency of smaller workshops or summer schools. But in case of the PsiK2010 I honestly have to admit that in spite of its extraordinary dimensions it still maintained the character of a small work meeting with plenty of opportunities for scientific exchange, planning of future projects or just chats about the good old times.

I'm looking forward to the next years' PsiK workshops and hopefully we will all meet again at PsiK2015.

Peter Kluepfel,

Chemistry Department, Science Institute,
University of Iceland
Dunhaga 3, 107 Reyjavik, Iceland

Comment by Angela Klautau (Belém - PA, Brazil)

The Psi-k Conference 2010 confirmed the well-known general opinion about the importance of *ab initio* calculations for many different fields of natural sciences. The huge number of participants, ranging from Ph.D. students to senior researchers, representing many countries, confirms the increasing importance of electronic structure theory and calculations. The Conference topics ranged from Magnetism and Spintronics to Biological Systems, from Superconductivity to Solar Energy Conversion, from Organic Electronics to Multiferroics and Oxides, etc. In the Conference Symposiums we heard and discussed about state of the art computer codes for calculations of different phenomena in different classes of systems, and implementations of new ideas for incorporating correlation effects beyond DFT. In this Conference we had the possibility to attend to seminars and discuss at posters sessions: we listened to senior researchers, who have contributed to the development of the electronic structure area, and learned from groups which have developed and continue working on the improvement of computational methods in this area. It was also an opportunity for listening to very bright young researchers, who contribute to our area in developing and applying computational quantum physics to deepen our understanding of materials. The Conference represented an efficient and unique opportunity to exchange and disseminate information among the computational material science community, giving high visibility to research progresses, as well as to get collaborations started.

Angela Burlamaqui Klautau[1,2]
e-mail: aklautau@ufpa.br
[1] *Falcudade de Física, Universidade Federal do Pará, Belém - PA, Brazil*
[2] *(on behalf of the Nanomol Group, http://nanomol.if.usp.br)*

2.3 Report on Psi-k Volker Heine Young Investigator Award

The Young Investigator Award of Psi-k was announced at the beginning of this year. All young scientists in our field, who had obtained a Ph. D. within the five years preceding the announcement could apply. In total 23 applications were received before the deadline of 17th April 2010. The award committee consisted of four Psi-k Trustees and three outstanding scientists from USA and Japan:

Volker Heine, Cambridge University
Matthias Scheffler, Fritz Haber Institute
Angel Rubio, University Pais Vasco
Peter Dederichs, Research Center Juelich
Marvin Cohen, UC Berkeley ,USA

Gerbrand Ceder, MIT, USA
Ferdi Aryasetiawan, Chiba University, Japan

The award committee had to select five finalists from the 23 applications who were to present their work at the Award Session of the Psi-k 2010 Conference. This selection turned out to be difficult, since there were about a dozen excellent proposals, which deserved a careful review. The selected five finalists were:

Claudio Attaccalite, Institute Neel, Grenoble
Christoph Freysoldt, MPI Iron Research, Duesseldorf
Mateo Gatti, ETSF-UPV, San Sebastian
Samir Lounis, UC Irvine and Research Center Juelich
Alexandre Tkatchenko, Fritz-Haber Institut, Berlin

They presented their work in the special Award Session on Wednesday afternoon of the Psi-k 2010 Conference. The session was very well attended and all candidates gave very good talks.

Thus the award committee, chaired by Marvin Cohen, had again a difficult choice. Special care was taken that the committee members, who had a conflict of interest, did not vote for .their . candidates. The final vote was close. The winner of the Psi-k Volker Heine Young Investigator Award was

Christoph Freysoldt, MPI Iron Research, Duesseldorf.

The Psi-k Chairman presented to him the award certificate and Volker Heine the cheque of 2500 Euro. Also the other finalists obtained each an award certificate as "Finalist of the Psi-k Volker Heine Young Investigator Award" and a cheque of 500 Euro. In the photographs below, we see both the winner and the whole group of finalists, respectively, in the company of Peter Dederichs and Volker Heine.

In summary, we consider the new Volker Heine Award as very successful. It was great to see that there were very many talented young researchers in our community. For this reason we intend to issue the prize also in the future.

Peter H. Dederichs, Psi-k Chairman

3 General Workshop/Conference Announcements

3.1 Joint NNIN & NCN Fall Workshop: Building a Collaborative Framework for Nanoscale Simulations

Cornell Nanoscale Facility, Ithaca, NY

November 15th-16th, 2010

Organized by
National Nanotechnology Infrastructure Network (NNIN/C)
&
Network for Computational Nanotechnology (NCN)

Sponsored by the National Science Foundation

Workshop Coordinator: Derek Stewart (stewart@cnf.cornell.edu)

`http://www.cnf.cornell.edu/cnf_fallworkshop2010.html`

Overview: Science, in the words of Isaac Newton, has always relied on current researchers advancing the field by *standing on the shoulders of giants*. Modern science rests not only on the easy communication of ideas via publications, but also on the transfer and analysis of vast stores of data from both experiments and simulations. Nanoscience also relies on extensive atomistic simulations that generate copious amounts of data. Since various *ab-initio* and empirical approaches exist in the field, there is a growing need to develop a **common collaborative framework** to enable meaningful comparisons between calculations and to also provide a set of robust technical components and results for the next generation of researchers.

This workshop will bring together researchers in atomistic simulations to help develop a set of *lingua franca* formats and libraries for easy translation of input and output files between codes. We will address the creation of international cyberinfrastructure resources that deliver trusted components such as pseudopotentials, basis sets, empirical potentials, and atomic coordinates.

Workshop Topics

- Standing on the Shoulders of Giants: Online Input File Databases & Common IO File Formats
- Trusted Components for Calculations (Pseudopotentials, Basis Sets, Empirical Potentials, Crystallographic Data)

- In the Trenches: An Overview of Current Efforts in the Field
- Fully Reproducible Science (linking calculations with publications)

Registration Deadline: October 31st, 2010
Seating is limited to 40 participants, please register early!

3.2 Meeting on Optical Response in Extended Systems MORE 2010

Vienna, Austria

November 3-5, 2010

supported by the Austrian ministry of science

organized by the association
Physicae et Chimicae Solidorum Amici

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

"MORE of LESS".
MORE 2010 follows the spirit of the highly successful Low Energy Spectrometry Symposiums (LESS 2007 and 2009) as well as MORE 2008 that took place in Vienna.

Optical absorption spectroscopy, resonant inelastic X-ray scattering, reflection EELS, and RAMAN 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.

Main intention of MORE 2010 is bringing together experimentalists and theoreticians of these different fields and to form a platform for leading experts to discuss and exchange their recent results and scientific concepts. 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.

The AMICI Young Scientist Support Fund (A.Y.S.S.) offers five grants of EUR 400,– to support young scientists. Selected applicants will get the opportunity to present the results of their work as a contributed talk (30 minutes including discussions). Young scientists are invited to apply for these grants by sending their CV, a short motivation letter, and an abstract for their presentation to amici@physics.at.

Invited Speakers (confirmed):

- **A. Alkauskas** (EPFL, Lausanne, Switzerland)
- **J. Braun** (LMU Munich, Germany)
- **F. J. Garcia de Abajo** (CSIC Madrid, Spain)
- **R. Hackl** (LMU Munich, Germany)
- **M. Kociak** (CNRS-Orsay, France)
- **M. A. L. Marques** (Université Lyon 1 et CNRS, France)
- **P. Oppeneer** (University of Uppsala, Sweden)
- **T. Pichler** (University of Vienna, Austria)
- **J. J. Rehr** (University of Washington, USA)
- **P. Rinke** (FHI Berlin, Germany)
- **G. Schütz** (MPI f. Metallforschung, Stuttgart, Germany)
- **E. Sherman** (University of the Basque Country, Spain)
- **W. Sigle** (MPI f. Metallforschung, Stuttgart, Germany)
- **J. Verbeeck** (Univ. of Antwerp, Belgium)

Venue:

MORE 2010 is hosted 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 available at http://www.physics.at/.
The final deadline for registration is October 15, 2010.

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:

K. Hummer (University of Vienna, Austria)

J. Luitz (luitz.com, Austria)

C. Ambrosch-Draxl (MU Leoben, Austria)

P. Schattschneider (TU Vienna, Austria)

Programme:

Wednesday, November 3

13:00 – 14:00	Registration and Refreshments
14:00 – 14:15	Opening
14:15 – 15:00	**Mathieu Kociak** *Nanoplasmonics and nanophotonics with electrons*
15:00 – 15:45	**Javier Garcia de Abajo** *Interaction of electrons with plasmons and evanescent light fields*
15:45 – 16:15	Coffee break
16:15 – 16:45	**Contributed talk**
16:45 – 17:30	**Wilfried Sigle** *Some new results of plasmon research in Stuttgart*
17:30 –	Aperitif

Thursday, November 4

09:00 – 09:45	**John J. Rehr** *Broad spectrum calculations of X-ray and electron spectra*
09:45 – 10:30	**Gisela Schütz** *to be announced*
10:30 – 11:00	Coffee break
11:00 – 11:45	**Miguel A. L. Marques** *The band-structure of delafossite transparent conductive oxides*
11:45 – 12:15	**Contributed talk**

12:15 – 14:15	Lunch
14:15 – 15:00	**Thomas Pichler** *Electronic properties and nature of the metallic ground state in pristine and interca*
15:00 – 15:30	**Contributed talk**
15:30 – 16:00	Coffee break
16:00 – 16:45	**Johan Verbeeck** *Electrons with a twist: production and application of electron vortex beams*
16:45 – 17:30	**Contributed talk**
17:30 – 18:30	**Poster session**
18:30 –	Dinner

**Friday,
November 5**

09:00 – 09:45	**Rudi Hackl** *Light scattering in unconventional metals and superconductors*
09:45 – 10:30	**Evgeny Sherman** *Raman scattering in solids from first principles*
10:30 – 11:00	Coffee break
11:00 – 11:45	**Jürgen Braun** *Recent developments in the theory of ARPES: Correlation, disorder and temperatu*
11:45 – 12:30	**Peter Oppeneer** *Theory of magnetic spectroscopy: The X-ray regime and towards femtosecond lase*
12:30 – 14:30	Lunch
14:30 – 15:15	**Audrius Alkauskas** *What do we learn from dynamic structure factors of metals*
15:15 – 16:00	**Patrick Rinke** *When many-body matters: From the f-electron challenge to F-centers in MgO*
16:00 –	Closing

3.3 First Euro-Mediterranean Meeting on Functionalized Materials EMM- FM 2011

March 17-21, 2011, Sousse, TUNISIA

CALL FOR PAPERS

Submission deadline: November 18, 2010

It is the first "Euro-Mediterranean Meeting on Functionalized Materials". This first conference of a new cycle of conferences involving Solid State Physics, Material Science and Chemistry including applications at all scales should offer a forum where the scientists of all the Mediterranean countries or regions and over the world will have the opportunity to renew old friendships and network with new contacts, have friendly discussions and get to know each other better.

The topics of the conference cover several aspects of functionalized materials (magnetism, superconductors, dielectric materials, ferroelectricity, multiferroic materials, biomaterials, materials for energy, characterization methods).

Prof. Albert Fert, the Nobel Prize in Physics 2007 accepted to attend this meeting and will deliver the first plenary talk during the scientific session on Friday morning, March 18-2011.

Our meeting will be preceded by a spring school (March 15-17, 2011) and followed by a three days tours in the south of Tunisia .

Besides this tour, a rich social program during the Conference will be organized and specially for accompanying persons. You will surely enjoy the vibrant culture and many points of interest in our beautiful and hospitable country Tunisia .

Please visit our website

`http://www.tu-mrs.org/emm-fm2011`

for more details about EMM-FM 2011.

Please do not hesitate to contact us at emmfm2011@tu-mrs.org for any inquiry.

Sousse is known as "the pearl of the Sahel" and located on the eastern coast of Tunisia , two hours from the capital - Tunis - in the central-east of the country.

The Organizers,

Bernard Barbara and Abdelwaheb Cheikhrouhou

Contact details:

Prof. Abdelwaheb CHEIKHROUHOU
Faculty of Sciences of Sfax
B.P. 1171, 3000 Sfax , Tunisia
Tel-Fax: 00216 74676607
Mobile: 00216 98656481; 00216 25656481
E-mail: abdcheikhrouhou@yahoo.fr

Dr. Bernard BARBARA
Institut Nel, CNRS
B.P. 166, 38042 Grenoble , France
Tel: 0033(0)4 76725353, Mobile 00(0)6 71701704
E-mail: bernard.barbara@grenoble.cnrs.fr

Conference Secretariat:

Ben ghozlen Afifa; Boughariou Sana
Faculty of Sciences of Sfax
B.P. 1171, 3000 Sfax , Tunisia
E-mail: emmfm2011@tu-mrs.org

4 General Job Announcements

Postdoctoral Position in Computational/Theoretical Chemistry or Solid State Physics (Band Structure)

State University of New York at Buffalo, USA

A position for a postdoctoral researcher in computational/theoretical chemistry will be available in the group of Eva Zurek. Our research interests are broad and currently we are focussing on the electronic structure, properties, and reactivity of: electrides and solvated electrons, carbon nanostructures, gold clusters, and solids under high pressure. We are also involved in developing evolutionary algorithms for predicting the structures of solids and molecules. For further information about our research, please visit our website:
http://www.acsu.buffalo.edu/∼ezurek/ .

The starting date is flexible, however sometime around Jan 2011 would be preferred. The initial contract will be for one year, with a possible extension for another year upon mutual agreement.

A PhD in computational chemistry, solid state physics, or a closely related area is required. I am looking for a creative individual whose strength lies in the application of molecular and periodic program packages (first principles, DFT and others). This particular project will focus on using evolutionary algorithms developed in our group to predict the structures of extended systems with novel stoichiometries. Good communication and writing skills in English are required.

Buffalo is a lively city located in the western part of the State of New York, USA, near the Canadian border. The greater Buffalo area has a population of about a million. There are many local attractions, galleries and museums, annual events and festivals, and recreational sites within easy reach. E.g. the nearest skiing resorts as well as the Niagara Falls are reached by car within 30 to 45 minutes. During the winter we are blessed with descent amounts of good quality snow for winter sports. Across the Canadian border, Toronto is close enough for a day trip.

If you wish to apply for the position please email your resume to ezurek@buffalo.edu. If it appears that we have compatible research interests I will subsequently ask you to arrange for two letters of recommendation from current and former research supervisors to be emailed to me.

Post Doctoral/Research Associate Positions in NREL's Solid State Theory Group

with Alex Zunger

`http://www.sst.nrel.gov`

NREL's Solid State Theory group is looking to fill Postdoctoral/Research Associate positions (depending on qualifications and experience). We are looking for candidates in two areas:

(i) Inverse Design of Materials, addressing the search and discovery of new materials for renewable energy, and

(ii) Atomistic Theory of Quantum Semiconductor Structures - electronic and optical properties. These positions are affiliated with the DOE's "Energy Frontier Research Center" (EFRC) at NREL.

The positions are made initially for one year, and renewable upon mutual consent for up to 3 years. The position is available at present. The salary range is $48,000 - $62,000 per year, depending on seniority, qualification and experience. Applicants are expected to have experience in electronic structure methods and a strong background in general solid-state theory.

Interested candidates should send now their curriculum vitae, list of publications (including preprints of unpublished papers, if possible), and arrange for two to three references addressed to:

Alex Zunger, M/S 3213
Solid State Theory Group
National Renewable Energy Laboratory
1617 Cole Boulevard
Golden, Colorado 80401

Clarification or further details can be obtained via email to `alex_zunger@nrel.gov`.

The Solid State Theory Group is headed by Alex Zunger and currently consists of ten PhD's in condensed matter theory and interacts with a broad range of experimentalists. The group has outstanding computational facilities, an excellent basic-research atmosphere, and is located in the beautiful Rocky Mountains. Consult our web page for additional information on the group, its history, research subjects, publications, current and past personnel and facilities.

NREL is an equal opportunity employer and proud of its commitment to diversity. Women and minorities are encouraged to apply.

5 Abstracts

Superconductivity in SnO: a Nonmagnetic Analogue to Fe-based Superconductors?

M. K. Forthaus[1], K. Sengupta[1], O. Heyer[1],
N. E. Christensen[2], A. Svane[2], K. Syassen[3]
D. I. Khomskii[1], T. Lorenz[1], and M. M. Abd-Elmeguid[1]
[1]*II. Physikalisches Institut, Universität zu Köln,*
Zülpicher Str. 77, D-50937 Köln, Germany
[2]*Department of Physics and Astronomy, Aarhus University,*
Ny Munkegade 120, DK-8000 Aarhus C, Denmark
[3]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1,*
D-70569 Stuttgart, Germany

Abstract

We found that under pressure SnO with α-PbO structure, the same structure as many Fe-based superconductors, e.g. β-FeSe, undergoes a transition to a superconducting state for $p \geq 6$ GPa with a maximum T_c of 1.4 K at $p = 9.3$ GPa. The pressure dependence of T_c reveals a dome-like shape and superconductivity disappears for $p \geq 16$ GPa. It is further shown from band structure calculations that SnO under pressure exhibits a Fermi surface topology similar to that reported for some Fe-based superconductors and that the nesting between the hole and electron pockets correlates with the change of T_c as a function of pressure.

(Phys. Rev. Letters, to appear)
Manuscript available from: svane@phys.au.dk

Quasiparticle self-consistent GW theory of III-V nitride semiconductors: bands, gap bowing and effective masses

A. Svane[1], N. E. Christensen[1], I. Gorczyca[2], M. van Schilfgaarde,[3],
A. N. Chantis[4], and T. Kotani[5]
[1]*Department of Physics and Astronomy, Aarhus University,*
Ny Munkegade 120, DK-8000 Aarhus C, Denmark
[2]*High Pressure Research Center, Polish Academy of Sciences, Warsaw, Poland,*
[3]*School of Materials, Arizona State University, Tempe, Arizona, 85287-6006, USA,*
[4]*Theoretical Division, Los Alamos National Laboratory,*
Los Alamos, New Mexico, 87545, USA,
[5]*Department of Applied Physics and Mathematics, Tottori University,*
Tottori 680-8552, Japan

Abstract

The electronic band structures of InN, GaN and a hypothetical ordered $InGaN_2$ compound, all in the wurtzite crystal structure, are calculated using the quasiparticle self-consistent GW approximation. This approach leads to band gaps which are significantly improved compared to gaps calculated on the basis of the local approximation to density functional theory, although generally overestimated by 0.2 - 0.3 eV in comparison with experimental gap values. Details of the electronic energies and the effective masses including their pressure dependence are compared with available experimental information. The band gap of $InGaN_2$ is considerably smaller than what would be expected by interpolation implying a significant band gap bowing in InGaN alloys.

(Phys. Rev. B 82, 115102 (2010))
Manuscript available from: svane@phys.au.dk

Coexistence of different charge states in Ta-doped monoclinic HfO_2, Theoretical and experimental approaches

M. A. Taylor[1], R. E. Alonso[1], L. A. Errico[1,2],
A. Lopez-Garcia[1], P. de la Presa[3], A. Svane[4], and N. E. Christensen[4]
[1]*Departemento de Fisica & Instituto de Fisica La Plata,*
(IFLP, CCT-La Plata, CONICET-UNLP),
Facultad de Ciencias Exactas, Universidad Nacional de La Plata,
Casilla de Coreo 67, 1900 La Plata, Argentina
[2]*Universidad Nacional del Nordeste Bonaerense (UNNOBA),*
Monteagudo 2772, Pergamino, CP 2700, Buenos Aires, Argentina
[3]*Instituto de Magnetismo Applicado, UCM-ADIF-CSIC,*
PO Box 155, 28230 Las Rozas, Spain
[4]*Department of Physics and Astronomy, Aarhus University,*
Ny Munkegade 120, DK-8000 Aarhus C, Denmark

Abstract

A combination of experiments and *ab-initio* quantum mechanical calculations has been applied to examine hyperfine interactions in Ta-doped hafnium dioxide. Although the properties of monoclinic HfO_2 have been the subject of several earlier studies, some aspects remain open. In particular, Time Differential Perturbed Angular Correlation (TDPAC) spectroscopy studies using ^{181}Ta as probe atom revealed the coexistence of two hyperfine interactions in this material, but an explanation was only given for the more populated one. Until now, no models have been proposed that explain the second interaction, and it has not yet been associated with a specific crystallographic site. In this work, a detailed study of the different charge states for the impurity-probe atom (Ta) was performed in order to understand the second interaction observed in Ta-doped monoclinic HfO_2. The combination of experiments and theory suggests that two different charge states coexist in this compound. Further, *ab-initio* calculations predict that, depending on the impurity charge state, a sizeable magnetic moment can be induced at the probe site. This is confirmed by a new analysis of experimental data.

(submitted to Phys. Rev. B)
Manuscript available from: svane@phys.au.dk

Band parameters and strain effects in ZnO and group-III nitrides

Q. Yan[1], P. Rinke[1,2], M. Winkelnkemper[3], A. Qteish[4], D. Bimberg[3],
M. Scheffler[1,2], and C. G. Van de Walle[1]
[1]*Materials Department, University of California, Santa Barbara,*
California 93106-5050, USA
[2]*Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany
[3]*Institut für Festkörperphysik, Technische Universität Berlin,*
Hardenbergstrasse 36, D-10623 Berlin, Germany
[4]*Department of Physics, Yarmouk University,*
21163-Irbid, Jordan

Abstract

We present consistent sets of band parameters (including band gaps, crystal-field splittings, effective masses, Luttinger, and E_P parameters) for AlN, GaN, InN, and ZnO in the wurtzite phase. For band-energy differences we observe a pronounced nonlinear dependence on strain. Consistent and complete sets of deformation potentials are then derived for realistic strain conditions in the linear regime around the experimental equilibrium volume. To overcome the limitations of density-functional theory (DFT) in the local-density or generalized- gradient approximations (LDA and GGA) we employ the Heyd-Scuseria-Ernzerhof (HSE) hybrid functional as well as exact exchange (OEPx) based quasi-particle energy calculations in the G_0W_0 approach.

(Semicond. Sci. Technol., accepted (June 15, 2010))
Contact person: Patrick Rinke (rinke@fhi-berlin.mpg.de)

First-principles modeling of localized d states with the GW@LDA+U approach

H. Jiang[1], R. I. Gomez-Abal[2], P. Rinke[2], and M. Scheffler[2]
[1] *Beijing National Laboratory for Molecular Sciences,*
National Laboratory of Rare Earth Material Chemistry and Application,
Institute of Theoretical and Computational Chemistry,
College of Chemistry and Molecular Engineering,
Peking University, 100871 Beijing, China
[2] *Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany

Abstract

First-principles modeling of systems with localized d states is currently a great challenge in condensed-matter physics. Density-functional theory in the standard local-density approximation (LDA) proves to be problematic. This can be partly overcome by including local Hubbard U corrections (LDA+U) but itinerant states are still treated on the LDA level. Many-body perturbation theory in the GW approach offers both a quasiparticle perspective (appropriate for itinerant states) and an exact treatment of exchange (appropriate for localized states), and is therefore promising for these systems. LDA+U has previously been viewed as an approximate GW scheme.We present here a derivation that is simpler and more general, starting from the static Coulomb-hole and screened exchange approximation to the GW self-energy. Following our previous work for f-electron systems [H. Jiang, R. I. Gomez-Abal, P. Rinke, and M. Scheffler, Phys. Rev. Lett. 102, 126403 (2009)] we conduct a systematic investigation of the GW method based on LDA+U (GW@LDA+U), as implemented in our recently developed all-electron GW code FHI-gap (Green's function with augmented plane waves) for a series of prototypical d-electron systems: (1) ScN with empty d states, (2) ZnS with semicore d states, and (3) late transition-metal oxides (MnO, FeO, CoO, and NiO) with partially occupied d states. We show that for ZnS and ScN, the GW band gaps only weakly depend on U but for the other transition-metal oxides the dependence on U is as strong as in LDA+U. These different trends can be understood in terms of changes in the hybridization and screening. Our work demonstrates that GW@LDA+U with physical values of U provides a balanced and accurate description of both localized and itinerant states.

(Phys. Rev. B **82**, 045108 (2010))
Contact person: Patrick Rinke (rinke@fhi-berlin.mpg.de)

Stacking and Registry Effects in Layered Materials: The Case of Hexagonal Boron Nitride

N. Marom[1], J. Bernstein[3], J. Garel[1], A. Tkatchenko[2],
E. Joselevich[1], L. Kronik[1], O. Hod[3]
[1]*Department of Materials and Interfaces, Weizmann Institute of Science,*
Rehovot 76100, Israel
[2]*Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany
[3]*School of Chemistry, The Sackler Faculty of Exact Sciences,*
Tel Aviv University, Tel Aviv 69978, Israel

Abstract

The interlayer sliding energy landscape of hexagonal boron nitride (h-BN) is investigated via a van der Waals corrected density functional theory approach. It is found that the main role of the van der Waals forces is to anchor the layers at a fixed distance, whereas the electrostatic forces dictate the optimal stacking mode and the interlayer sliding energy. A nearly free-sliding path is identified, along which band gap modulations of ≈ 0.6 eV are obtained. We propose a simple geometric model that quantifies the registry matching between the layers and captures the essence of the corrugated h-BN interlayer energy landscape. The simplicity of this phenomenological model opens the way to the modeling of complex layered structures, such as carbon and boron nitride nanotubes.

(Phys. Rev. Lett. **105**, 046801 (2010))
Contact person: Alexandre Tkatchenko (tkatchen@fhi-berlin.mpg.de)

Structural metastability of endohedral silicon fullerenes

A. Willand[1], M. Gramzow[2], A. Ghasemi[1], L. Genovese[3], T. Deutsch[4],
K. Reuter[2,5] and S. Goedecker[1]
[1] *Department of Physics, Universität Basel,*
Klingelbergstr. 82, 4056 Basel, Switzerland
[2] *Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany
[3] *European Synchrotron Radiation Facility,*
6 rue Horowitz, 38043 Grenoble, France
[4] *INAC/SP2M/L_Sim, Commissariat à l'Energie Atomique,*
17 rue des Martyrs, 38054 Grenoble, France
[5] *Dept. Chemie, Technische Universität München,*
Lichtenbergstr. 4, 85747 Garching, Germany

Abstract

Endohedrally doped Si_{20} fullerenes appear as appealing building blocks for nanoscale materials. We investigate their structural stability with an unbiased and systematic global geometry optimization method within density-functional theory. For a wide range of metal-doping atoms, it was sufficient to explore the Born-Oppenheimer surface for only a moderate number of local minima to find structures that clearly differ from the initial endohedral cages but are considerably more favorable in terms of energy. Previously proposed structures are thus all metastable.

(Physical Review B **81**, 201405(R) (2010))
Contact person: Matthias Gramzow (gramzow@fhi-berlin.mpg.de)

First-principles study of the mechanism of ethylene epoxidation over Ag-Cu particles

S. Piccinin[1,2], N. L. Nguyen[3], C. Stampfl[2], and M. Scheffler[4]
[1]*DEMOCRITOS Simulation Center, CRN-IOM,*
Theory@Elettra Group, Trieste, Italy
[2]*School of Physics, The University of Sydney,*
Sydney, New South Wales 2006, Australia
[3] *Scuola Internazionale Superiore di Studi Avanzati (SISSA),*
Via Bonomea 265, 34136 Trieste, Italy
[4]*Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany

Abstract

Silver-copper alloys have been proposed as catalysts for ethylene epoxidation due to their superior selectivity compared to pure silver, the predominant catalyst for this reaction. Under reaction conditions it has been previously shown that, rather than a two-dimensional (2D) Ag-Cu alloy, a thin copper oxide-like layer forms on top of silver, and several possible surface structures were identified (Phys. Rev. Lett. **104**, 035503 (2010)). By means of density-functional theory calculations, we study the mechanism of ethylene epoxidation catalyzed by the thin oxide-like surface structures. We identify different reaction pathways that will compete and/or synergetically interplay in the catalysis. In general, the reaction mechanism is structure-dependent and often the reaction does not always proceeds through the formation of (meta)stable intermediates, in contrast to clean Ag and the 2D alloy. Analyzing the competing reactions, we discuss how the addition of Cu improves the selectivity and stress the overall importance of accounting for the effect of ambient conditions.

(submitted to: J. Mater. Chem. (June 18, 2010))
Contact person: Matthias Scheffler (scheffler@fhi-berlin.mpg.de)

Towards a first-principles chemical engineering: Transport limitations and bistability in in situ CO oxidation at $RuO_2(110)$

S. Matera[1] and K. Reuter[1,2]
[1]*Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany
[2]*Department Chemie, Technische Universität München,*
Lichtenbergstr. 4, D-85747 Garching, Germany

Abstract

We present a first-principles based multiscale modeling approach to heterogeneous catalysis that integrates first-principles kinetic Monte Carlo simulations of the surface reaction chemistry into a fluid dynamical treatment of the macro-scale flow structures in the reactor. The approach is applied to a stagnation flow field in front of a single-crystal model catalyst, using the CO oxidation at $RuO_2(110)$ as representative example. Our simulations show how heat and mass transfer effects can readily mask the intrinsic reactivity at gas-phase conditions typical for modern *in situ* experiments. For a range of gas-phase conditions we furthermore obtain multiple steady-states that arise solely from the coupling of gas-phase transport and surface kinetics. This additional complexity needs to be accounted for when aiming to use dedicated *in situ* experiments to establish an atomicscale understanding of the function of heterogeneous catalysts at technologically relevant gas-phase conditions.

(submitted to: Phys. Rev. B (June 1, 2010))
Contact person: Sebastian Matera (matera@fhi-berlin.mpg.de)

An Introduction to the Theory of Crystalline Elemental Solids and their Surfaces

A. Michaelides[1] and M. Scheffler[2]
[1] *London Centre for Nanotechnology and Department of Chemistry,*
University College London, London WC1E 6BT, UK
[2] *Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany

Abstract

A basic and general introduction to the physical properties and electronic structures of elemental crystalline solids and their surfaces is presented. This begins with a brief discussion of a few preliminary concepts of crystalline solids, such as an introduction to the common crystal types and a brief discussion of the cohesive properties of solids. Following this, the most widely used electronic structure technique for interrogating the properties of solids and their surfaces, namely density-functional theory will be introduced. We then discuss cohesion in bulk metals and semiconductors in more depth before reaching the main body of the chapter which involves a discussion of the atomic structures of crystalline solid surfaces, their energies, and their electronic structures in turn. We close with some general conclusions and perspectives for future work.

(To appear in: Textbook of Surface and Interface Science, Vol. I. (Ed.) K. Wandelt, Wiley-VCH (2010))

Contact person: Matthias Scheffler (scheffler@fhi-berlin.mpg.de)

Role of strain in polarization switching in semipolar InGaN/GaN quantum wells

Qimin Yan[1], Patrick Rinke[1,2], Matthias Scheffler[1,2], Chris G. Van de Walle[1]
[1] *Materials Department, University of California,*
Santa Barbara, California 93106-5050, USA
[2] *Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany

Abstract

The effect of strain on the valence-band structure of $(11\bar{2}2)$ semipolar InGaN grown on GaN substrates is studied. A **k·p** analysis reveals that anisotropic strain in the c-plane and shear strain are crucial for deciding the ordering of the two topmost valence bands. The shear-strain deformation potential D_6 is calculated for GaN and InN (D_6=-3.95 eV and -3.02 eV, respectively) using density functional theory with the Heyd-Scuseria-Ernzerhof hybrid functional. Using our deformation potentials and assuming a pseudomorphically strained structure, no polarization switching is observed. We propose that partial strain relaxation (e.g., due to dislocations) plays a role in the observed polarization switching.

(submitted to: Appl. Phys. Lett. (August 17, 2010))
Contact person: Patrick Rinke (rinke@fhi-berlin.mpg.de)

Direct assessment of quantum nuclear effects on hydrogen bond strength by constrained-centroid ab initio path integral molecular dynamics

Brent Walker and Angelos Michaelides
London Centre for Nanotechnology and Department of Chemistry,
University College London, London, WC1E 6BT, UK

Abstract

The impact of quantum nuclear effects on hydrogen (H-) bond strength has been inferred in earlier work from bond lengths obtained from path integral molecular dynamics (PIMD) simulations. To obtain a direct quantitative assessment of such effects, we use constrained-centroid PIMD simulations to calculate the free energy changes upon breaking the H-bonds in dimers of HF and water. Comparing *ab initio* simulations performed using PIMD and classical nucleus molecular dynamics (MD), we find smaller dissociation free energies with the PIMD method. Specifically, at 50 K the H-bond in $(HF)_2$ is about 30% weaker when quantum nuclear effects are included, while that in $(H_2O)_2$ is about 15% weaker. In a complimentary set of simulations, we compare unconstrained PIMD and classical nucleus MD simulations to assess the influence of quantum nuclei on the structures of these systems. We find increased heavy atom bond lengths, indicating weakening of the H-bond consistent with that observed by direct calculation of the free energies of dissociation.

(Submitted to Journal of Chemical Physics)
Contact person: angelos.michaelides@ucl.ac.uk

Theory of gold on ceria

Changjun Zhang[1], Angelos Michaelides[1], and Stephen J. Jenkins[2]
[1] *London Centre for Nanotechnology and Department of Chemistry,*
University College London, London, WC1E 6BT, UK
[2] *Department of Chemistry, University of Cambridge,*
Lenseld Road, Cambridge, CB2 1EW, UK

Abstract

The great promise of ceria-supported gold clusters as catalysts of the future for important industrial processes, such as the water gas shift reaction, has prompted a flurry of activity aimed at understanding the molecular-level details of their operation. Much of this activity has focused on experimental and theoretical studies of the structure of perfect and defective ceria surfaces, with and without gold clusters of various sizes. The complicated electronic structure of ceria, particularly in its reduced form, means that at present it is highly challenging to carry out accurate electronic structure simulations of such systems. To overcome the challenges, the majority of recent theoretical studies on these systems have adopted a pragmatic approach, applying the so-called DFT+U technique. Here we will briefly discuss some recent studies of Au on $CeO_2\{111\}$ that mainly use this approach. We will show that considerable insight has been obtained into these systems, particularly with regard to Au adsorbates and Au cluster reactivity. We will also briefly discuss the need for improved electronic structure methods, which would enable more rigorous and robust studies in the future.

(To appear on PCCP as a review article)
Contact person: angelos.michaelides@ucl.ac.uk

Magnetic-field control of the electric polarization in BiMnO$_3$

I. V. Solovyev
National Institute for Materials Science, 1-2-1 Sengen, Tsukuba,
Ibaraki 305-0047, Japan
Z. V. Pchelkina
Institute of Metal Physics, Russian Academy of Sciences - Ural Division,
620041 Ekaterinburg GSP-170, Russia

Abstract

We present the microscopic theory of improper multiferroicity in BiMnO$_3$, which can be summarized as follows: (1) the ferroelectric polarization is driven by the hidden antiferromagnetic order in the otherwise centrosymmetric $C2/c$ structure; (2) the relativistic spin-orbit interaction is responsible for the canted spin ferromagnetism. Our analysis is supported by numerical calculations of electronic polarization using the Berry-phase formalism, which was applied to the low-energy model of BiMnO$_3$ derived from the first-principles calculations. We explicitly show how the electric polarization can be controlled by the magnetic field and argue that BiMnO$_3$ is a rare and potentially interesting material where ferroelectricity can indeed coexist and interplay with the ferromagnetism.

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Contact person: Solovyev.Igor@nims.go.jp

Hybrid functionals within the all-electron FLAPW method: Implementation and application of PBE0

Markus Betzinger, Christoph Friedrich, and Stefan Blügel
Institut für Festkörperforschung and Institute for Advanced Simulations,
Forschungszentrum Jülich and JARA,
52425 Jülich, Germany

Abstract

We present an efficient implementation of the Perdew-Burke-Ernzerhof hybrid functional PBE0 within the full-potential linearized augmented-plane-wave (FLAPW) method. The Hartree-Fock exchange term, which is a central ingredient of hybrid functionals, gives rise to a computationally expensive nonlocal potential in the one-particle Schrödinger equation. The matrix elements of this exchange potential are calculated with the help of an auxiliary basis that is constructed from products of FLAPW basis functions. By representing the Coulomb interaction in this basis the nonlocal exchange term becomes a Brillouin-zone sum over vector-matrix-vector products. The Coulomb matrix is calculated only once at the beginning of a self-consistent-field cycle. We show that it can be made sparse by a suitable unitary transformation of the auxiliary basis, which accelerates the computation of the vector-matrix-vector products considerably. Additionally, we exploit spatial and time-reversal symmetry to identify the nonvanishing exchange matrix elements in advance and to restrict the $\mathbf{k}$ summations for the nonlocal potential to an irreducible set of $\mathbf{k}$ points. Favorable convergence of the self-consistent-field cycle is achieved by a nested density-only and density-matrix iteration scheme. We discuss the convergence with respect to the parameters of our numerical scheme and show results for a variety of semiconductors and insulators, including the oxides ZnO, EuO, $\mathrm{Al_2O_3}$, and $\mathrm{SrTiO_3}$, where the PBE0 hybrid functional improves the band gaps and the description of localized states in comparison with the PBE functional. Furthermore, we find that in contrast to conventional local exchange-correlation functionals ferromagnetic EuO is correctly predicted to be a semiconductor.

Phys. Rev. B **81**, 195117 (2010)
Contact person: m.betzinger@fz-juelich.de

Fractional occupation in Kohn-Sham density-functional theory and the treatment of non-pure-state v-representable densities

Eli Kraisler[1,2], Guy Makov[2,3], Nathan Argaman[2], Itzhak Kelson[1]
[1] *Raymond and Beverley Sackler Faculty of Exact Sciences,*
School of Physics and Astronomy, Tel Aviv University, Tel Aviv 69978, Israel
[2] *Physics Department, NRCN, P.O. Box 9001, Beer Sheva 84190, Israel*
[3] *Physics Department, Kings College London,*
The Strand, London WC2R 2LS, United Kingdom

Abstract

In the framework of Kohn-Sham density-functional theory, systems with ground-state densities that are not pure-state v-representable (PSVR) in the noninteracting reference system occur frequently. In the present contribution, an algorithm, which allows the solution of such systems, is proposed. It is shown that the use of densities which do not correspond to a ground state of their noninteracting reference system is forbidden. As a consequence, the proposed algorithm considers only noninteracting ensemble v-representable densities. The Fe atom, a well-known non-PSVR system, is used as an illustration. Finally, the problem is analyzed within finite-temperature density-functional theory, where the physical significance of fractional occupations is exposed and the question of why degenerate states can be unequally occupied is resolved.

(Published in Phys. Rev. A **80**, 032115 (2009))
Contact person: ekraisler@gmail.com

Projector self-consistent DFT+U using nonorthogonal generalized Wannier functions

David D. O'Regan[1], Nicholas D. M. Hine[1,2], Mike C. Payne[1], and Arash A. Mostofi[2]
[1]*Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue,*
Cambridge CB3 0HE, United Kingdom
[2]*The Thomas Young Centre, Imperial College London,*
London SW7 2AZ, United Kingdom

Abstract

We present a formulation of the density-functional theory+Hubbard model (DFT+U) method that is self-consistent over the choice of Hubbard projectors used to define the correlated subspaces. In order to overcome the arbitrariness in this choice, we propose the use of nonorthogonal generalized Wannier functions (NGWFs) as projectors for the DFT+U correction. We iteratively refine these NGWF projectors and, hence, the DFT+U functional, such that the correlated subspaces are fully self-consistent with the DFT+U ground state. We discuss the convergence characteristics of this algorithm and compare ground-state properties thus computed with those calculated using hydrogenic projectors. Our approach is implemented within, but not restricted to, a linear-scaling DFT framework, opening the path to DFT+U calculations on systems of unprecedented size.

(Published in Phys. Rev. B 82, 081102(R) (2010).)
Contact person: ddo20@cam.ac.uk

Special Issue on Theory of Superhard Materials

Edited by Artem R. Oganov
State University of New York, Stony Brook

Abstract

A special issue 'Theory of superhard materials' of the Journal of Superhard Materials has just appeared, with me as editor. The exciting field of superhard materials is strongly related to high pressure, as many of the novel superhard materials are discovered through the application of pressure. Examples are many - e.g., BC_2N and BC_5 discovered by Solozhenko in 2001 and 2009, respectively, nanodiamond by Irifune in 2003, γ-B28 phase of boron established in 2004-2006 by a collaboration of Chen, Solozhenko and Oganov, and a new superhard allotrope of carbon prepared in 2003 by Mao. Our Special Issue is dedicated to the latest theoretical advances in the understanding and prediction of materials' hardness, and theoretical search for novel superhard phases. Given the technical complexity of high-pressure experiment, theory is becoming a necessary instrument in the design of superhard materials and allows one to judge controversial results (such as the claimed high hardness of TiO_2-cotunnite). I hope that the emerging theoretical methods in this field will attract an increasing attention throughout the high-pressure community. The Special Issue is published online at:
`http://www.springerlink.com/content/u6jwv4501575/?p=5ec1ee31eef54688891cbf224a91e9e8&pi=0`

We have successfully negotiated open access until the end of 2010.

The contents are as follows:

1. Towards the theory of hardness of materials (A.R. Oganov and A.O. Lyakhov)
2. Microscopic models of hardness (F.M. Gao and L.H. Gao)
3. Thermodynamic model of hardness: Particular case of boron-rich solids (V.A. Mukhanov, O.O. Kurakevych and V.L. Solozhenko)
4. Intrinsic hardness of crystalline solids (J.S. Tse)
5. Predicting new superhard phases (Q. Li, H. Wang and Y.M. Ma)
6. Electronic structures and mechanical properties of boron and boron-rich crystals (Part I) (K. Shirai)

Let me also add, as a brand new member of the Editorial Board, that Journal of Superhard Materials welcomes submission of high-quality papers on all theoretical and experimental aspects related to the field of superhard materials.

Contact person:
Prof. Artem R. Oganov
Department of Geosciences and Department of Physics and Astronomy,
State University of New York, Stony Brook, NY 11794-2100
Phones: (631)-632-1429 (office), x1449 (lab), x8241 (admin. assistant)
Fax (631) 632 8240
E-mail artem.oganov@sunysb.edu
Web-site: http://mysbfiles.stonybrook.edu/ aoganov

6 Presenting USPEX Method/Code

USPEX is a method/code for crystal structure prediction and is now available for downloading from the web:

`http://han.ess.sunysb.edu/~USPEX/downloads.html`

USPEX is an evolutionary method/code, which was developed by A. Oganov's group in 2005 at ETH Zurich, and has been instrumental in many important results published in over 50 articles. There are important new methodological developments (see list of publications on the website). Its applications in high-pressure science have yielded a large number of unexpected results.

The code is now used by well over 100 researchers worldwide. In its early stages, the code was distributed through licenses issued by ETH Zurich. Now the licenses are superseded by the online registration procedure. The code is distributed as an open source and is free for non-commercial research. The code is user-friendly, and we gladly provide guidance for new users.

It may be of interest that later in 2010 a book on crystal structure prediction will be published by Wiley-VCH:

`http://www.wiley-vch.de/publish/en/books/forthcomingTitles/NT00/3-527-40939-4/?sID=pk4rq15hcr5b64p5huhfk98p80`

Among many other things, the book contains the first blind test of structure searching methods, which has demonstrated compelling performance of USPEX.

We are in the process of finalizing the next version of USPEX (v.8), which features new ideas for enhancing efficiency, variable-composition simulations (where one determines both the stable compositions and structures in a multicomponent system), optimization of certain physical properties, and prediction of structures of nanoparticles and surfaces.

We truly hope that you will find USPEX to be useful in your research. Happy USPEXing!

Best regards,

USPEX developers team (Andriy, Qiang, Artem)

Prof. Artem R. Oganov
Department of Geosciences and Department of Physics and Astronomy,
State University of New York, Stony Brook, NY 11794-2100
Phones: (631)-632-1429 (office), x1449 (lab), x8241 (admin. assistant)
Fax (631) 632 8240
E-mail artem.oganov@sunysb.edu
Web-site: http://mysbfiles.stonybrook.edu/ aoganov

7 SCIENTIFIC HIGHLIGHT OF THE MONTH: Correlation effects in transition metals and their alloys studied by the fully self consistent KKR+DMFT method

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Correlation effects in transition metals and their alloys studied by the fully self consistent KKR+DMFT method

Ján Minár

Universität München, Department of Chemistry, Physical Chemistry
Butenandtstr. 5-13, D-81377 München, Germany

Abstract

Recent results on magnetic and spectroscopic properties of transition metals, alloys and their surfaces, which were obtained by the LSDA+DMFT approach will be reviewed in this article. We exploit the various advantageous features offered by a LSDA+DMFT implementation on the basis of the KKR band structure method. This fully self consistent approach, with respect to charge and self-energy, allows to investigate in a most realistic way the impact of correlation effects in ordered as well as disordered systems. For the latter case the coherent potential approximation (CPA) is exploited. The fully-relativistic formulation implemented for space filling potentials gives the opportunity to study various ground state properties as for example orbital magnetic moments or total energies. Experimentally, the most detailed information on the electronic properties of correlated materials can be accessed by means of angle-resolved photoemission and inverse photoemission. In several earlier studies the measured spectra were described either within a single-particle approach based on the local spin density approximation, including matrix-element- and surface effects within so called one-step model or by sophisticated many-body approaches neglecting these effects. Here, we demonstrate that the combination of the one-step model with the LSDA+DMFT gives a detailed insight and the most stringent comparison between theory and corresponding experimental data.

1 Introduction

Density functional theory (DFT) is the state of the art technique for calculating the electronic and magnetic properties of a wide spectrum of systems ranging from atoms and molecules to surfaces and solids [1]. Its successes in treating the many-body problem is mainly due to the virtues of the local spin density approximation (LSDA) [2–4]. However, in spite of numerous impressive success it faces serious difficulties for strongly correlated materials such as Mott insulators, heavy fermion systems, high-temperature superconductors and many others. Nowadays, there exist various ways to improve the rather simple but successful LSDA. One way is to stay within the DFT formalism and to develop improved exchange correlation functionals like, for example, generalized gradient approximation (GGA) based functionals [5], orbital dependent functionals based on the optimized potential (OEP) method [**?**, 7] or self-interaction correction (SIC) [8] derived functionals. In order to determine excitation spectra quantitatively, time dependent DFT has been developed, but in practice the exchange-correlation kernel based on LSDA is still of limiting applicability.

To include many-body effects beyond LSDA and to describe different types of excitations, various many-body techniques have been developed. One of the most successful approaches is the dynamical mean field theory (DMFT) [9] that was introduced about 20 years ago. Within this approach more or less all local correlation effects like the Mott insulating state or quasiparticle excitations have been successfully described. The only approximation made within the DMFT in its standard formulation is the neglect of non-local, spatial correlations. In practice the DMFT has been primarily applied to solve various model Hamiltonians with limiting applicability to the real materials.

To go beyond the level of a model-like treatment and to investigate real materials with strong correlation effects a combination of DFT based methods supplemented by many-body theories have been suggested. In practice this leads to a complex, energy-dependent and non-local self-energy which has to be combined coherently into the existing DFT based methods by means of the Dyson equation for the Green's function. As a consequence, this combination allows for a realistic description of one-particle excitations in correlated systems. As an example, let us mention the GW-approximation [10] well suited for the case of insulators and semiconductors. Another approach is to consider Hubbard-type models where Coulomb-type interaction terms are included explicitly, which are assumed to be treated insufficiently within LSDA. Already the simplest Hartree-Fock like realization of such an approach called LSDA+U [11] scheme allows one to improve considerably the description of strongly correlated materials. Later on more advanced methods that represent correlation effects in terms of a complex and energy-dependent self-energy have been proposed. For example, the so called 3-body scattering method (3BS) [12] has been developed, which in principle is very well suited for the description of photoemission. Without any doubts, nowadays, the most successful approach consists in a combination of DFT with DMFT, the so called LSDA+DMFT method, that was developed during the last ten years or so (for reviews see [13, 14]). In spite of impressive progress in the development and application of the LSDA+DMFT method this framework has still to be improved. Recently Albers et al. [15] critically reviewed various issues connected with the LSDA+DMFT. In particular, to turn the LSDA+DMFT into a *method with predictive power for strongly correlated materials* [1] various important problems have to be solved, for example a fully self consistent coherent interface between LSDA and DMFT, reliable calculations of the Coulomb parameter U, the choice of the correlated orbitals, reliable DMFT impurity solvers applicable to a wide range of systems. In addition, a comparison to various spectroscopic properties is an important tool to check the achievements of theoretical developments. Concerning this, however, often only the pure spectral function is compared with experiment ignoring spectroscopic specific issues like e.g. matrix elements.

The aim of this contribution is to review the various advantageous features of the LSDA+DMFT implementation within the multiple scattering Korringa-Kohn-Rostoker (KKR) method [16]. This

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[1] This words have been borrowed from the title of new collaborative Research Unit FOR1346 (funded by German DFG). The main general objective is to ultimately create a new standard of computational electronic structure schemes beyond LDA on a level which is suitable to predict and compute the properties of complex, correlated materials.

approach has been developed because the KKR method [17] represents the electronic structure by the corresponding single-particle Green's function leading to a number of attractive features of this scheme. In particular, several of the above mentioned questions could be addressed within the present approach. In Sec. 2 a combination of the LSDA+DMFT with the KKR band structure method is discussed. Various calculations of ground state properties are shown in Sec. 3.1.1. The second part of this review is devoted to the calculation of angle-resolved photoemission within so called one-step model including in particular more or less all relevant spectroscopy issues like, e.g. matrix elements and surface effects. Recent technical developments allow to perform calculations for ordered as well as of chemically disordered systems including electronic correlation effects. This topic is presented in Secs. 2.2.4 and 3.2.2.

2 Theory

In the late 1980s the dynamical mean field theory (DMFT) became a very important concept when dealing with many body Hubbard Hamiltonians for solids. Within the DMFT the infinite interacting many body lattice problem is mapped onto a single quantum impurity exposed to a mean field given by an effective bath. This central concept of the standard DMFT becomes exact in the limit of high lattice coordination number [18]. As a consequence of the DMFT equations for the on-site complex energy dependent self-energy and fully interacting Green's function these has to be calculated self consistently. Within this approach, even when dealing with a model Hubbard Hamiltonian, a lot of insight for example into the Mott-Hubbard transition has been obtained [9]. However, it has also been realized that in order to calculate the electronic structure properties of realistic materials the combination of the LSDA with the DMFT is desirable. First successful steps in this direction had been done by Anisimov et al. [19] and Katsnelson et al. [20]. In the LSDA+DMFT the local one particle Green's function needed as an input to the DMFT self consistent cycle is obtained from the LSDA method. Up to now most of the LSDA+DMFT implementations (see for example reviews [13, 14] and references given there) have been performed using only DMFT self consistency. This means the LSDA charge density and consequently the effective LSDA potential has been fixed. Therefore, one neglects the impact of correlation effects on the charge distribution. Until now only few completely self consistent schemes, e.g. with respect to charge density and self-energy, have been proposed and implemented. To underline the importance of the complete LSDA+DMFT self consistency we mention the problem of phase diagram calculations and localization in f-electron systems [21, 22], the description of non-quasiparticle states in the Heusler alloys [23] and the problem to describe angle-integrated [24] and angle-resolved photoemission of 3d-ferromagnets [25]. The first fully self consistent implementations of the LSDA+DMFT have been done on the basis of the linear muffin tin orbital method (LMTO) [21], followed by the EMTO method [26]. One of the first self consistent schemes based on a full potential scheme has been proposed by Minar et al. [16] for the KKR Green's function method. Most of the work done by the LSDA+DMFT method deals with electronic and magnetic properties of strongly correlated materials. However, correlation effects are also often of crucial importance to describe the cohesive energy, the equilibrium lattice constant and the bulk modulus as demonstrated for the case of plutonium [22, 27] and cerium [28]. After these first experiences, the need for more systematic implementations and investigations on numerical problems of LSDA+DMFT emerged.

A very detailed study of the effects of self- consistency on the total energy has been presented by Pourovskii et al. [29]. In spite of various numerical problems, recently, the huge potential of the LSDA+DMFT method concerning total energies has been demonstrated. For example, the collapse of the magnetic moment of MnO [30], equilibrium volume and bulk modulus of Ni and Mn [31] or correlation driven structural properties of complex materials like $KCuF_3$ and $LaMnO_3$ [32] had been discussed.

In this section we briefly review a full potential KKR implementation of the DMFT theory. We will discuss the various advantages of this approach like, for example, fully self-consistency, the total energy problem and the combination of the LSDA+DMFT with the coherent potential approximation and its fully relativistic extension. In addition, the combination of the DMFT with the one step model of photoemission allows for a direct comparison to the angle-resolved photoemission of ordered as well as of disordered systems including matrix elements, final state and surface effects.

2.1 Formulation of the problem

All standard approaches for a realistic electronic structure calculation of strongly correlated materials start from a choice of the set of orbitals that are not accurately described enough within the standard DFT LSDA method. We call them "correlated orbitals" and indicate them with $\phi_i^n(\vec{r})$, where n specifies the Bravais lattice site and the i is an index that enumerates the orbitals within the unit cell of the crystal. The choice of "correlated orbitals" is dictated by physical motivations for the problem under consideration and always implies some degree of arbitrariness. Usually the correlated orbitals are derived from d or f atomic states and the index i stands for the atomic quantum numbers l, m, σ. Natural choices can be the Linear Muffin-Tin Orbitals [22] or the Wannier functions [33, 34] or downfolded orbitals constructed via NMTO approach [35].

After having decided for the set of "correlated orbitals", we correct the standard DFT LSDA Hamiltonian with an additional Hubbard interaction term [13] that explicitly accounts for the local Coulomb repulsion for the orbitals in the set:

$$H = H_{LSDA} + H_U - H_{DC} \; . \tag{1}$$

Here H_{LSDA} stands for the ordinary LSDA Hamiltonian, H_U describes the additional effective electron-electron interaction and the one-particle Hamiltonian H_{DC} serves to eliminate double counting of the interactions already accounted for by H_{LSDA}. H_{DC} used in our implementation for transition metals and compounds will be discussed later in Sec. 2.2. Similar to the LSDA+U method [11], this many-body Hamiltonian is specified by a number of parameters representing Coulomb matrix elements. Among these parameters the Hubbard U that represents the screened on-site Coulomb interaction is the most important one. Accordingly, several schemes have been suggested in the literature to determine U from LSDA calculations [14, 36]. However, in this work we use it as a parameter. The resulting many-particle Hamiltonian can not be diagonalized exactly, thus various methods were developed in the past to find an approximate solution [9]. Among them one of the most promising approaches is to deal with Eq. (1) within the DMFT.

The main idea of DMFT is to map a periodic many-body problem onto an effective single-impurity problem that has to be solved self consistently. For this purpose one describes the electronic properties of the system in terms of the one particle Green's function $\hat{G}(E)$, being the solution of the equation:

$$[E - \hat{H}_{LSDA} - \hat{\Sigma}(E)]\hat{G} = \hat{1} \,, \tag{2}$$

where E is the complex energy and the effective self-energy operator $\hat{\Sigma}$ is assumed to be a single-site quantity for site n:

$$\hat{\Sigma}(E) = \sum_{ij} |\phi_{ni}\rangle \Sigma_{ij}(E) \langle \phi_{nj}| \,. \tag{3}$$

Within DMFT, the self-energy matrix $\Sigma_{ij}(E)$ is a solution of the many-body problem of an impurity placed in an effective medium. This medium is described by the so called *bath* Green's function matrix $\mathcal{G}$ defined as:

$$\underline{\mathcal{G}}^{-1}(E) = \underline{G}^{-1}(E) + \underline{\Sigma}(E) \,, \tag{4}$$

with the underline indicating matrices with respect to orbital index i. The Green's function matrix $G_{ij}(E)$ is calculated as a projection of $\hat{G}(E)$ onto the impurity site:

$$G_{ij}(E) = \langle \phi_i^n | \hat{G}(E) | \phi_j^n \rangle \,. \tag{5}$$

Because the self-energy $\Sigma_{ij}(E)$ depends on the *bath* Green's function $\mathcal{G}_{ij}(E)$ the DMFT equations have to be solved self consistently. From a technical point of view the problem can be split into two parts. One is dealing with the solution of Eq. (2) and the second one is the effective many-body problem to find the self-energy $\Sigma_{ij}(E)$ according to Eq. (4). Within the present work, the first task is solved by the KKR band structure method, as described below in Sec. 2.2. The choice and details of solving the many-body effective impurity problem, often referred to as DMFT-solver, can be found in various LSDA+DMFT reviews for example [13, 14].

2.2 Multiple scattering implementation of the LSDA+DMFT method

In the following section we shortly review a fully self consistent (with respect to charge density and self-energy) LSDA+DMFT implementation within the full potential fully relativistic multiple scattering Korringa Kohn Rostoker (KKR) method [16]. This approach was motivated because the KKR method represents the electronic structure by the corresponding single-particle Green's function leading therefore to a number of very attractive features of the scheme that are exploited in the subsequent work [2]. This allows to combine the KKR method with the DMFT straightforwardly. In addition, an outstanding feature of the KKR method is the possible use of the Dyson equation which relates the Green's function of a perturbed system with the Green's function of the corresponding unperturbed reference system. Using the Dyson equation allows to calculate various properties of low dimensional systems like e.g. semi infinite 2D-surfaces, nano-structures or embedded 3D- or 2D- systems. In addition, the KKR Green's function method allows to deal with substitutional disordered alloys in the framework of the coherent potential approximation (CPA) [37].

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[2] For a recent review where various advantages of the KKR method are listed see Ψ_k highlight of the month [17] and references therein.

The idea of the LSDA+DMFT approach presented here is to account for the general non-local, site-diagonal, complex and energy-dependent self-energy Σ^{DMFT} into the KKR method already when calculating the basis functions, i.e. when solving the single site Schrödinger (or Dirac) equation. This allows directly to exploit all features of the KKR Green's function method within LSDA+DMFT calculations and consequently allows to quantitatively account for correlation effects.

As already mentioned, there are various approaches available to combine the LSDA with the DMFT method [13, 14]. In most implementations as a first step the corresponding LSDA problem is solved variationally using a given basis set (e.g. LMTO). The corresponding local Green's function is determined by the spectral representation of the Kohn-Sham Hamiltonian. The resulting local self-energy Σ^{DMFT} and local Green's function can be in turn used to calculate a new charge density and an effective LSDA potential. However, in order to combine coherently the LSDA with the DMFT method (in the spirit of spectral density functional theory [22]) one needs to solve self consistently the following Dyson equation:

$$\begin{aligned} G(\vec{r},\vec{r}\,',E) &= G_0(\vec{r},\vec{r}\,',E) \\ &+ \int \mathrm{d}^3r'' \int \mathrm{d}^3r''' G_0(\vec{r},\vec{r}\,'',E)\left[V_{\mathrm{eff}}(\vec{r}\,'')\delta(\vec{r}\,''-\vec{r}\,''') + \Sigma(\vec{r}\,'',\vec{r}\,''',E)\right] G(\vec{r}\,''',\vec{r}\,',E), \end{aligned} \tag{6}$$

where $G_0(\vec{r},\vec{r}\,'',E)$ is the free electron Green's function. The potential $V_{\mathrm{LSDA}}(\vec{r})$ denotes the (effective) potential (in the relativistic spin DFT defined as $V_{\mathrm{eff}}(\vec{r}) = [\overline{V}_{\mathrm{eff}}(\vec{r}) + \beta\boldsymbol{\sigma}\mathbf{B}_{\mathrm{eff}}(\vec{r})]$ where $\overline{V}_{\mathrm{eff}}(\vec{r})$ denotes the spin-independent potential, and $\mathbf{B}_{\mathrm{eff}}(\vec{r})$ is the magnetic field [38]). β and $\boldsymbol{\alpha}$ used below are standard Dirac matrices with the latter one given by $\alpha_k = \sigma_x \otimes \sigma_k$ $(k = x, y, z)$ in terms of the 2×2 Pauli-matrices σ_k.

A very efficient way to solve Eq. (6) is offered by the multiple scattering KKR method. Having decomposed the system into atomic regions (Wigner-Seitz-cells) and considering that Σ^{DMFT} is an on-site quantity the equation can be solved using the standard KKR formalism. This implies that first one must solve the so-called single site scattering problem to obtain the single site t-matrix and the corresponding scattering wave function $\Psi(\vec{r})$. In the relativistic spin density functional theory [1, 39] the corresponding single site Dirac equation reads:

$$\left[\frac{\hbar}{i}c\vec{\alpha}\cdot\vec{\nabla} + \beta mc^2 + V_{\mathrm{eff}}(\vec{r}) + \int d^3r'\Sigma(\vec{r},\vec{r}\,',E)\right]\Psi(\vec{r}) \quad = \quad E\Psi(\vec{r})\,. \tag{7}$$

Here, the $\Psi(\vec{r})$ are four-component spinor functions with corresponding energies E. To be able to solve Eq. (7) one makes the following ansatz for the wave function $\Psi = \sum_\Lambda \Psi_\Lambda$, with the combined relativistic quantum number $\Lambda = (\kappa, \mu)$. In addition, in the spirit of the DMFT $\Sigma(\vec{r},\vec{r}\,',E)$ one has to project onto the localized set of orbitals $\phi^n_\Lambda(\vec{r})$. The corresponding matrix $\Sigma_{\Lambda,\Lambda'}(E)$ is obtained as an output of the DMFT solver. In practice, $\Sigma_{\Lambda\Lambda'}(E)$ is used only for correlated d- or f- orbitals (see above). It is worth to note that even in the case of the spherical muffin-tin or atomic sphere approximation to the potential, full-potential like coupled Eqs. (7) have to be solved. This implies that the full-potential version of the KKR has to be used. After having solved the set of coupled equations for the wave functions one gets the corresponding single site t matrix by standard matching to the Hankel and Bessel functions as free electron solutions. When solving the single-site problem, obviously the entire complexity of the underlying complex non-local potential within LSDA+DMFT is accounted for. Accordingly, the resulting regular and irregular scattering wave functions $Z_\Lambda(\vec{r},E)$ and $J_\Lambda(\vec{r},E)$ as well as the corresponding single-site t-matrix carry all information on the underlying LSDA+DMFT Hamiltonian. This means that in contrast to other LSDA+DMFT implementations, the effect of the self-energy is also reflected in the wave functions Ψ. This becomes important, for example, in a total energy calculation and for the photoemission matrix elements (see Sec. 2.2.4).

With the single-site t matrix available the next step of the KKR calculation is to solve the multiple scattering problem. This task can be done by using the scattering path operator τ [17] and it is independent from the DMFT. For a finite system this can be done straight forwardly by inverting the so called KKR-matrix, $\underline{\underline{\tau}}(E) = [\underline{\underline{t}}(E)^{-1} - \underline{\underline{G}}_0(E)]^{-1}$ with the double underline indicating matrices with respect to site and spin-angular (Λ) character. Dealing with a three-dimensional periodic system this equation can also be solved exactly by Fourier transformation. As a result the retarded site diagonal Green's function $G(\vec{r},\vec{r}\,',E)$ can be written as [37, 40]:

$$\begin{aligned} G(\vec{r},\vec{r}\,',E) &= \sum_{\Lambda,\Lambda'} Z_\Lambda(\vec{r},E)\tau^{nn}_{\Lambda,\Lambda'}(E)Z^\times_{\Lambda'}(\vec{r}\,',E) \\ &- \sum_{\Lambda}\{Z_\Lambda(\vec{r},E)J^\times_\Lambda(\vec{r}\,',E)\Theta(\vec{r}\,'-\vec{r}) \\ &+ J_\Lambda(\vec{r},E)Z^\times_\Lambda(\vec{r}\,',E)\Theta(\vec{r}-\vec{r}\,')\}\;, \end{aligned} \tag{8}$$

where $\vec{r}$ ($\vec{r}\,'$) lies in the atomic cell n representing cell-centered coordinates and $\times$ indicates a so-called left-hand side solution [41]. With the Green's function $G(\vec{r},\vec{r}\,',E)$ available all properties of interest as e.g. , the charge density, can be calculated straight forwardly and in this way the calculated Green's function G includes all effects of the self-energy Σ^{DMFT}. The definition of the Green's function and the expressions given above are not restricted to real energies E but also holds for arbitrary complex energies z. The fact that $G(\vec{r},\vec{r}\,',E)$ is analytical [42] allows, in particular, to perform the energy integration for the charge density on a contour in the complex energy plane [43,44] with typically around 30 energy mesh points. On the other hand the self-energy Σ^{DMFT} is often calculated for a mesh of Matsubara frequencies. This implies that it is necessary to use analytical continuation techniques to transform Σ^{DMFT} from Matsubara frequencies ω onto the KKR complex energy contour. It is worth to note that in general Σ^{DMFT} is not Hermitian and for low symmetry systems one has to consider right and left handed solutions of (7) when constructing the Green's function $G(\vec{r},\vec{r}\,',E)$ [41].

In order to construct the bath Green's function needed as the input of the DMFT solver, the localized Green's function is calculated by projecting the total Green's function onto the correlated atomic site. This is done by projecting $G(\vec{r},\vec{r}\,',E)$ with a localized set of orbitals $\phi_\Lambda(\vec{r})$:

$$G_{\Lambda\Lambda'}(E) = \int d^3r \int d^3r\,'\phi_\Lambda(\vec{r})G(\vec{r},\vec{r}\,',E)\phi_{\Lambda'}(\vec{r}\,')\;. \tag{9}$$

The multiple scattering formalism provides the natural choice of the projectors $\phi_\Lambda(\vec{r})$ which are the regular single-site solutions of the Kohn-Sham-Dirac equations. For transition metal systems only the d-d sub-block of the $\underline{G}^{nn}(E)$ is considered with $\phi_\Lambda(\vec{r})$ wave functions with $l = 2$. In principle the choice of the $\phi_\Lambda(\vec{r})$ is arbitrary as long as $\phi_\Lambda(\vec{r})$ is a complete set of functions. This implies that a localized basis set is calculated at a given reference energy E_{ref} (set to be the center of gravity of the occupied d- or f-band) with the magnetic field set to zero in the relativistic case. In

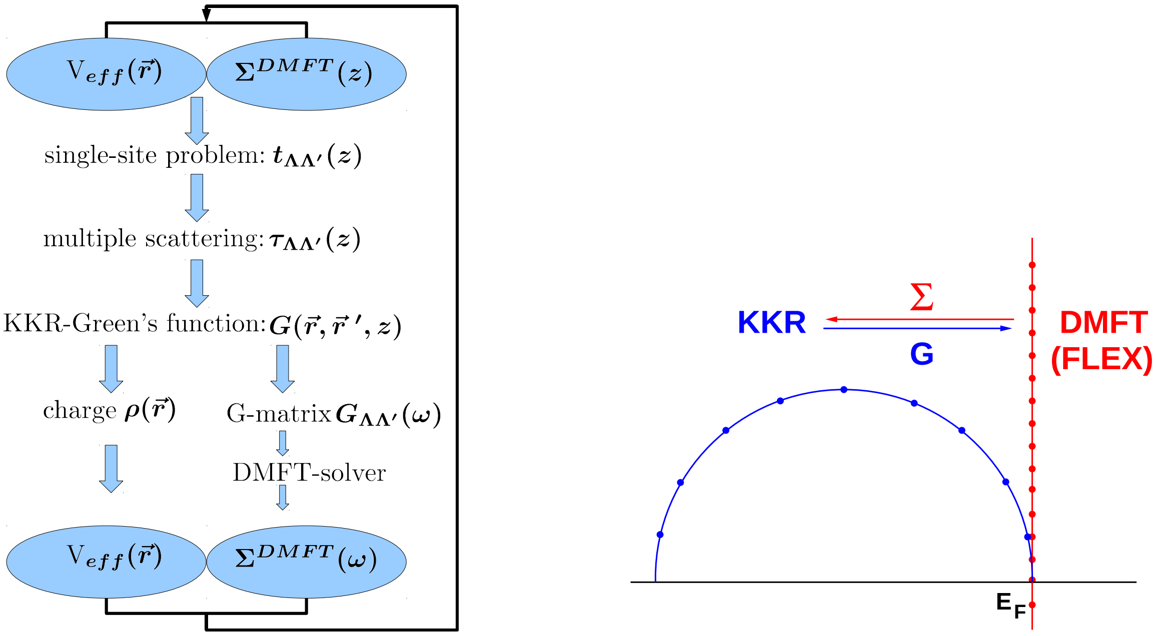


Figure 1: Left: Schematic overview of the KKR+DMFT scheme. Right: Illustration of the energy paths involved. The blue semicircle is the complex energy path used by KKR to calculate the charge density. After the bath Green's function G is obtained, it is analytically continued onto the imaginary axis (red) to calculate the self-energy Σ^{DMFT} via the DMFT impurity solver. The latter is analytically extrapolated back to the semicircle.

the full-potential case couplings to the other l-channels as a consequence of crystal symmetry have to be suppressed.

Finally, a description of the flow diagram of the self consistent LSDA+DMFT approach is presented in Fig. 1. Eq. (7) provides the set of regular (Z) and (J) irregular solutions of the single-site problem. Together with the t matrix and the scattering path operator τ the KKR Green's function is constructed from Eq. (8). To solve the many-body problem the projected impurity Green's function is constructed according to Eq. (9). The LSDA Green's function $G_{\Lambda\Lambda'}(E)$ is calculated on the complex contour which encloses the valence band one-electron energy poles. The Padé analytical continuation scheme is used to map the complex local Green's function $G_{\Lambda\Lambda'}(E)$ on the set of Matsubara frequencies or real axis, respectively, which is used when dealing with the many-body problem. In the current fully relativistic implementation the perturbative SPTF (spin-polarized T-matrix + FLEX) [45] as well as $T = 0K$ spin-polarized T-matrix [46] solvers of the DMFT problem are used. In fact any DMFT solver could be included which supplies the self-energy $\Sigma(E)$ as a solution of the many-body problem. The Padé analytical continuation is used once more to map back the self-energy from the Matsubara axis to the complex plane, where the new KKR Green's function is calculated. As was described in the previous sections, the key role is played by the scattering path operator $\tau^{nn}_{\Lambda\Lambda'}(E)$, which allows us to calculate the charge in each SCF iteration and the new potentials that are used to generate the new single particle Green's function.

Of course double counting corrections H_{DC} have to be considered explicitly. The problem of the double-counting correction is definitely one of the main challenges towards first-principle calculations within LSDA+DMFT. Until know various schemes for double counting correction have been suggested [14]. The simplest choice, i.e. the idea of the static LSDA+U scheme has been used here. We apply the double counting corrections to the self-energy when solving the many-body problem. In the case of metallic transition metals, their compounds and alloys the so called around mean field (AMF) double counting correction seems to be most appropriate. It is worth to note here, that until now it is more or less impossible to derive an exact analytical equation for the double counting correction. An alternative way to get an exact solution of the double counting problem seems to be possible on the level of the GW+DMFT scheme [47]. Therefore, it is important to use a comparison to angle-resolved photoemission (ARPES) experiments as a stringent criterium for the choice of the optimal H_{DC}. However, to be able to make a decision between various suggestions for H_{DC} it is helpful to calculate not only the bare spectral function, e.g. ImG, but instead to perform a complete photoemission calculation. (for example with the one step model of photoemission, see Sec. 2.2.4). In fact, using the one step model of photoemission one can clearly see that the AMF H_{DC} is an appropriate choice for at least transition metals [25, 48, 49].

2.2.1 LSDA+DMFT treatment of disordered alloys

An outstanding feature of the KKR method is the Dyson equation relating the Green's function of a perturbed system with the Green's function of the corresponding unperturbed reference system. Because of this property, the KKR Green's function method allows to deal with substitutional disorder including both diluted impurities and concentrated alloys in the framework of the CPA [50, 51]. Within this approach (KKR-CPA) the propagation of an electron in an alloy is regarded as a succession of elementary scattering processes due to random atomic scatterers, with an average taken over all configurations of the atoms. This problem can be solved assuming that a given scattering center is embedded in an effective medium whose choice is open and can be determined in a self consistent way. The physical condition corresponding to the CPA is simply that a single scatterer embedded in the effective CPA medium should produce no further scattering on the average. A similar philosophy is applied also when dealing with many-body problems for crystals in the framework of DMFT. Thus it seems to be rather natural to combine the DMFT and KKR-CPA method. In fact the combination of the KKR-CPA for disordered alloys and the DMFT scheme is based on the same arguments as used by Drchal et al. [52] when combining the LMTO Green's function method for alloys [53] with the DMFT.

The combination of the CPA and LSDA+DMFT turned out to be a quite powerful technique to calculate electronic structure properties of substitutionally disordered correlated materials [16, 54–56]. Within the CPA the configurationally averaged properties of a disordered alloy are represented by a hypothetical ordered CPA-medium, which in turn may be described by a corresponding site-diagonal scattering path operator τ^{CPA}, which in turn is closely connected with the electronic Green's function. For example for a binary system $\mathrm{A}_x\mathrm{B}_{1-x}$ composed of components A and B with relative concentrations x_{A} and x_{B} the corresponding single-site t-matrix t^{CPA} and the multiple scattering path operator τ^{CPA} are determined by the so called CPA-condition:

$$x_{\mathrm{A}}\tau^{\mathrm{A}} + x_{\mathrm{B}}\tau^{\mathrm{B}} = \tau^{\mathrm{CPA}}. \tag{10}$$

The above equation represents the requirement that embedding substitutionally an atom (of type A or B) into the CPA medium should not cause additional scattering. The scattering properties of an

A atom embedded in the CPA medium, are represented by the site-diagonal component-projected scattering path operator τ^{A} (angular momentum index omitted here)

$$\tau^{\mathrm{A}} = \tau^{\mathrm{CPA}} \left[1 + \left(t_{\mathrm{A}}^{-1} - t_{\mathrm{CPA}}^{-1} \right) \tau^{\mathrm{CPA}} \right]^{-1} \quad , \tag{11}$$

single-site matrices of the A component and of the CPA effective medium. A corresponding equation holds also for the B component in the CPA medium. The coupled sets of equations for τ^{CPA} and t^{CPA} have to be solved iteratively within the CPA cycle.

The above scheme can straightforwardly be extended to include the many-body correlation effects for disordered alloys [16]. Within the KKR approach the local multi-orbital and energy dependent self-energies ($\Sigma_A^{DMFT}(E)$ and $\Sigma_B^{DMFT}(E)$) are directly included into the single-site matrices t_A and t_B, respectively by solving the corresponding Dirac Eq. (7). Consequently, all the relevant physical quantities connected with the Green's function, as for example, the charge density contain the electronic correlations beyond the LSDA scheme.

2.2.2 Combination of the LSDA+DMFT with disordered local moment approach

In the following subsection we address the question to which extent a regime of strong correlations can be studied by means of an implementation suited to deal with moderately correlated systems. In particular the spin-polarized T-matrix fluctuation-exchange solver (SPTF) [45, 57] has been implemented to treat the problem of magnetic fluctuations in transition metals, and has been successfully applied to the ferromagnetic phases of Fe, Co, Ni [25, 57, 58] and to the antiferromagnetic phase of γ-Mn [59], as well as to half-metallic ferromagnets [23]. It is quite stable, computationally rather cheap and deals with the complete four-indices interaction matrix. On the other hand, its perturbative character restricts its use to relatively weakly, or moderately, correlated systems.

Not surprisingly, the SPTF performs well when starting from a spin-polarized solution, since the spin-splitting contains already the main part of the exchange and correlation effects. On the other hand, the direct application of SPTF to a non-magnetic reference state can create stability problems. This is because one tries to attribute the strong and essentially mean-field effect of the formation of a local magnetic moment to dynamical fluctuations around the non-spin-polarized state. This is fine when one uses the quantum monte carlo (QMC) method, which has no formal restrictions on the value of fluctuations, but seems problematic for the perturbative approaches. As a way to reduce such a limitation we propose a combination of SPTF with the disordered local moment (DLM) approach [60, 61]. As already shown for the case of actinides [62] the inclusion of the fluctuations of randomly oriented local moments can improve drastically the description of energetics in the paramagnetic phase. Therefore, one can hope that it allows us to extend the range of applicability of SPTF.

In DLM the itinerant electrons form self-maintaining "local moments" which are analogous - but physically different - to the localized spins of the Heisenberg model. With $\hat{\mathbf{e}}_i$ the orientations of the moments at the sites i, we can describe the system through the generalized grand-canonical potential $\Omega(\{\hat{\mathbf{e}}_i\})$. Then a mean-field approximation of the true potential is constructed as expansion around a single-site spin Hamiltonian [61]:

$$\Omega_0(\{\hat{\vec{e}}_i\}) = -\sum_i \vec{h}_i \hat{\vec{e}}_i. \tag{12}$$

The self consistent parameters h_i define a set of probabilities $P_i(\hat{\vec{e}}_i)$ of finding the moments oriented along $\hat{\vec{e}}_i$. Explicit calculations can now be made using standard methods developed for substitutionally disordered alloys, as the Coherent Potential Approximation (CPA), which is straightforwardly implemented within the multiple-scattering theory of KKR [16] (see above). The study of the paramagnetic phase is especially simple, since the problem can be reduced to a binary alloy, where the half of the sites are occupied by "up" moments and the other half by "down" moments.

2.2.3 Total energy calculations

In the Sec. 2.1 we have presented the equations that define the LSDA+DMFT scheme in terms of the local problem and a self consistent bath. These equations can be obtained by many different techniques [9], but in perspective of total energy calculations we have already adopted the point of view of the spectral density-functional theory of Savrasov and Kotliar [22]. In the case of the KKR implementation presented above the LSDA+DMFT total energy can be calculated from:

$$E_{LSDA+DMFT} = E_{LSDA}\left[\rho(\mathbf{r}), G(\omega)\right] - \langle H_U \rangle \, . \tag{13}$$

It should be noted that E_{LSDA}, in particular its band energy contribution, already contains the self-energy contribution and therefore the Galitskii-Migdal term has to be removed, since it is already added twice with the band energy.

The evaluation of Eq. (13) requires only the additional calculation of the Galitskii-Migdal energy, since the band energy results from the DFT part of the KKR code. While it might be simpler to evaluate the Galitskii-Migdal correction directly within the local problem (inside the DMFT solver integrating over Matsubara frequencies), we prefer to work again on the semicircular complex energy contour, retaining to the same formalism for both contributions of the total energy. This implies we calculate

$$\langle H_U \rangle = -\frac{1}{2\pi} \mathrm{Im} \sum_{\Lambda_1 \Lambda_2} \int dz \, \Sigma_{\Lambda_1 \Lambda_2}(z) G_{\Lambda_2 \Lambda_1}(z) \, . \tag{14}$$

The integration is performed over the contour starting close to the real energy axis at the bottom of a valence band and ending at the Fermi energy. It turned out that this procedure is numerically more stable than the evaluation of the Galitskii-Migdal correction using an integration over the Matsubara frequencies inside the DMFT solver.

2.2.4 Fully relativistic one-step model of photoemission for alloys

Spectroscopy is an extremely important experimental tool providing information on the electronic structure of the probed system that has to be seen as a stringent benchmark for the success of any electron structure theory. Photoemission spectroscopy (PES) or its inverse – the Bremsstrahlung isochromate spectroscopy (BIS) – in their angle-integrated form should reflect the density of states

(DOS) rather directly – in particular in the high photonenergy regime (XPS). For that reason it is quite common to check the DMFT-based calculations by comparing the calculated DOS directly to the PES spectra (see the reviews [13, 14, 63] for example).

However, this approach ignores the influence of the specific PES matrix elements that in general will introduce an element- and energy-dependent weight to the partial DOS. In case of angle-resolved photoemission (ARPES) the situation is even move severe as the surface as well as dipole selection rules may have a very pronounced impact on the spectra [64] demanding for a coherent description within the one-step model of photoemission [65]. To achieve a reliable interpretation of experiments, however, it is inevitable to deal with so-called matrix-element effects which considerably modify the raw spectrum. Above all, the wave-vector and energy dependence of the transition-matrix elements has to be accounted for. These issues are known to be important and actually cannot be neglected. They result from strong multiple-scattering processes which dominate the electron dynamics in the low-energy regime of typically 1-200 eV [66]. The transition-matrix elements also include the effects of selection rules which are not accounted for in the raw spectrum. Loosely speaking, it can be said that the main task of a theory of photoemission is to close the gap between the raw spectrum obtained by LSDA+DMFT electronic-structure calculations and the experiment. The most successful theoretical approach concerning this is the so-called one-step model of photoemission as originally proposed by Pendry and co-workers [66–68]. In the following a short overview will be given on the recent extensions of the one-step model which are connected with correlation effects and disordered alloys.

The main idea of the one-step model is to describe the actual excitation process, the transport of the photoelectron to the crystal surface as well as the escape into the vacuum [69] as a single quantum-mechanically coherent process including all multiple-scattering events. Within this model self-energy corrections, which give rise to damping in the quasi-particle spectrum, are properly included in both the initial and the final states. This for example allows for transitions into evanescent band gap states decaying exponentially into the solid. Similarly the assumption of a finite lifetime for the initial states gives the opportunity to calculate photoemission intensities from surface states and resonances. Treating the initial and final states within the fully relativistic version of layer KKR theory [70, 71], it is a straight forward task to describe complex layered structures like thin films and multilayers within the photoemission theory. Furthermore, the surface described by a barrier potential can be easily included into the multiple-scattering formalism as an additional layer. A realistic surface barrier model which shows the correct asymptotic behavior has been introduced, for example, by Rundgren and Malmström [72].

We start our considerations by a discussion of Pendry's formula for the photocurrent which defines the one-step model of PES [67]:

$$I^{\mathrm{PES}} \propto \mathrm{Im}\, \langle \epsilon_f, \mathbf{k}_{||} | G_2^+ \Delta G_1^+ \Delta^\dagger G_2^- | \epsilon_f, \mathbf{k}_{||} \rangle \, . \tag{15}$$

The expression can be derived from Fermi's golden rule for the transition probability per unit time [73]. Consequently, I^{PES} denotes the elastic part of the photocurrent. Vertex renormalizations are neglected. This excludes inelastic energy losses and corresponding quantum-mechanical interference terms [67, 73, 74]. Furthermore, the interaction of the outgoing photoelectron with the rest system is not taken into account. This "sudden approximation" is expected to be justified for not too small photon energies. We consider an energy-, angle- and spin-resolved photoemission experiment. The state of the photoelectron at the detector is written as $|\epsilon_f, \mathbf{k}_{\parallel}\rangle$, where $\mathbf{k}_{\parallel}$ is the component of the wave vector parallel to the surface, and ϵ_f is the kinetic energy of the photoelectron. The spin character of the photoelectron is implicit by included in $|\epsilon_f, \mathbf{k}_{\parallel}\rangle$ which is understood as a four-component Dirac spinor. The advanced Green function G_2^- in Eq. (15) characterizes the scattering properties of the material at the final-state energy $E_2 \equiv \epsilon_f$. Via $|\Psi_f\rangle = G_2^-|\epsilon_f, \mathbf{k}_{\parallel}\rangle$ all multiple-scattering corrections are formally included. For an appropriate description of the photoemission process we must ensure the correct asymptotic behavior of $\Psi_f(\mathbf{r})$ beyond the crystal surface, i. e. a single outgoing plane wave characterized by ϵ_f and $\mathbf{k}_{\parallel}$. Furthermore, the damping of the final state due to the imaginary part of the inner potential $iV_{0\mathrm{i}}(E_2)$ must be taken into account. We thus construct the final state within spin-polarized low-energy electron diffraction (SPLEED) theory considering a single plane wave $|\epsilon_f, \mathbf{k}_{\parallel}\rangle$ advancing onto the crystal surface. Using the standard layer-KKR method generalized for the relativistic case [65, 75], we first obtain the SPLEED state $-T\Psi_f(\mathbf{r})$. The final state is then given as the time-reversed SPLEED state ($T = -i\sigma_y K$ is the relativistic time inversion). Many-body effects are included phenomenologically in the SPLEED calculation, by using a parametrized, weakly energy-dependent and complex inner potential $V_0(E_2) = V_{0\mathrm{r}}(E_2) + iV_{0\mathrm{i}}(E_2)$ as usual [66]. This generalized inner potential takes into account inelastic corrections to the elastic photocurrent [73] as well as the actual (real) inner potential, which serves as a reference energy inside the solid with respect to the vacuum level [76]. Due to the finite imaginary part $iV_{0\mathrm{i}}(E_2)$, the flux of elastically scattered electrons is continuously reduced, and thus the amplitude of the high-energy wave field $\Psi_f(\mathbf{r})$ can be neglected beyond a certain distance from the surface.

For disordered alloys the CPA scheme can straightforwardly be extended to include the many-body correlation effects for disordered alloys [16]. According to the LSDA+DMFT approach realized in the framework of the fully relativistic SPR-KKR multiple scattering theory we use the self-energy $\Sigma_A^{DMFT}(E)$ calculated self consistently using dynamical mean field theory [9]. Within the KKR approach the local multi-orbital and energy dependent self-energies ($\Sigma_A^{DMFT}(E)$ and $\Sigma_B^{DMFT}(E)$) are directly included into the single-site matrices t_A and t_B, respectively by solving the corresponding Dirac Eq. (7). Consequently, all the relevant physical quantities connected with the Green's function and used for photoemission calculations, as for example, the matrix elements or scattering matrices like t^{CPA} contain the electronic correlations beyond the LSDA scheme. A detailed description of the generalized one-step model for disordered magnetic alloys can be found in [56]. Here we present the expression for the CPA photocurrent only. Following Durham *et al.* [77, 78] we have to perform the configurational average of these different contributions to the photocurrent, leading to the fully relativistic expression:

$$\begin{aligned} < I^{\mathrm{PES}}(\epsilon_f, \mathbf{k}_{\parallel}) > &= < I^{\mathrm{a}}(\epsilon_f, \mathbf{k}_{\parallel}) > + < I^{\mathrm{m}}(\epsilon_f, \mathbf{k}_{\parallel}) > \\ &+ < I^{\mathrm{s}}(\epsilon_f, \mathbf{k}_{\parallel}) > + < I^{\mathrm{inc}}(\epsilon_f, \mathbf{k}_{\parallel}) > \\ &. \end{aligned} \tag{16}$$

When dealing with disorder in the alloys, an additional term I^{inc}, the so called "incoherent" term appears. This contribution to the alloy photocurrent appears because the spectral function of an disordered alloy [37] is defined as a non single-site quantity. In fact this contribution is closely connected with the presence of the irregular wave functions well-known from the spherical representation of the Green function G_1^+.

3 Applications

In this section we review some results that have been obtained using the LSDA+DMFT as implemented in the full-potential KKR method (2.2). The use of a fully relativistic formulation on the basis of the four-component Dirac formalism allows to deal with all spin-orbit coupling induced properties like for example orbital magnetic moments of ferromagnetic transition metals. Magnetic moments and other ground state properties will be discussed in Sec. 3.1.1. Investigations on ground states properties are complemented by investigations on spectroscopic properties with an emphasis on valence band photoemission calculated on the basis of the so-called one-step model. In Sec. 3.2.1 the spin-orbit coupling induced Fano effect observed in angle-integrated photoemission is discussed. To achieve more stringent and detailed information about the electronic structure of correlated materials, the calculation of ARPES is highly desirable. As already mentioned our approach combines coherently LSDA+DMFT and ARPES relevant issues: the surface geometry, final state effects and the transition matrix elements accounting for the selection rules. Examples will be shown for the low index surfaces of transition metals like Ni or Fe in Sec. 3.2.2) and for the disordered NiPd-alloy system in Sec. 3.2.3. In the last Sec. 3.3 recent developments of the KKR formalism concerning $\vec{k}$ dependent self-energies will be discussed for the case of the electron-phonon coupling.

3.1 Ground state properties

3.1.1 Orbital magnetic moments

Various important properties of magnetic materials, like magnetic anisotropies, magneto-optical effects, and for example magnetic dichroism in various types of electronic spectroscopies are caused by spin-orbit coupling. While spin magnetic moments for $3d$ transition metals and their alloys are described rather accurately by the LSDA, the orbital magnetic moments are typically underestimated. To improve the description of orbital magnetism the so-called orbital polarization correction (OP) scheme was introduced by Brooks *et al.* [79–81] as an additional *ad hoc* term added to the LSDA Hamiltonian. Later on it had been realized that the OP term can be systematically derived from current DFT [82]. Despite of a quite accurate description of the orbital moments for bulk systems, calculations based on the OP-term fails to predict the experimental values for impurities [83–85] and clusters on surfaces [86].

An alternative approach is based on the explicit account of local (on-site) many-body correlations. In particular, Solovyev *et al.* [87, 88] have shown by calculations based on the random-phase approximation that the OP-picture is one of the limits of the more general LSDA+U concept [89]. On the other hand, the LSDA+U approach fails to give a proper description for the spectral properties of the $3d$ transition metals. Examples for these problems are found in the calculated bandwidths, the spin splitting, the energetics of the Ni satellite and in the absence of quasiparticle damping, etc. [58, 90]. On the other hand, the explicit account for local correlations within the LSDA+DMFT approach should improve the description of the orbital magnetic moments [54, 55] as well as of the spectral properties [24, 25, 48, 56, 91]. As an example LSDA+DMFT results for the ferromagnetic transition metals Fe, Co and Ni are shown in Fig. 2. It was found that the use of DMFT hardly

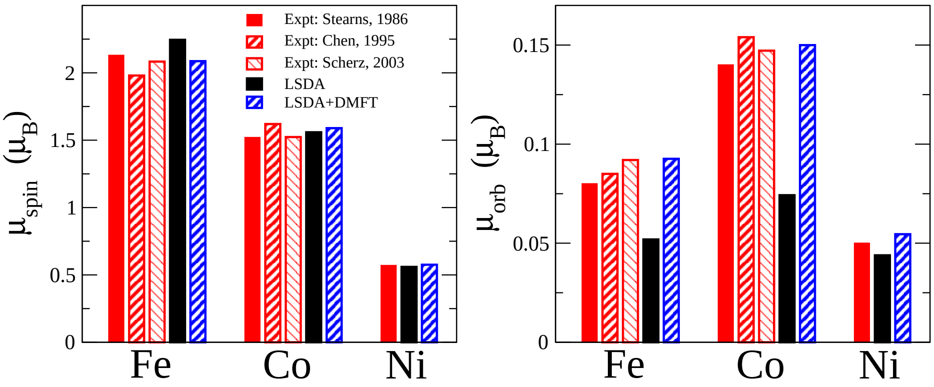


Figure 2: Spin (left panel) and orbital (right panel) magnetic moments in bcc Fe, hcp Co and fcc Ni calculated using LSDA+DMFT (hatched blue bars) compared with plain LSDA calculations (black filled bars) and experimental data (red bars).

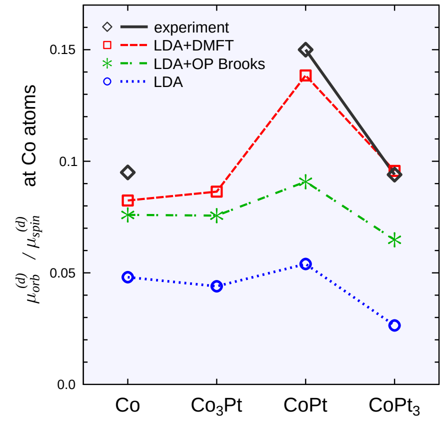
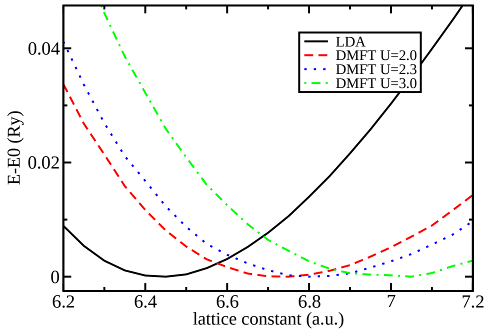


Figure 3: Ratios between the d components of μ_{orb} and μ_{spin} for Co in ordered CoPt obtained via various computational schemes compared with experiment [92].

changes the spin magnetic moments compared to plain LSDA-based calculations, that in turn give results already in very good agreement with experiment. Inclusion of the DMFT, on the other hand, increases the orbital moment and brings the theoretical values in very satisfying agreement with experiments. Concerning this a very important issue is the treatment of the double-counting term. A direct comparison of the calculated orbital magnetic moments with the corresponding experimental data clearly show that the AMF setting is most adequate for metallic transition metal systems. By construction the dynamical part of the self-energy Σ^{DMFT} behaves in the vicinity of the Fermi level as for a Fermi liquid. Therefore Σ^{DMFT} cannot noticeably affect integral quantities like spin and orbital magnetic moments. On the other hand, the applied AMF static double counting which splits the different orbitals only slightly at the Fermi level, has no impact on the renormalization of the density of states. As already mentioned, the application of the OP term for diluted magnetic moment systems is not much successful. Because the LSAD+DMFT method leads to more accurate results in this case, we performed LSDA+DMFT calculations for various CoPt ordered phases [55]. In Fig. 3 we show the *ratios* between the orbital and spin magnetic moments, which can be deduced from x-ray magnetic circular dichroism (XMCD) measurements. It follows from Fig. 3 that the LSDA systematically underestimates $\mu_{\text{orb}}^{(d)}/\mu_{\text{spin}}^{(d)}$ for Co in various



		EXP	LSDA	$U = 2.0$ eV
V_0(a.u.^{3})	FP-LMTO	73.52	67.88	76.20
	KKR		66.86	76.28
B(GPa)	FP-LMTO	186	260	163
	KKR		280	171

Figure 4: Left: Total energy of Ni calculated by means of the LSDA and LSDA+DMFT as a function of the lattice parameter a for various values of the parameter U. Right: Computed values of the equilibrium atomic volume V_0 and the bulk modulus B for the the standard LSDA-DFT method and for the LSDA+DMFT scheme in comparison with experiment [31].

compounds. This deficiency increases with increasing Pt content: the experimental $\mu^{(d)}_{\text{orb}}/\mu^{(d)}_{\text{spin}}$ ratio exceeds the LSDA result twice for bulk Co, three times for CoPt and four times for CoPt$_3$. Employing the OP scheme of Brooks increases the $\mu^{(d)}_{\text{orb}}/\mu^{(d)}_{\text{spin}}$ ratio and partially compensates for the deficiency of the LSDA. This compensation is nearly complete for bulk Co but it becomes insufficient as the Pt content increases. Employing the LSDA+DMFT, on the other hand, leads to a nearly perfect reproduction of the experimental $\mu^{(d)}_{\text{orb}}/\mu^{(d)}_{\text{spin}}$ ratio at Co atoms for bulk Co, CoPt, as well as CoPt$_3$.

3.1.2 Total energy calculations

As already mentioned, most of the LSDA+DMFT applications up to now are connected with spectral function calculations and related properties. However the predictive power of any LSDA+DMFT approach can be achieved only by an accurate description of the total energies and related phenomena like forces or lattice relaxations. A step towards investigations on the equilibrium structure of correlated systems, which are based on total energy calculations in the framework of the full potential KKR implementation of the LSDA+DMFT method, is found in [31]. Results obtained for ferromagnetic Ni are presented in Fig. 4. It is well known that in the framework of LSDA the equilibrium value of the lattice constant is slightly underestimated (typically by 3%) with respect to the experimental value. Concerning the calculations based on the LSDA+DMFT, one notices that the results are strongly dependent on the value of the Hubbard U. The best results are obtained for $U = 3$eV, i.e. for a value which is smaller as commonly accepted. This discrepancy can be assigned to the perturbative nature of the SPTF solver [31, 57]. However, for $U = 3$eV we obtained a reasonable agreement also for the bulk modulus. In the case of Ni a satisfying result can also be obtained by using gradient corrected functionals (GGA) [5]. On the other hand, in the case of γ-Mn which is considered to be a material on the border between moderate and strong correlations [59, 93] the change of the exchange correlation potential from LSDA to GGA does not solve all the problems and it remains a significant difference between theory and experiment. The atomic volume is underestimated and the anomalously low value of the bulk modulus ($B = 90 - 130$GPa [94, 95]) is strongly overestimated by LSDA. This problem can be solved by

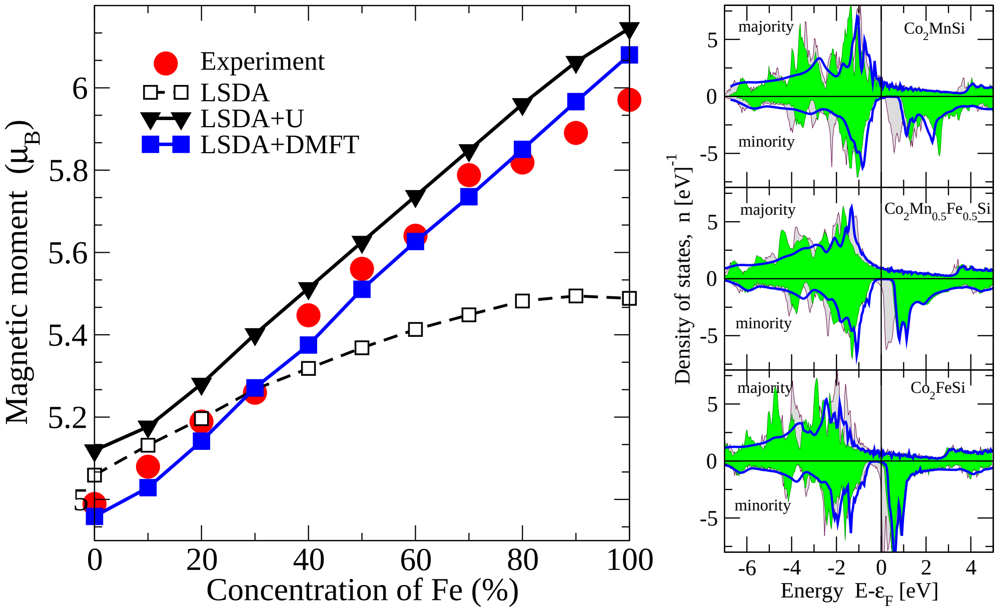


Figure 5: Left: Comparison of the total magnetic moments for $Co_2Mn_xFe_{1-x}Si$ compounds calculated within LSDA (black dashed line, opened squares), LSDA+U (black triangles) and LSDA+DMFT (blue squares) with the results of the SQUID magnetic measurements (red circles) [98]. Right: The total spin-resolved DOS curves for Co_2MnSi, $Co_2Mn_{0.5}Fe_{0.5}Si$ and Co_2FeSi calculated within LSDA (light/grey filled area), LSDA+U (dark/green filled area) and LSDA+DMFT (blue line). ($U_{Co,Mn,Fe} = 3.0$eV and $J_{Co,Mn,Fe} = 0.9$eV)

the LSDA+DMFT [31]. Interestingly a combination of the GGA and the DMFT in general leads only to moderately changes compared to plain GGA results, therefore demonstrates the general applicability of the LSDA+DMFT.

3.1.3 LSDA+DMFT for disordered alloys

Representing the electronic structure in terms of the Green's function allows for a straightforward combination of the LSDA+DMFT method with the CPA alloy theory, as both schemes are used up to now on a single site-level. This combined approach was applied to study the magnetic properties of a number of disordered substitutional transition metal alloys [16, 54, 56, 96]. An interesting class of materials for magneto-electronic applications is found in the family of half-metallic Heusler ferromagnets [97]. These materials show, for example, interesting properties connected with correlation effects, like as for example so called non-quasi particle states [23]. Special attention had been paid to Co_2MnSi, Co_2FeSi and to the substitutional series $Co_2Mn_xFe_{1-x}Si$ of Heusler half-metals which show a large minority band gap (of about 0.4eV) and a very high Curie temperature of about 1000K [98]. In Fig. 5 we present the total magnetic moments of the substitutional disordered alloy $Co_2Mn_xFe_{1-x}Si$. Our calculations show a monotonous increase with Fe content in the magnetic moment in analogy to the Slater-Pauling curve within the whole range of Fe concentrations. It turned out that the static treatment of correlation effects (on the level of LSDA+U) is important for a reliable description of the minority band gap [99] for this class of materials. The additional dynamical effect appearing within LSDA+DMFT only slightly reduces the spin magnetic moment.

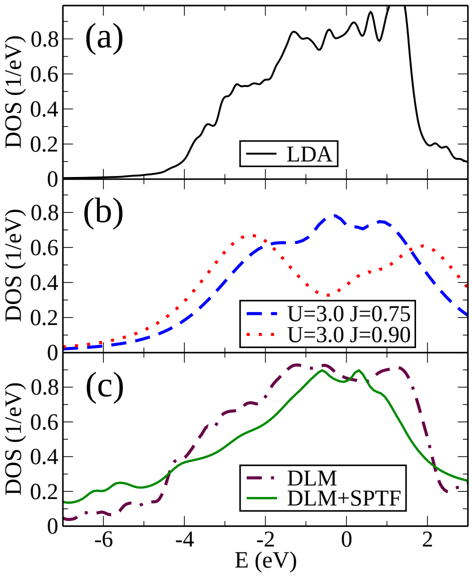


Figure 6: Comparison of the density of states of the $3d$ electrons in γ-Mn obtained by different approaches: (a) DFT-LDA (b) QMC with $U = 3.0$eV and different values of J. (c) DLM and DLM+SPTF for $U = 3.0$eV and $J = 0.8$eV.

The dynamical correlations appear to be more significant for the spectroscopic properties. As it follows from the right panel of Fig. 5, the LSDA+U enlarges the minority spin gap. Adding the DMFT causes a shift of the d-states towards the Fermi level and a substantial broadening of the DOS within the range between -2 and -8eV is observed. This effect can be seen directly in the angle-integrated valence band photoemission spectra [96].

Another interesting application of the CPA is to deal with thermal fluctuations of magnetic moments by use of the so called disordered local moment model (see Sec. 2.2.2). Its combination with the DMFT was recently demonstrated by a corresponding application to γ-Mn [59]. Here, it turned out that instead of the cumbersome and computationally expensive QMC calculations, the proposed combination of SPTF and the DLM approach can be used to describe adequately the high temperature properties of γ-Mn. In Fig. 6 a comparison of the densities of states of $3d$ electrons in γ-Mn obtained by different computational schemes is presented. The upper panel (a) and the middle panel (b) display the bare LDA results and the LDA+DMFT results with the QMC solver for $U = 3\,\mathrm{eV}$ and $J = 0.75\,\mathrm{eV}$ or $J = 0.9\,\mathrm{eV}$. In the lower panel (c) the density of states obtained within the DLM approach is reported, both for LSDA and LSDA+DMFT by use of the SPTF solver ("DLM+SPTF"). As the bare DLM describes the fluctuations in a very simple way, only a weak indication of three peak structures is observed. However the width of the $3d$-band is too large, since it derives from the single particle LDA density of states. On the other hand, in DLM+SPTF we can properly describe the shrinking of the $3d$ band, as already observed for Fe, Co and Ni [25, 58]. In comparison with the QMC calculations, the peak around $-2\,\mathrm{eV}$ is less pronounced. This is due to the perturbative nature of SPTF, which tends to shift the correlation effects related to the formation of non-coherent satellites. For example, the famous $-6\,\mathrm{eV}$ satellite of Ni is found at about $-8\,\mathrm{eV}$ [24]. In the case of γ-Mn the spectral weight is transferred to the region between -4 and $-6\,\mathrm{eV}$. In conclusion, it seems that the DLM+SPTF can reproduce QMC results for the moderately correlated regime. But, as it is shown in panel

(b) of Fig. 6 only a slight change of the parameter J (from 0.75 to 0.9eV) drives the γ-Mn into the strong correlation regime. This is because of the appearance of the Hubbard band. In fact, this tendency also strongly changes the temperature dependence of the magnetic moment. For the smaller value of J (moderately correlated system) spin fluctuations are reduced because of the presence of strong orbital fluctuations. For the higher value of J (strongly correlated limit) a strong fluctuating local moment is formed. This transition happens close to the physical value of Hund's exchange J. These different tendencies had been studied before in the two-orbital Hubbard model [100], but it is interesting to see that they can be quantitatively described in a real material with five orbitals and realistic hybridizations.

3.2 LSDA+DMFT for calculations of spectroscopic properties

3.2.1 Angle-integrated valence band photoemission: Fano effect

Spin-orbit coupling gives rise to many interesting phenomena in the electron spectroscopy of magnetic solids. A rather straightforward access to the understanding of these phenomena is provided by the study of the Fano effect. This effect was predicted by Fano at the end of the sixties and denotes the fact that one obtains a spin-polarized photoelectron current even for non-magnetic systems if the excitation is done using circularly polarized light [101]. At which, for non-magnetic samples the spin polarization of the photocurrent is reversed, this symmetry is in general broken for magnetically ordered systems leading to magnetic circular dichroism. However, in the case of magnetic materials, the spin polarization is usually due to the interplay between spin-orbit coupling and exchange splitting. Recently, we demonstrated by investigations on Fe, Co and Ni that the pure Fano effect can also be observed in angle-integrated valence band XPS (VB-XPS) for ferromagnets if circularly polarized light impinges perpendicular to the magnetization direction and if a subsequent spin analysis is done with respect to the direction of the photon beam [24]. This is demonstrated in Fig. 7 where the VB-XPS of the Fe, Co and Ni at a photon energy of 600eV is shown. The photon energy of 600eV has been used in order to increase the bulk sensitivity of the photoemission process. In the upper panel of Fig. 7 we compare experimental data with corresponding LSDA and LSDA+DMFT VB-XPS data based on the one-step model of photoemission. In all three cases the LSDA+DMFT considerably improves the agreement with the experiment. In particular, in the case of Ni LSDA+DMFT leads to the shrinking of the d-band width. In addition, use of the LSDA+DMFT scheme leads to a pronounced increase of the intensity in the regime of the 6eV satellite. For the total intensity of the Fe and Co signal we observed an pronounced improvement in the energy region from -2 to -8eV. A decomposition of the theoretical spectrum according to the angular momentum character of the initial state shows that the d-contribution is far dominating and that the spectrum essentially maps the corresponding DOS. This, in some sense, supports the common practice of comparing experimental XPS directly with the DOS. In the lower panel of Fig. 7 the corresponding spin difference $\Delta I^+ = I^+_\uparrow - I^+_\downarrow$ (i.e. the difference of the currents of photoelectrons with spin-up and spin-down electrons, for excitation with left circularly polarized radiation) is shown. The occurrence of this spin current is a pure matrix element effect induced by spin-orbit coupling. In fact, one finds that the shape of the ΔI^+ curves are very similar to those that can be found for non-magnetic Nobel metals [102, 103]. In fact, the amplitudes scale with the spin-orbit coupling parameter of the Fe, Co and Ni d-states. To achieve this rather good

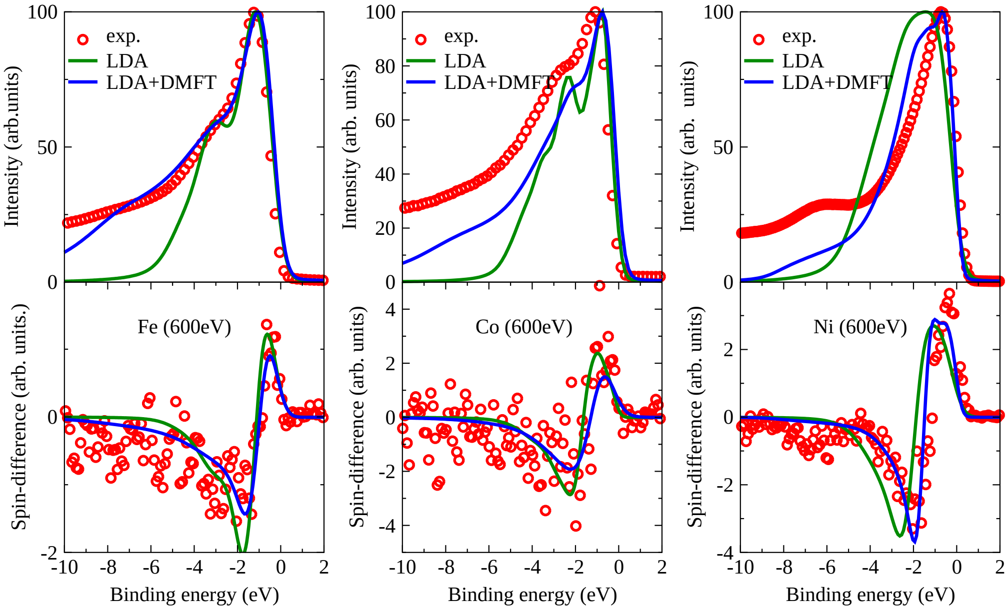


Figure 7: Top panel: The experimental (dots), LSDA (green line) and LSDA+DMFT (blue line) angle integrated valence band XPS spectra of bcc Fe, hcp Co and fcc Ni for a photon energy of 600eV . Lower panel: Spin difference $\Delta I^{+} = I^{+}_{\uparrow} - I^{+}_{\downarrow}$ of the photocurrent for excitation with left circularly polarized light.

agreement with the experimental data for the ΔI^{+} intensity distribution the fully self consistent LSDA+DMFT approach is obviously needed.

3.2.2 Angle-resolved photoemission within one-step model

In the previous Sec. 3.2.1 we showed angle-integrated XPS spectra which can be directly compared to the DOS ignoring matrix element effects. However, the most complete description of the band structure of correlated materials can be obtained by spin- and angle-resolved valence band photoemission. In the following section we present various examples of angle-resolved photoemission calculations done within the one-step model. These examples clearly demonstrate the need for such calculations in order to obtain a quantitative understanding of the corresponding experimental data.

The following examples concern the ferromagnetic transition metal systems Ni and Fe as prototype materials to study electronic correlations and magnetism beyond the LSDA scheme. In Fig. 8 we present a comparison between experimental photoemission data [64] and calculated spectra using different theoretical approaches [25]. In the upper row spin-integrated ARPES measurements from Ni(011) along $\overline{\Gamma Y}$ for different angles of emission are shown. The dotted lines represent the experimental data, whereas the solid lines denote the single-particle approach to the measured spectral function. Obviously, the LSDA-based calculation completely fails to describe the experimental data. The energetic positions of the theoretical peaks deviate strongly from the measured ones. Furthermore, the complicated intensity distributions that appear for higher emission angles are not accounted for by the LSDA-based calculations. In contrast, the non self consistent quasi-particle 3BS calculation provides a significant improvement when compared to the measured spectra. For

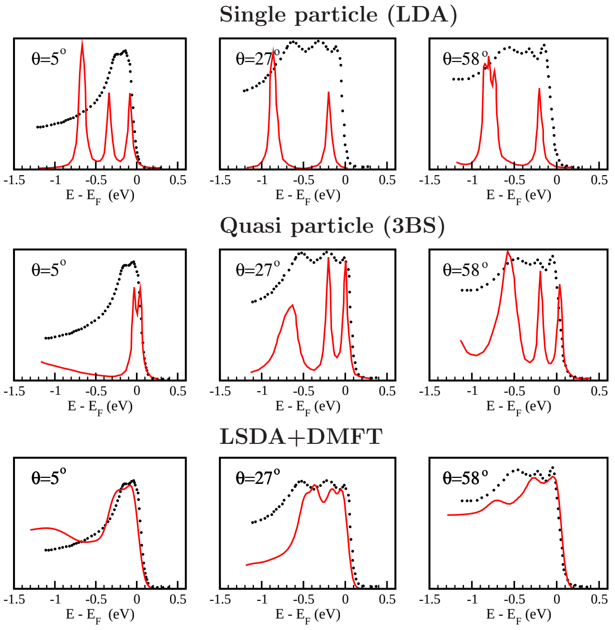


Figure 8: Spin-integrated ARPES spectra from Ni(011) along $\overline{\Gamma}Y$ for three different angles of emission. Upper row: comparison between LSDA-based calculation and experiment [64]; middle row: comparison between experiment and non self consistent quasi-particle calculations neglecting matrix element and surface effects [64]; lower row: spin-integrated LSDA+DMFT spectra including photoemission matrix elements (this work). Theory: solid red line, experiment: black dots.

the complete range of emission angles the energetic peak positions coincidence with the experiment within about 0.1 eV. Only the overall shape of the measured spectral intensities deviate from the calculations because of the neglect of multiple scattering and surface-related as well as matrix-element effects. In the experiment the various peaks seem to be more broadened and the spectral weight especially for nearly normal emission is shifted by about 0.1 eV to higher binding energies. In addition it seems that for very high emission angles like 60° an even more complicated peak structure is hidden due to limited experimental resolution. An additional spin-analysis is therefore highly desirable for these experiments.

Within our work we could go far beyond previous theoretical studies by combining a sophisticated many-body approach as the self consistent LSDA+DMFT method with the one-step based calculation of the corresponding spectral function. The self-energy within the DMFT has been applied only for d-states and can be calculated in terms of two parameters - the averaged screened Coulomb interaction U and the exchange interaction J. The screening of the exchange interaction is usually small and the value of J can be calculated directly and is approximately equal to 0.9 eV for all $3d$ elements. This value has been adopted for all our calculations presented here. For U we used a value of 3 eV. However, our test calculations showed that a somewhat different choice of U does not substantially changes the various features and trends in the calculated spectra. The intensity distributions resulting from the corresponding photoemission calculation are shown in the lower row of Fig. 8. A first inspection reveals very satisfying quantitative agreement between experiment and theory for all emission angles. Let us concentrate first on the excitation spectrum calculated for the emission angle $\Theta = 5°$. The spin-integrated spectrum exhibits a pronounced double-peak structure with binding energies of 0.1 eV and 0.3 eV. The second peak is slightly reduced in intensity

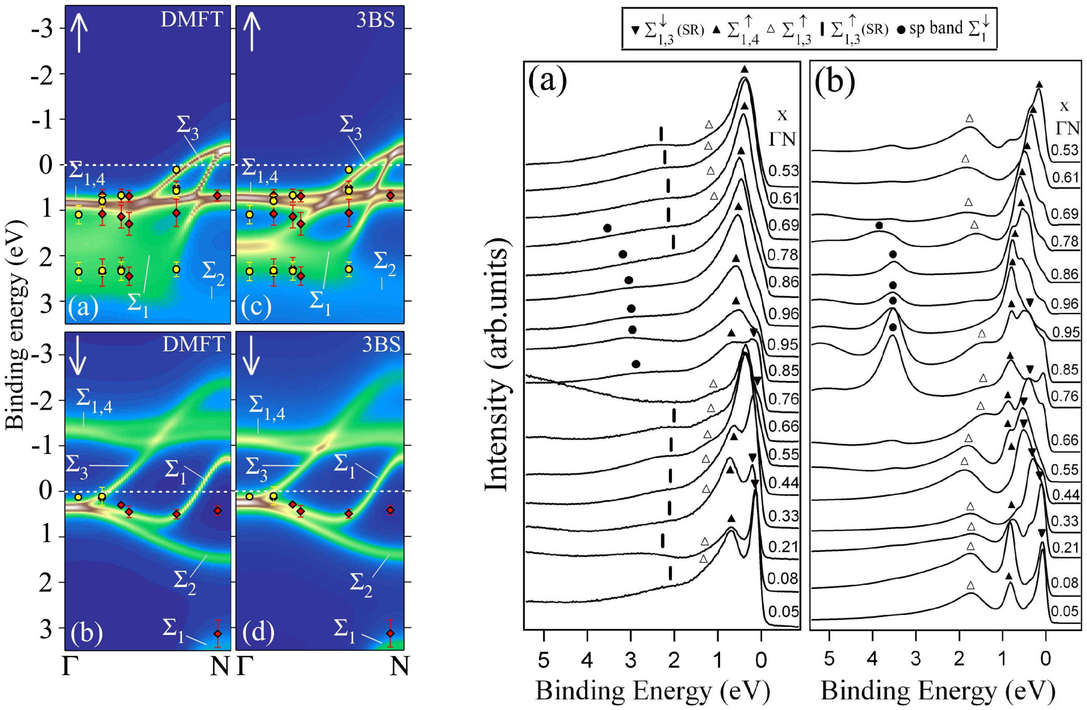


Figure 9: Left panel: Spin resolved Bloch spectral functions calculated within LSDA+DMFT and 3BS formalism . Corresponding experimental data points have been deduced from the normal emission spectra along the ΓN direction. Right panel: (a) Experimental spin-integrated photoemission spectra of the Fe(110) surface measured with p-polarization in normal emission along the ΓN direction of the bulk Brillouin zone. The curves are labeled by the wave vectors in units of ΓN=1.55 $\AA^{-1}$. (b) Corresponding one-step model calculations based on the LSDA+DMFT method which include correlations, matrix elements and surface effects.

which is also in accordance with the experimental findings. Furthermore, the width of the spectral distribution is quantitatively reproduced. The calculated binding energies are related to the real part of the self-energy that corrects the peak positions due to a dynamical renormalization of the quasiparticles which is missing in a typical LSDA-based calculation. The relative intensities of the different peaks, on the other hand, must be attributed to the matrix-element effects which enter our calculations from the very beginning via the one-step model of photoemission. The double-peak structure originates from excitation of the spin-split d-bands in combination with a significant amount of surface-state emission [104]. The two spectra calculated for high emission angles show the more broadened spectral distributions observed in experiment. An explanation can be given in terms of matrix-element effects, due to the dominating dipole selection rules. The spin-resolved spectra reveal a variety of d-band excitations in both spin channels, which in consequence lead to the complicated shape of the spectral distributions hardly to be identified in the spin-integrated mode.

The second example within this section concerns a spectroscopic study of ferromagnetic Fe [48]. In the left panel of Fig. 9 we compare the experimental peak positions from bulk like transitions with spin-resolved LSDA+DMFT spectral functions. In addition to these investigations we accounted for correlation effects within the 3BS approach [12]. Within the 3BS approach the self-energy is calculated using a configuration interaction-like expansion. In particular three-particle configurations like one hole plus one electron-hole pair are explicitly taken into account within 3BS-based calculations. The corresponding output can be directly related to the photoemission process and allows for a detailed analysis of various contributions to the self-energy (e.g., electron-hole lifetime). A more detailed quantitative comparison is shown in right panel of Fig. 9. Here we display a comparison between spin-integrated ARPES data and theoretical LSDA+DMFT based one-step photoemission calculations of Fe(110) along the ΓN direction of the bulk Brillouin zone (BZ) with p-polarized radiation. In our LSDA+DMFT investigation underlying the ARPES calculations we use for the averaged on-site Coulomb interaction U a value U=1.5 eV which lies between the experimental value U ≈1 eV [105] and a value U ≈2 eV obtained from theoretical studies [36, 54]. The k values associated with the spectra were calculated from the used photon energies ranging from 25 to 100 eV. Near the Γ point (k∼0.06 ΓN), the intense peak close to the Fermi level corresponds to a $\Sigma^{\downarrow}_{1,3}$ minority surface resonance, as indicated on top of Fig. 9. Experimentally, its $\Sigma^{\downarrow}_{3}$ bulk component crosses the Fermi level at k ∼0.33 ΓN, leading to a reversal of the measured spin-polarization and to a strong reduction of the intensity at k =0.68 ΓN in the minority channel. The peak at the binding energy BE∼0.7 eV, visible mainly for p-polarization in a large range of wave vectors between Γ and N, can be assigned to almost degenerate $\Sigma^{\uparrow}_{1,4}$ bulk-like majority states. A $\Sigma^{\uparrow}_{3}$ feature at BE∼1.1 eV dominates the spectrum close to the Γ-point. Depending on the polarization the degenerate $\Sigma^{\uparrow}_{1}$ states form a shoulder around the same BE. The broad feature around 2.2 eV, visible at various k-points, but not at the N-point, is related to a majority $\Sigma^{\uparrow}_{1,3}$ surface state. Around the N-point (0.76≤ k ≤1.0) and at BE≥3 eV we observe a $\Sigma^{\downarrow}_{1}$ band having strong sp character. The pronounced difference between its theoretical and experimental intensity distributions can be attributed to the fact that in the present calculations only the local Coulomb repulsion between d electrons is considered, without additional lifetime effects for the sp bands. Finally, we notice that the background intensity of the spectrum at k=0.66 ΓN, corresponding to a photon energy of 55 eV, is strongly increased due to the appearance of the Fe 3p resonance. The direct comparison of the calculated and experimental spectra turned out to be a very stringent check of Coulomb parameter U used in the calculations. This also applies to the DMFT self-energy, that was compared to its counterpart deduced from the experimental band dispersion and line width.

In conclusion, we have presented spectral function calculations of ferromagnetic Ni and Fe, which coherently combine an improved description of electronic correlations, multiple-scattering, surface emission, dipole selection rules and other matrix-element related effects that lead to a modification of the relative photoemission intensities. A similar study has been recently performed for Co(0001) [49]. This approach allows on the one hand side a detailed and reliable interpretation of high-resolution angle-resolved photoemission spectra of 3d-ferromagnets. On the other hand, it also allows for a very stringent test of new developments in the field of DMFT and similar many-body techniques.

3.2.3 One-step model of photoemission of disordered Ni$_x$Pd$_{1-x}$(001) alloy

In this section alloying effects in combination with electronic correlations are considered [56]. Fig. 10 shows a series of spectra of Ni$_x$Pd$_{1-x}$ as a function of the concentration x calculated for a

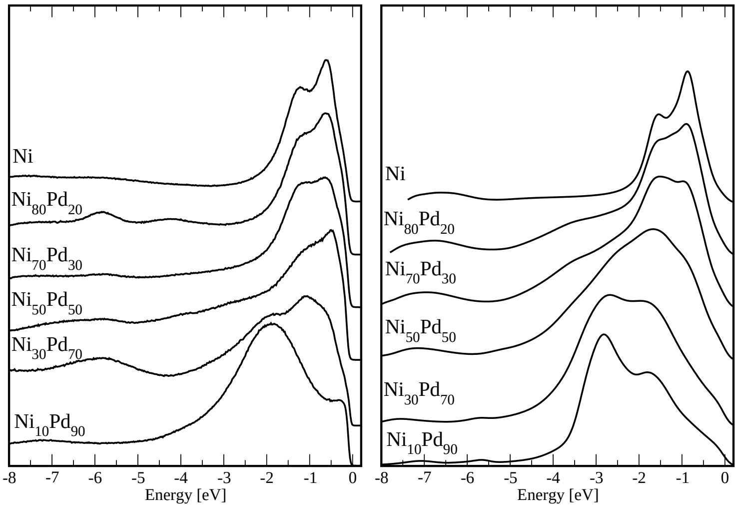


Figure 10: ARPES spectra taken from the Ni$_x$Pd$_{1-x}$(001) alloy surfaces as a function of the concentration x for a fixed photon energy of $h\nu$=40.0 eV along ΓX in normal emission. Experimental data shown in the left panel calculated spectra presented in the right panel. Depending on the concentration x a pronounced shift in spectral weight towards the Fermi level is visible.

photon energy $h\nu$=40 eV with linear polarized light. The experimental data are shown in the left panel and the corresponding LSDA+DMFT-based photoemission calculations are presented in the right one. Our theoretical analysis shows that starting from the pure Ni, the agreement is fully quantitative with deviations less than 0.1 eV binding energy, as expected. Going to the Ni$_{0.80}$Pd$_{0.20}$ alloy the agreement is comparable good for binding energies between the Fermi energy and 2 eV. Inspecting the density of states (DOS) for the Ni$_{0.80}$Pd$_{0.20}$ alloy this fact becomes explainable, because this energy interval represents the Ni-dominated region. The Pd derived states start to appear at about 2 eV below E_{F} besides of the small dip at the Fermi level. For higher binding energies the agreement is also very good, although a bit more structure is observable in the theory, especially around 3.5 eV. An explanation for this behavior can be found in terms of lifetime effects, but it should be mentioned here that background in the experimental spectra due to secondary electrons was not considered in the theoretical analysis. From the Ni$_{0.70}$Pd$_{0.30}$ alloy system it becomes clearly visible that the deviation between theory and experiment is mainly introduced for increasing concentration of Pd. This can be seen from the spectra for Ni$_{0.50}$Pd$_{0.50}$ and Ni$_{0.30}$Pd$_{0.70}$ alloys shown next in the series. In addition, the spectra of Ni$_{0.30}$Pd$_{0.70}$ reveal some deviations near the Fermi level. Also, the spectral intensity of the Ni surface resonance, that appears at about 0.5 eV binding energy is underestimated in the calculation when compared to the experiment.

Our spectroscopic analysis has clearly demonstrated that the electronic properties of the Ni$_x$Pd$_{1-x}$ alloy system depend very sensitively on the interplay of alloying and electronic correlation. A description within the LSDA approach in combination with the CPA results in a quantitative description of the electronic structure of Ni$_x$Pd$_{1-x}$ [56]. This example may illustrate that the use of the CPA alloy theory self consistently combined with LSDA+DMFT approach serves as a powerful tool for electronic structure calculations, whereas the application of the fully relativistic one-step model of photoemission, which takes into account chemical disorder and electronic correlation on equal footing guarantees a quantitative analysis of the corresponding spectroscopic data.

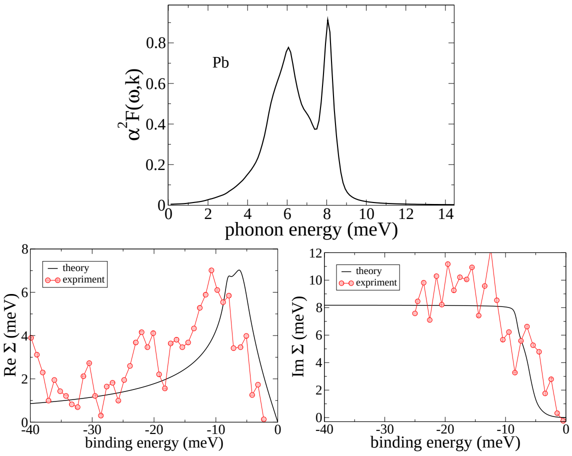


Figure 11: Upper panel: Calculated Eliashberg function $\alpha^2 F_k(\omega)$ for Pb(110) for $\mathbf{k} = \frac{\pi}{a}(0.17, 1., 0.17)$. Lower panel: corresponding k-dependent self-energy, real part shown on left, imaginary part shown on the right.

3.3 Effects of electron-phonon interaction in angle-resolved photoemission

In the following section we want to demonstrate that the formalism presented in Sec. 2.2 can be extended in order to include a $\vec{k}$-dependent self-energy. In detail, we discuss the effect of electron-phonon coupling on the angle-resolved photoemission of Pb(110).

Nowadays, high resolution photoemission measurements allow to investigate in great detail the electronic structure of materials close to the Fermi level. This gives access to energy bands modified by electron-phonon interaction, and therefore serves as a very important spectroscopic tool to characterize transport properties or, for example, properties of super-conducting materials [106–109]. Modifications of the electronic structure generated by electron-phonon interaction are typically not accounted for in LSDA-based band-structure calculations. The theoretical treatment of electron-phonon effects requires the use of many-body techniques which originally had been developed to give a quantitative description of strong electronic correlation effects. The self-energy approach often applied in many-body calculations can be used as well to represent the electron-phonon interaction. Accordingly, having the electronic structure obtained by an ab-initio calculation, the modifications due to electron-phonon interaction can be described via an electron-phonon self-energy $\Sigma(\mathbf{k}, E_k)$ [110, 111]. The electron-phonon self-energy is derived using many-body perturbation theory within second order and is expressed in terms of the momentum-resolved Eliashberg function $\alpha^2 F_k(\omega)$. To obtain explicitly the electron-phonon self-energy, we focus our attention on the Eliashberg function as the main quantity determined by all coupled electronic and phononic states of the system. This function can efficiently be reformulated in terms of the Green's function [112, 113] and accordingly by calculated directly using KKR formalism. For this propose the phonon dispersions and corresponding matrix elements have been calculated within the real space KKR formulation of the rigid sphere approximation.

The complex self-energy in general consists of contributions due to electron-electron and electron-impurity scattering, as well as due to electron-phonon scattering (Σ_{el-ph}). The real part of Σ_{el-ph} is responsible for band renormalization around E_{F}. On the other hand the imaginary part part of

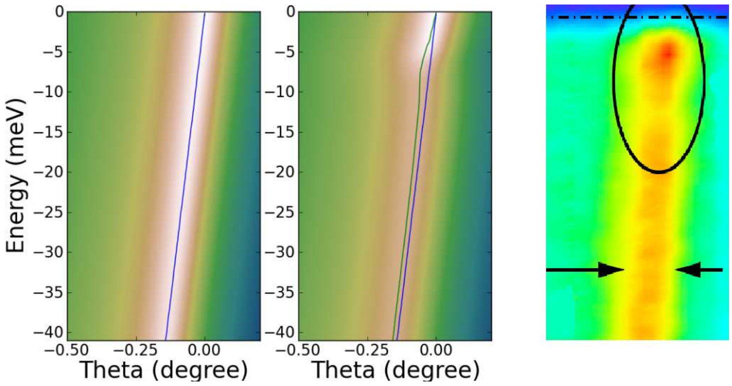


Figure 12: ARPES spectra taken from the Pb(110) surface excluding (left panel) and including (middle panel) phonons via a corresponding electron-phonon self-energy Σ_{el-ph}. Corresponding experimental data are shown on the right panel [115].

Σ_{el-ph} describes lifetime effects. These quantities are determined by electron-phonon scattering induced quasiparticle lifetimes, measurable in photoemission experiments. To demonstrate the effect of electron-phonon interactions on the electronic quasi-particle states near E_{F} we present photoemission calculations for Pb, which serves as a prototype system and compare our theoretical results to corresponding experimental data [114]. Pb is of special interest because of the presence of strong electron-phonon interactions which in particular lead to the superconducting state at low temperatures [106, 109].

The calculations have been performed for energies close to the Fermi level. The wave vectors in the BZ ($\mathbf{k} = \frac{\pi}{a}(0.17, 1., 0.17)$) correspond to the experimental geometry [109]. Fig. 11 shows a comparison between the experimental and theoretical real and imaginary parts of the self-energy Σ_k. Obviously, the agreement is very satisfying. In general, it is possible to deduce a phononic self-energy from measured photoemission spectra. The imaginary part of the self-energy can be estimated from the full width at half maximum of the energy distribution curves fitted by a Lorentzian. The real part is then obtained by a Kramers-Kronnig transformation. However, in cases where for example several transitions appear close to the Fermi level or even additional surface contributions to the photocurrent take place, this analysis might be questionable. Therefore, it seems to be more reliable to make a direct comparison between angle-resolved photoemission calculations and corresponding experimental data. For this purpose we developed a scheme that allows to include directly a $\mathbf{k}$-dependent self-energy into the one-step model of photoemission. In Fig. 12 we present photoemission spectra calculated for the two cases without (left panel) and with (middle panel) electron-phonon interactions and compare them to the corresponding measurements [115]. The contour plot has been calculated for HeI radiation along $\bar{\Gamma}\bar{\mathrm{X}}$. More precisely, bands forming the locally tubular Fermi surface of Pb(110) crosses E$_F$ at about 5° off-normal emission along $\bar{\Gamma}\bar{\mathrm{X}}$. In the middle panel of Fig. 12 a clear renormalization of the bare band (often called *kink*) at 8meV binding energy is observable. This effect appears just where the real part of the self-energy reaches its maximum. Furthermore, an additional broadening of the spectral features due to the imaginary part of the phononic self-energy is visible in the calculated spectra. This, of course, completes a lifetime analysis of spectral features in a quantitative sense because the imaginary part of the phononic self-energy can be identified as the last contribution that has to be considered in addition to impurity scattering and electronic correlation related broadening mechanisms of photoemission intensities. As an outlook the above scheme can straightforwardly be extended to combine the electron-phonon interaction with LSDA+DMFT calculations. This will allow a electron-phonon interaction studies of the correlated materials like for example fcc-Ni [116].

4 Summary and concluding remarks

In summary, we have reviewed a LSDA+DMFT implementation within the KKR method. In particular this approach was chosen because the KKR method represents the electronic structure by the corresponding single-particle Green's function leading therefore to a number of very attractive features of the scheme. The fully-self consistent full-potential implementation turned out to be a very powerful tool to study ground state as well as spectroscopic properties of transition metals, alloys and their surfaces. The fully-relativistic formulation allows to study spin-orbit coupling induced properties as, e.g. orbital magnetic moments or magnetic dichroic phenomena in photoemission. The TMA and SPTF solver used so far clearly imposes a restriction to moderately correlated materials. To get access to strongly correlated systems the adaption of QMC based solvers to the KKR implementation is needed.

Of course, the LSDA+DMFT method in its standard form is not an *ab-initio* method mostly due to use of the Coulomb parameter U. Various schemes have been proposed to calculate U like the constrained random phase approximation or constrained LSDA. The scheme developed by Cococcioni et al. [36] seems to be especially attractive, as it might be applicable without modifications to surfaces and nano-structures within the embedding KKR technique by avoiding the super-cell approach. With reliable values for the Coulomb parameter U the KKR implementation of the LSDA+DMFT will open the way to investigate the impact of correlation effects on structural and volume dependent properties in a quantitative way. In particular systems with reduced dimensionality, like 2D surfaces or nano-structures deposited on surfaces can be straightforwardly calculated within the KKR implementation of LSDA+DMFT. Furthermore, as it has been demonstrated, a $\vec{k}$-dependent self-energy can be included into the KKR method. This allows to discuss non-local correlation effects, for example, on the basis of cluster DMFT.

All these developments can be directly tested by a direct comparison with the corresponding experiments like i.e. angle-resolved photoemission, magnetic Compton profiles or magneto optics. Finally, developments connected with resonant and time-dependent spectroscopy become more and more important to gain detailed insights into the physics of correlated materials.

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