HCM Newsletter

Ψ_k Network

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

Number 23 October 1997

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Contract: ERBCHRXCT930369
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Contents

1 Editorial

As can be seen from the content, in this Newsletter we have a report, including abstracts of presented papers, on the '**GMR Workshop**' of the TMR Network on *'Interface Magnetism'*. The spectroscopies workshop of the latter Network, planned for 21-23 November, has been postponed until spring 1998. Thus the TMR Network plans to organize the following activities in 1998: workshops on spectroscopies and TB-LMTO, and an annual general meeting. Please keep on reading the Newsletters for further announcements.

In the section presenting reports of collaborative visits we have a report by J. Goss et al. in the form of a small scientific paper, presenting results of his one month secondment to Aveiro in Portugal. In the **Highlight of the Month** section readers will find an interesting article by T. Korhonen, N. Papanikolaou, R. Zeller, P.H. Dederichs, and N. Stefanou on **"Forces and Lattice Relaxations Calculated by a Full-Potential KKR-Green's Function Method**. We would also like to turn reader's attention to the WEB announcement which tells how to find and enjoy the beauty of a variety of Fermi surfaces.

The *Network* has a home page on World Wide Web (WWW). Its *Uniform Resource Locator* (URL) is: **http://www.dl.ac.uk/TCSC/HCM/PSIK/main.html**

The above contains pointers to several other nodes: *O.K. Andersen (Stuttgart)*, *P. Blöchl (IBM, Zürich)*, *M. Gillan (Keele)*, *E.K.U. Gross (Univ. Würzburg)*, *B.L. Györffy (Bristol)*, *V. Heine (Univ. Cambridge)*, *R. Jones (Univ. Exeter)*, *J. Kübler (TH, Darmstadt)*, *J.L. Martins (INESC, LISBON)*, *R. Nieminen (TU, Helsinki)*, *J. Nørskov (TU, Lyngby)* with information on CAMP, *M. Scheffler (FHI Berlin)*, *K.-H. Schwarz (TU, Vienna)*, *G.P. Srivastava (Univ. Exeter)*, and *A. Walker (UEA Norwich)*. Additionally, recently we have also included a pointer to the WWW home page of the Solid State Theory Group at National Renewable Energy Laboratory, Golden, CO 80401 *(http://www.sst.nrel.gov)*. If you maintain a home page on your activities we will be happy to include a pointer from the *Network's* home page to your home page.

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

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

	function
psik-coord@daresbury.ac.uk	**messages to the coordinator & newsletter**
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2 News from the Network

As the end of the calendar year is fast approaching, so is the time of a final report to Brussels on the Network's activity. This time, however, it is to be the report summarizing the four year activity of the Network whose lifetime comes to an end in December. Therefore we would need the cooperation of all the official nodes-associated members of the Network. We would appreciate all information on your Network's activity during the past 4 years, scientific publications that you have published this year where the Network was explicitly acknowledged. We would need about a page write-up from each of you on a scientific highlight of your research that has evolved due to the collaborations within the Network. We would like to hear how each of you benefited from the Network, be it in the form of attending workshops, conferences, getting computer codes, or making collaborative visits to start or continue scientific projects. Please do send your contributions by November 1. The last Newsletter of this calendar year will be the December issue and we would like to make it a special issue, summarizing our 4 year activity.

The Ψ_k-HCM Network is to be succeeded by a TMR Network which is to start formally at the beginning of next year. The Newsletter will of course continue pretty much as now, however may change its first page and content design to incorporate all the different networks that have originated from the Ψ_k-HCM Network. One of them is the TMR Network on 'Interface Magnetism', which was created by the Magnetism Working Group of the present Network. It has been in existance for the last year or so, and its workshops and other scientific material have been present in the past Newsletters, including the present one. The other, already mentioned, TMR Network, which is a direct continuation of the present Ψ_k-HCM Network, will partly continue the networking activity of the Ψ_k-Network, although in most part it will be much more projects oriented. A possible third 'mega-network' which the Newsletter will cover is the ESF Programme that was mentioned in our previous Newsletters. Its official launching is expected some time in spring next year.

3 News from the Working Groups

3.1 Reports on Collaborative Visits

Report on the collaborative visit of J.B.A.N. van Hoof (Eindhoven and Delft) to Cardiff 9–13 June, 1997

Dr J.B.A.N. van Hoof (Eindhoven and Delft) visited Cardiff from 9 – 13 June, to work with John Inglesfield on developing the embedding method. Over the last few years, Jeroen van Hoof has developed the embedding method so that the self-consistent electronic structure of bulk, surfaces and interfaces can be calculated accurately: the system is divided into layers, and the Green function for each distinct layer is calculated. From the Green function, the transmission and reflection properties of each layer can be found, and these can be put together to find the reflection properties and embedding potential for a semi-infinite substrate. The embedding potential on each side of a particular layer provide the right boundary conditions to embed it, either into the bulk, the surface or an interface. Jeroen has coded this up, and during the visit to Cardiff this was mounted on the local work stations and some test examples were run. At present we are using the codes in Cardiff to study screening of electric fields at surfaces.

(J.E. Inglesfield)

Report on the collaborative visit of Charles Patterson (Dublin) to York 17–18 July, 1997

The Trinity College group is developing a many-body code which will use DFT wavefunctions obtained from the crystal95 code as input. This code uses a Gaussian orbital basis and this will make it possible to treat systems containing 1st row elements such as C and materials containing transition metals. Silicon and other semiconductors are easier materials to begin with and we discussed using the space-time method used by Godby and coworkers for calculations on silicon. The use of a Gaussian orbital basis for calculating the self energies of silicon is now being tested in York using wavefunctions generated in Dublin on a real space grid using the crystal95 DFT wavefunctions as input. Comparison with self energies obtained from well converged plane wave basis set wave functions will give some indication of how well the Gaussian basis will perform in self energy calculations.

(Charles Patterson)

Report on a Collaborative Visit of C.Molteni (Max Planck Institut, Stuttgart), to the Theory of Condensed Matter group, University of Cambridge

10–23 July 1997

I visited Prof. V. Heine and Dr. M.C. Payne at the Theory of Condensed Matter group of the Cavendish Laboratory (University of Cambridge) from 10 to 23 August 1997.

The goal of my visit was to investigate the possibility to study the interaction between a dislocation and a grain boundary, a process that is believed to be of great importance for the mechanisms of mechanical failure in metals, by means of a recently developed scheme for metals based on ensemble density functional theory (N. Marzari, D. Vanderbilt and M.C. Payne, Phys. Rev. Lett.**79**, 1337 (1997)).

This project wants to expand and complete our previous work on mechanical properties of metallic grain boundaries, that has been recently published (C. Molteni, N. Marzari, M.C. Payne and V. Heine, Phys. Rev. Lett. **79**, 869 (1997)).

My visit benefited also from the presence of N. Marzari from Rutgers University, who was in Cambridge at the same time, and from previous discussions with Prof. M.W. Finnis, from the Queen's University of Belfast.

It was very useful to be in Cambridge and to have the possibility to discuss and clarify a number of issues related to the development of new grain boundary calculations, and I am grateful to the network for its support.

(C.Molteni)

Report on a Collaborative Visit of K. Capelle (Würzburg) to B.L. Gyorffy (Bristol)

September 8-14, 1997

The purpose of this visit was the continuation of the Bristol-Würzburg collaboration on superconductivity.

During the visit we had discussions about various topics which are part of the Bristol-Würzburg collaboration on superconductivity, such as dichroism in superconductors, density-functional theory for superconductors and relativistic effects in superconductors.

We also discussed a new common project, namely the scattering of X-rays from the vortex lattice, and performed some first calculations. In this context we drafted a proposal to the European Union for a Marie Curie scholarship, which would found a longterm stay of mine in Bristol, to work on this latter project.

(K. Capelle)

Report on the Collaborative Visit of M. Lüders (Würzburg) to Daresbury

July 14 - Aug 9, 1997

The main purpose of this visit was to finalise work on the superconductivity project started during my previous visit. Specifically, the aim was to finish the development of the computer code for calculating matrix-elements of the mutual Coulomb interaction, which appear in both the Kohn-Sham-gap equation and the exchange and correlation contributions to the pairing potential. Calculations for the free electron gas, for which these matrix-elements can be verified analytically, were completed. The results of the calculation, using the tight-binding LMTO in the atomic sphere approximation (ASA), revealed a severe disagreement with the analytical values. Careful examination of the different constituents of the matrix-elements showed, that the poor results could be traced back to poor wavefunction coefficients. These coefficients, and thus also the Coulomb matrix-elements, could be drastically improved by including the so-called combined correction term (CCT), which corrects the ASA description of the interstitial region.

Due to the rather big enhancement of the wavefunction coefficients, we also incorporated the CCT into the self-interaction corrected LMTO-ASA-computer code of the Daresbury group. Here the better wavefunctions led to a better angular momentum convergence, and the improved treatment of the interstitial region reduced the necessity of introducing empty spheres. Hence the secular matrix becomes smaller and the convergence gets better. Both smaller matrices and better convergence, of course, substantially reduce the CPU time.

Additionally, we had many interesting and valuable discussions on both the LMTO method and the common superconductivity project.

(Martin Lüders)

Report on the Collaborative Visit of S. Beiden (Sheffield) to Aarhus

August 4 - 29, 1997

The Self-Interaction Correction to the Local Spin Density (LSD) approximation provides a successful description of the localized-delocalized transiton in $4f$ rare earth metals and their compounds, such as Ce, cerium monopnictides [1]. However to study a similar behaviour of $5f$ systems, such as Am, U and its compounds etc, one might need to apply the fully relativistic approach to the self-interaction correction method. Americium is the first element in the actinide series which behaves like a rare earth: experimental results indicate that the 5f electrons are localized. It has been expected that americium under compression will undergo a transition to a dense phase where $5f$ electrons are itinerant. During the visit the relativistic self-interaction correction approach, which had been used before to study orbital and magnetic moments in γ-Ce [2], was applied to obtain preliminary results on localized-delocalized transition in Am, similar to the one in elemental Ce. Preliminary calculations for elemental U were also performed to study the influence of different SIC-corrected orbitals on the ground state energy and such quantities as magnetic and orbital moments, with the intention of gaining experience so that more complex compounds like e.g. UPd_3 can be studied. The present relativistic SIC-LSD code had been extended to the case of several atoms per unit cell and was then tested by applying it to UPd_3. Various experiments on UPd_3 have ascertained that there are two f electrons per U which are localized. To simulate the localized f electrons, two were placed in the core in the previous calculations of M. Norman et al.[3]. Self-interaction correction approach has been applied to simulate this situation and we are doing futher calculations now. Some work was also done to make a parallel version of the code and adjust it to the MPL platform on SP/2.

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(Sergey Beiden)

Report on the one month secondment of J. Goss to V. Torres

July 1st – July 31th 1997: The extraordinary optical properties of S donors in GaP

Theory of GaP:S_P

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Abstract

Recent infrared absorption spectroscopy experiments (Grosche *et al.* 1997) have revealed the presence of a vibrational mode at 272.5 cm^{-1} (in the gap between the acoustic and optic branches of the phonon dispersion) associated with the ionised S donor in GaP. No absorption is detected for neutral defects. We have carried out local-density-functional calculations on large atomic-clusters terminated by H. We find that the ionised defect absorbs strongly around 270 cm^{-1} but the oscillator strength for the transition is greatly reduced for the neutral defect. The calculations rule out structural changes as being responsible for the effect. The reduction in the strength of the transition completely explains the experimental findings and the switching between absorbing and transparent modes with the speed of an electronic excition may be of technological interest. The calculations also show DX behaviour of GaP:S is unlikely.

Introduction

V. Torres (Universidade de Aveiro) and J. Goss (University of Exeter) have had a collaborative relationship through the theoretical physics group in Exeter for several years. J. Goss' primary area of expertise is in the electronic, optical and structural properties of point defects in diamond, Si and GaAs. V. Torres has experience in first principles calculations of centres in GaN and GaP and importantly sulphur centres in III-Vs and Si.

The support of the PsiK network has enabled J. Goss to stay in Aveiro to benefit from this experience as well as the local computing facilities. The visit extended throughout the month of July. This work was also performed with reference to and the cooperation of the experimental group that made the recent observations (R. C. Newman *et al.*, Semiconductor IRC, Imperial College, London) and with M. Sangster of the Theoretical Physics group at Reading.

The collaboration has proved very useful and a publication will follow shortly.

Scientific Content

We have found an extraordinary optical property of sulphur donors in GaP. In their positive charge state they absorb strongly at 272.5 cm^{-1}, but the material is transparent if the donor level is occupied. The switching between absorbing and transparent modes with the speed of an electronic excition may be of technological interest and should be pursued. The effect is not that the absorption causes a structural change, possibly involving a barrier, which leads to a different vibrational frequency, but rather the oscillator strength of the transition is profoundly changed.

Sulphur acts as a donor in Ga-(group-V) compound semiconductors, lying at the group-V lattice site. These centres have relatively deep donor levels: for example, GaP:S_P is reported to have a donor level at 107 meV below the conduction band [1]. Furthermore, S in GaSb, GaAs and $GaAs_{1-y}P_y$ can possess a DX character, where either heavy doping levels or applied pressure can induce a change of structure for the S centre as it captures an electron to form S^-.

EPR has been measured on S doped GaP [2], as in the neutral charge state S_P possesses an unpaired electron. The resonance signal was reported to be a single line, Lorentzian in shape and structureless. GaP:S_P has also been examined using electron nuclear double resonance (ENDOR) [3] which suggests tetrahedral symmetry.

In zinc-blende materials made up from group III and group V elements of very different masses, a substantial gap exists between the acoustic and optical branches of the one-phonon dispersion. For GaP, this gap lies between 255 and 326 cm^{-1}. Normally, impurities with a mass close to that of the host material can only be observed as resonances in the one-phonon absorption, which tends to be very broad. However, where a substantial gap is present impurity related vibrational frequencies can lie in the one-phonon frequency range but still be detected as sharp absorption peaks, and fine structure resolved. This is true for GaP:As_P which possesses a local vibrational mode at 269 cm^{-1} [4]. This band is very sharp (a line width of only 0.15 cm^{-1}) and the fine structure due to the Ga isotopes neighbouring the As_P centre have been resolved [4].

Recent infrared absorption experiments [5] have revealed a gap mode in n-type and compensated GaP doped with S. In the as-grown (n-type) material, the mode appears at 272.5 cm^{-1} along with a broad band in the acoustic branch from 210-255 cm^{-1}, centred at 245cm^{-1}. The gap mode in the as-grown sample is described as weak, an overall width of 1.6 cm^{-1}, and possesses no fine structure. However, when the S donors are compensated by irradiation with 2 MeV electron, the gap mode becomes much more intense, and a second mode at 266.2 cm^{-1} is also observed. The integrated absorption from these two peaks correlate with the natural abundances of ^{32}S and ^{34}S (95:4.2), confirming the source of the vibrational modes. The third isotope of sulphur (^{33}S) is much less abundant (0.76%), and would be likely to coincide with the As_P absorption.

Significantly, there is only one mode observed for each S isotope. Furthermore, the 272.5 cm^{-1} mode only changes intensity when the neutral S donors are ionised. In Ref. [5] it is suggested that there are two possible explanations: either the mode due to neutral S lies outside of the gap (i.e. the S-Ga force constant has sufficiently different from that of S^+ that the mode is resonant

with bulk-phonon modes), or the frequencies are *the same* (to within experimental resolution) but the effective charge of the S^0 vibration is much smaller than that of S^+. It is argued that the gap absorption seen at 272.5 cm^{-1} in the uncompensated sample arises from a small amount of S^+ and the remainder from S^0. It is assumed that the compensated sulphur is present wholly due to compensation of C_P. It is not clear that this is the case.

They support the effective charge hypothesis by saying that a shallow donor will be effective-mass-like and therefore the extra electron will be somewhat dispersed in an orbit about the S atom (the first Bohr radius is $\sim$5Å [5]). The presence of this donor electron should therefore have only a minimal effect on the local geometry of S_P, and a minimal effect on the local force constants. However, the S impurity is a reasonably deep for a shallow donor at 107 meV [1]. In fact the EPR data cannot be interpreted in a purely effective mass like model [2]. Therefore local perturbations in the geometry or force constants cannot be ruled out.

Sulphur is known to behave as a DX centre in III-V materials. For example, in GaSb sulphur gives a deep donor DX centre [6, 7, 8]. DX centres in GaAs are known to be stabalised by the application of pressure [8]. Furthermore local internal strains due to alloying gives rise to sulphur DX centres in $GaAs_{0.6}P_{0.4}$ at ambient pressures [9]. There are three main microscopic models for these centres. The first is a simple tetrahedral 'breathing mode' expansion of the donor neighbours, and the second is where an extra electron is accommodated in a dilated bond termed the broken-bond-model (C_{3v}). The final model is where two next-neighbour cations move to become bonded in a C_{2v} symmetry system [8], termed the cation-cation-bond model. The three configurations are determined theoretically to be typically a few meV apart [8].

We report the results of local-spin-density-functional calculations performed on large, hydrogen terminated atomic clusters using AIMPRO (*ab initio* modeling program). The method has been described in detail elsewhere [10]. Electron-ion interactions are treated using the norm-conserving pseudopotentials from Ref.[11], except in the case of hydrogen, for which the bare Coulomb potential is used. The basis for the central 17 Ga, P, and S atoms at the core of the defect consists of 4, 4, and 5 sets of independent atom-sited s- and p_x-, p_y- and p_z-Gaussian orbitals with different exponents for the expansion of the electronic wavefunction, and a linear combination of 4, 4, and 8 s-Gaussian functions for the fit to the charge density. The basis used at each of the remaining atomic sites consisted of fixed linear combinations of 4(2) different s- and p-Gaussian orbitals for the wavefunction expansions for the bulk atoms (surface H). The charge density fit for the H atoms consisted of 3 s-Gaussians. The clusters used in this present study contain 309 atoms (including terminating H atoms): $Ga_{92}P_{93}H_{124}$. More details of these types of calculations can be found in Ref. [10], and references therein.

To determine the vibrational modes of the systems, each cluster was relaxed and the second derivatives of the energy with respect to the coordinates of the inner 17 atoms found. These were then used to construct the dynamical matrix of the cluster. The remaining entries were described by a Musgrave-Pople potential [12] which was deduced by a fit to the second derivatives of a relaxed H-terminated cluster containing only Ga and P atoms. This potential for atom i is given by:

$$V_i = \quad 1/4 \sum_j k_r^{(i)} (\Delta r_{ij})^2 + r_0^2/2 \sum_{j>k} k_\theta^{(i)} (\Delta\theta_{jik})^2 + r_0 \sum_{k>j} k_{r\theta}^{(i)} (\Delta r_{ij} + \Delta r_{ik}) \Delta\theta_{jik}$$

$$+\sum_{k>j} k_{rr}^{(i)} \Delta r_{ij} \Delta r_{ik} + r_0^2 \sum_{l>k>j} k_{\theta\theta}^{(i)} \Delta\theta_{jik} \Delta\theta_{kil}. \quad (1)$$

Here Δr_{ij} and $\Delta\theta_{jik}$ are the changes in the length of the $i-j$ bond and angle between the $i-j$ and $i-k$ bond, respectively, and r_0=2.402 Å.. The sum is over the nearest neighbour atoms only. Table 1 gives the coefficients $k_r^{(i)}$, $k_\theta^{(i)}$, $k_{r\theta}^{(i)}$, $k_{rr}^{(i)}$ and $k_{\theta\theta}^{(i)}$. This potential was used to generate the dynamical matrix of the whole cluster.

Table 1: Musgrave-Pople potential parameters for GaP (eV/Å^2), as described in Eq. 1.

Atom	k_r	k_θ	$k_{r\theta}$	k_{rr}	$k_{\theta\theta}$
Ga	10.097	0.736	0.537	1.834	0.918
P	10.097	0.564	0.623	0.173	0.213

Table 2 shows the frequencies at high symmetry points which are compared with the results of Ref [1]. The top of the optic band is due to modes at Γ and L. The highest experimental frequency is 409 cm^{-1} which is about 30 cm^{-1} below the calculated optic branch at L. Of great concern here, is the gap between the acoustic and optic branches. The gap is calculated to lie 251-292 cm^{-1} and the observed gap lies 255-326 cm^{-1}. Thus our gap is about 30 cm^{-1} too small.

Table 2: Phonon Modes of GaP, cm^{-1}.

Branch	Calculated	Experimental [1]
TO, LO(G)	336	355, 409
TA(L)	54	85
LA(L)	255	212
TO(L)	292	357
LO(L)	441	373
TA(X)	200	107
LA(X)	251	249
TO(X)	345	353
LO(X)	296	366

The clusters are centred at the S_P site. All atoms were relaxed in this cluster in the positive and neutral charge states. The local environment of the S atom was then perturbed to trigonal and tetragonal in turn and relaxed again. In the positive charge state, the perturbed structures do not have a lower energy than the tetrahedral structure. The S-Ga bonds are rather dilated to 2.48 Å (5% longer than the experimental value for the Ga-P bond at 2.36 Å). The calculated vibrational modes are listed in Table 3.

In the neutral charge state, as in the 1+, the 131 atom cluster yields similar energies for the T_d and C_{2v} symmetry structures. However, the experimental evidence clearly indicates an on site centre. The larger cluster (309 atoms) has a diameter of ∼11 Å which is of the order of the

Table 3: Gap and Local Modes of S defects in GaP, cm^{-1}, and their downward isotopic shifts.

Defect	^{32}S	^{34}S	^{32}S-^{71}Ga
Calc. S^+	256.3	4.5	0.1 (2), 0.0
Calc. S^0	260.2	5.0	0.1 (2), 0.0
Experimental [5]			
S	272.5	6.3	-

orbit of the donor electron quoted [5] at ~5 Å. This gives the T_d system to be lowest in energy. The energy double derivatives for the on site neutral centre were calculated and the dynamical matrix solved. The resultant gap mode is listed in Table 3). Clearly there is only a modest frequency shift of the donor in its two charge states.

The effective charge of the gap mode has also been calculated. This is found by calculating the induced dipole moment when the atoms in the cluster are displaced according to the normal coordinates of the appropriate normal mode. The magnitude of the induced dipole leads to an effective charge of the impurity. The ionised defect has then an effective charge of $0.57e$, assuming a mass appropriate to ^{32}S. This is nearly 50 times that of the neutral system ($0.013e$). This implies that there would be no detected absorption for the neutral defect. The origin of the effect is that it is the displacement of the ion core of the sulphur impurity, together with the surrounding displacements of the Ga shell, that dominate the induced dipole moment. The displacement of the additional donor electron, bound to these atoms in a Bohr radius of about 7 Å, then reduces the induced dipole moment of the sulphur core [5].

To address possible DX behaviour, the defect has also been relaxed in C_{2v}, C_{3v}, and T_d symmetries in a negative charge state. In each case the system was started in a highly perturbed state to model the cation-cation-bond, broken-bond and breathing mode relaxations. The total energies do not show a distortion from T_d is stable. Thus DX behaviour may only be stabalised by a local strain in the samples, either from applied pressure, high concentrations of impurities, or the alloying such as in $GaAs_{1-y}P_y$.

In conclusion, the calculations demonstrate that the neutral GaP:S_P centre does have a mode close to that of the ionised centre, as suggested by Grosche *et al.* and that the oscillator strength of the S-related normal modes are completely different. Finally, the calculations give no support to GaP:S_P^- being a DX centre in the absence of strain.

Acknowledgements

The work presented here was performed in part (JG) with the support of the PsiK network. S.Ö thanks NFR and TFR in Sweden for financial support, and also PDC at KTH for computer time on the SP2. We thank the HPCI for an allocation of computer time on the Cray T3D at Edinburgh and the SP2 at Daresbury. We gratefully thank E. G. Grosche, R. C. Newman and M. Sangster for discussions and access to experimental data prior to publication.

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3.2 Reports on Workshops

Report on the

Workshop on 'GMR'

TMR-Network on 'Interface Magnetism'

Dresden, September 13th - 14th, 1997

Organized by: **Ingrid Mertig** (mertig@ptprs2.phy.tu-dresden.de)

The workshop was held at the Institute of Theoretical Physics, University of Technology in Dresden, on the weekend of 13th-14th September. It was organized by I. Mertig and was sponsored by the TMR-Network on 'Interface Magnetism'.
The workshop was attended by 23 participants. As the enclosed program and abstracts indicate, we covered the area of Giant Magnetoresistance (GMR) and Tunnel Magnetoresistance (Inoue) in magnetic multilayers, in particular, the field of ab initio calculations of these properties.

Starting from the electronic structure of a layered system the GMR effect was considered in the diffusive and ballistic regime. Spin-dependent scattering at domain walls (Brataas), at impurities (Zahn) and at substitutional interface alloys (Weinberger, Szunyogh, Blaas) was discussed as origin of GMR. Although spin-dependent scattering is extremely important for GMR the spin-dependent electronic structure of the layered systems seems to be a significant ingredient to establish GMR already which shows in reflection and transmission coefficients of layered systems and which was demonstrated for GMR in Uranium compounds (Richter, Oppeneer). We heard a useful summary of the current state on how to calculate the electrical conductivity of disordered semi-infinite systems using the Kubo-Greenwood approach within the Screened-KKR-CPA (Weinberger, Szunyogh) and within the TB-LMTO scheme (Kudrnovský). The influence of vertex-corrections was debated in detail (Bauer).

In summary, it was a scientifically interesting meeting with many lively discussions.

(Ingrid Mertig)

PROGRAM:

Saturday, September 13th, 1997
Morning session:
Chair-person: Paul J. Kelly

9.00		**Opening**		
9.05	9.45	Effect of roughness on the CPP-GMR and tunnel MR	J. Inoue	Nagoya
9.45	10.25	Resistivity of a domain wall	A.Brataas	Delft
10.25	11.00	**Coffee break**		
11.00	11.40	Quantum confinement in magnetic multilayers and GMR	P.Zahn	Dresden
11.40	12.20	Reflection and transmission for $Fe-$, $Co-$, $Ni-$(001)-layers in Cu	P.H. Dederichs	Jülich
12.20	14.00	**Lunch**		

Afternoon session:
Chair-person: Peter H. Dederichs

14.00	14.40	Periods evolution and damping in the oscillatory exchange coupling through Cu_xNi_{1-x} alloy spacer	N.N.Lathiotakis	Bristol
14.40	15.20	Electrical conductivity of disordered semi-infinite systems	P.Weinberger	Vienna
15.20	15.40	**Coffee break**		
15.40	16.20	Implementation of the Kubo-Greenwood formula to layered systems in terms of the spin-polarized relativistic screened-KKR-CPA method	L.Szunyogh	Vienna

16.20	17.00	Numerical evaluation of the off-diagonal contributions to the electrical conductivity in layered systems	C.Blaas	Vienna
17.00	17.40	Transport in magnetic multilayers: TB-LMTO approach	J.Kudrnovský	Vienna
20.00		**Joint dinner**		

Sunday, September 14th, 1997
Morning session:
Chair-person: P. Weinberger

9.00	9.40	Theoretical investigations on the magneto-optical properties of transition metal multi- and surface layer systems	H.Ebert	Munich
9.40	10.20	Density functional calculation of GMR in uranium intermetallics	M.Richter	Dresden
10.20	10.50	**Coffee break**		
10.50	11.30	Analysis of colossal magnetoresistance mechanisms in intermetallic compounds from ab initio calculations	P.M.Oppeneer	Dresden
11.30	12.10	On the shift of the chemical potential in the double exchange model in infinite dimension	M.S. Laad	Dresden
12.10	12.15	**Closing**		
12.15	14.00	**Lunch**		

Please note:
All talks are 40 minutes including 10-15 minutes discussion!

Effect of roughness on the CPP-GMR and tunnel MR

J. Inoue
Nagoya University, Japan

The interfaces of the multilayered systems play an important role on the magnetoresistance (MR) in these systems. There often exists roughness at the interfaces. In order to study the effect of roughness on MR, we have formulated the conductance for currents perpendicular to planes using the Kubo formula and coherent potential approximation. The geometry studied is the same with that of the ferromagnetic tunnel junctions. We will report the numerical results for CPP-GMR and tunnel MR and show that the roughness brings about a large effect on the MR.

This work has been done in collaboration with H. Itoh, A. Shibata, T. Kumazaki and Prof. S. Maekawa.

Resistivity of a domain wall

A. Brataas* and G. Bauer*
* *Delft University of Technology, Faculty of Applied Physics,*
Lorentzweg 1, 2628 CJ Delft, The Netherlands

We consider transport through a diffuse ferromagnet with a domain wall. The conductivity is calculated to the lowest order in the impurity scattering and the scattering due to the domain wall following Tatara and Fukuyama. In contrast to these authors we consider also the possibility to include a realistic band-structure of the ferromagnet and different life-times for the quasi-particles.

Approximations to make the calculation scheme feasible for ab-initio band- structure calculations are discussed.

Quantum confinement in magnetic multilayers and GMR

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it * Technische Universität Dresden, Institut für Theoretische Physik,
D-01062 Dresden, Germany

Ab initio calculations are presented for Giant Magnetoresistance (GMR) in magnetic multilayers. The electronic structure of the magnetic multilayers is calculated within the spin-density functional theory. Impurity scattering is included be means of a Green function technique. Transport is described quasiclassically. GMR is calculated in both configurations - current in plane (CIP) and current perpendicular to the plane (CPP). The character of the wave function

is analyzed with respect to confinement in quantum wells. It will be demonstrated that quantum well states contribute mainly to CIP transport, whereas extended states are responsible for CPP transport.

Reflection and Transmission for Fe-, Co- and Ni(001) Layers in Cu

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The reflection and transmission at the interface between a nonmagnetic and a magnetic layer represents a key quantity for the understanding of interlayer coupling, quantum well states as well as ballistic transport. For this reason we calculate the reflection coefficient of a Cu Bloch electron at the (001) interface of fcc Fe, Co and Ni layers of finite thickness. The main emphasis lies on the understanding of the size-oscillations arising from the interferences of the waves scattered at the front and back interface of the magnetic layer. In the first part we give a general analysis of the scattering in one dimension in terms of scattering phase shifts and present analytical formulas for the reflection coefficients and the integrated density of states. We further discuss the consequences of time inversion symmetry. In the second part we present KKR-Green's function calculations for Fe, Co and Ni layers of various thicknesses and discuss resonance effects and other anomalies in the reflectivity as well as the transition to the halfspace geometry. Finally we demonstrate that the reflectivity can be strongly influenced by a magnetic adlayer, e.g. a Fe monolayer on top or back of a Co or Ni layer.

Periods' Evolution and Damping in the Oscillatory Exchange Coupling through Cu_xNi_{1-x} Alloy Spacer

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[c] *Technical University, Vienna, Getreidemarkt 9/158, 1060 Vienna, Austria*

The topology of the Fermi surface surface obtained by KKR-CPA calculations was used to find the evolution of the oscillation periods with concentration in the case of the Oscillatory Exchange Coupling through an alloy spacer (Cu_x Ni_{1-x}). The quasiparticles' coherent lengths were also calculated over the Fermi surface and the consistency of the absence of exponential dumping in experiments with the residual resistivity was investigated.

The oscillation periods evaluated by the Fermi surface extremal vectors were found in a very good agreement with experimental data on (111) [1,2] and (110) [3] growth directions. On the other hand the calculated coherent lengths were found large enough to reproduce the residual

resistivity measurement. A scale down by a factor of about 2 in the coherent lengths is neccesary to bring convinience between calculated and experimental residual resistivity. Even scaled the coherent lengths are large enough to prevent dumping of the oscillations in consistency with experimental observations [1-3].

References

[1] Bobo F-J., Hennet L., Piecuch M. and Hubsch J., J.Phys.: Condens. Matter **6** 2689 (1994).

[2] Parkin, S.S.P., Chappert C., Herman, F., Europhys. Lett. **24**, p 71 (1993).

[3] S. N. Okuno, K. Inomata, Phys. Rev. Lett. **70**, 1711 (1993).

Electrical conductivity of disordered semi-infinite systems

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[c] *Fraunhofer-Institut für Angewandte Materialforschung, Lesumer Heerstrasse 36, D-28717 Bremen, Germany*

The Kubo-Greenwood formula for the electrical conductivity of disordered semi-infinite systems is formulated in terms of the (Screened) Korringa-Kohn-Rostoker method and the Coherent Potential Approximation. It is shown that by neglecting vertex corrections in the same manner as in the bulk case, the elements of the electrical conductivity tensor can be described in terms of "layer-diagonal"- and "layer-off-diagonal" contributions. In order to discuss effects of the underlying electronic structure on the electrical conductivity, but also to point out particular contributions to it, Bloch spectral functions are formulated. A spin-polarized (relativistic) application of the present theory to non-collinear disordered magnetic multilayers allows to discuss giant magnetoresistance (GMR) on an ab-inito level in quite a general context.
This contribution is supposed to recall main ideas already discussed in the literature.

Implementation of the Kubo–Greenwood formula to layered systems in terms of the spin–polarized relativistic Screened–KKR–CPA method

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[b] *Department of Theoretical Physics, TU Budapest, Hungary*

[c] *Fraunhofer–Institut für Angewandte Materialforschung, Bremen, Germany*

By employing the Screened Korringa–Kohn–Rostoker method we present a full computational scheme to solve the conditions of an inhomogeneous Coherent Potential Approximation (CPA), to evaluate the Bloch spectral functions and the Kubo–Greenwood formula for the *dc* conductivity for disordered layered systems. For the latter one, we discuss in some details the computation of the matrices of the current operator within a relativistic and a non–relativistic formalism and we establish relations between quantities involved in the formula with energy arguments $\epsilon \pm i\delta$ which considerably reduce the computational efforts. Special care is taken to the evaluation of the Surface Brillouin Zone (SBZ) integral in the *layer–off–diagonal* terms, where group theoretical methods are used to keep the computation to be tractable.

Numerical evaluation of the off-diagonal contributions to the electrical conductivity in layered systems

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[b] *Department of Theoretical Physics, TU Budapest, Hungary*
[c] *Fraunhofer–Institut für Angewandte Materialforschung, Bremen, Germany*

Employing the Kubo–Greenwood formula, the electrical conductivity of disordered layered systems is computed in terms of the Screened Korringa–Kohn–Rostoker method and the Coherent Potential Approximation. Numerical implications in evaluating the Surface-Brillouin-Zone integral that enters the expression for the *layer–off–diagonal* terms to the electrical conductivity are discussed, partially in connection to the corresponding Bloch spectral functions. Preliminary numerical experiences are presented for an interdiffused $Cu/Co_1/Cu_2/Co_1/Cu(100)$ and the $(Fe_{0.5}Co_{0.5})_6/Cu(100)$ systems, representing weakly and strongly disordered cases, respectively.

Transport in magnetic multilayers: TB-LMTO approach

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[c] *Institute of Physics of Materials, ASCR,*
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The Kubo-Greenwood approach to the electronic transport in metallic multilayers is developed within the TB-LMTO method which allows for two different formulations. The first approach, formulated along the lines of the KKR method[1,2], expresses the velocity operator in terms of wave functions on a given atomic sphere. In the other approach, very much in the spirit of the TB method, we use the equation of motion to obtain the expression for the velocity operator. For the perpendicular transport in multilayers the latter approach leads to the formulation developed by Lee and Fisher [3], Asano [4], and Mathon [5].

The simple s-cubium model of the trilayer is used to discuss the pecularities of numerical calculations. We present results illustrating the oscillatory behavior of the GMR ratio with the spacer and magnetic slabs thicknesses. The effect of alloying in a system on the GMR ratio is also studied for a simple s-cubium model and the results are discussed.

1. W.H. Butler et al., Phys. Rev. B **52**, 13399 (1995)
2. P. Weinberger et al., J. Phys.: Condens. Matter **8**, 7677 (1996)
3. P.A. Lee and D.S. Fisher, Phys. Rev. Lett. **47**, 882 (1981)
4. Y. Asano et al., Phys. Rev. B **48**, 6192 (1993)
5. J. Mathon et al., Phys. Rev. B **52**, R6983 (1995)

Theoretical investigations on the magneto-optical properties of transition metal multi- and surface layer systems

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The basis of theoretical investigations on the magneto-optical properties of transition metal surface and multilayer systems in the visible and X-ray regime of light is outlined. An appropriate description of the underlying electronic structure has to account for both the spin-orbit coupling and the magnetic state of the investigated system at the same time. As will be shown, this can be achieved in a very satisfying way within the framework of the fully relativistic Dirac formalism. Results of corresponding calculations on the magneto-optical Kerr effect (MOKE) in the visible regime of light, as well as of the magnetic dichroism in X-ray (MXD) absorption for Fe- and Co-based surface and multilayer systems are presented and compared with experimental data. Concerning the MOKE it is most interesting to study the changes induced by a second component compared to the properties of pure Fe or Co, respectively. As will be shown, the MXD on the other hand allows in particular to study the induced magnetic moments induced by Fe or Co on adjacent layers of atoms that are otherwise nonmagnetic.

Density functional calculations of the giant magnetoresistance in uranium intermetallics

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* *Technische Universitt Dresden, MP Arbeitsgruppe,*
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*** *Department of Metal Physics, Charles University,*
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Based on the coherent electronic structure of the ideal crystal, the giant magnetoresistance (GMR) of U_2Pd_2In at field-induced transitions from the antiferromagnetic ground state to spin-flipped non-collinear states is calculated using linear transport theory. An inverse effect of up to 20% is predicted, that we relate to symmetry lowering in the field. Calculations on UNiGe and U_2Pd_2X (X = In, Sn) for the case of model magnetic structures demonstrate that spin-dependent scattering should be of minor importance for actinide compounds due to strong spin-orbit coupling.

Analysis of colossal magnetoresistance mechanisms in intermetallic compounds from ab initio calculations

P.M. Oppeneer, A.Ya. Perlov, V.N. Antonov, A.N. Yaresko, and S.V. Halilov
Max-Planck Research Group "Electron Systems", University of Technology, D-01062 Dresden, Germany

We have investigated the various mechanisms that may cause a colossal or giant magnetoresistance (MR) in crystalline uranium and transition-metal intermetallics. Our approach is based on *ab initio* electronic structure calculations in the framework of the LSDA. Linear-response theory is used to evaluate the field dependent conductivity tensor. Several mechanisms that may bring a giant MR about are identified from our calculations. In the case of UNiGa, where an anisotropic, colossal MR of -87% for CPP and -58% for CIP was measured, we find that at the field induced metamagnetic transition an appreciable superzone reconstruction of the Fermi surface occurs, which contributes to the MR through a reduction of the total Fermi surface area, and through a substantial, anisotropic, reshaping of the Fermi surface. The latter leads accordingly to a very anisotropic MR. In the case of UNiSn, which also exhibits a colossal MR, the basic mechanism is the proximity of the metamagnetic transition to an anomalous metal-insulator transition. For FeRh we calculated an isotropic giant MR of -75%, which we find to be due to superzone Fermi surface gapping. Upon doping the MR is predicted to become colossal, i.e. -86%, in good agreement with experiment. Results for several other compounds are also discussed. Our calculations illustrate particularly that the MR phenomenon in intermetallics can be well understood from their underlying electronic structure.

On the shift of the chemical potential in the double exchange model in infinite dimensions

Mukul S. Laad and Jean-Marc Robin
MPI für Physik Komplexer Systeme, Nthnitzer Strasse 38, Dresden

The double-exchange (DE) model with itinerant carriers coupled to core spins and static lattice distortions or disorder is solved exactly in infinite dimensions. The coupling to the lattice is described within the adiabatic approximation, which we argue to be reasonable for this model. We find that increasing magnetic field triggers a jump in the carrier density as (geh/2mc)B crosses the chemical potential. It is argued that disorder could lead to similar features. Our

results are in qualitative agreement with indications from Hall measurements on La-Sr-Mn-O, which indicate that the CMR in these materials is driven by the field induced increase in the carrier concentration, rather than by an increase in the effective hopping between Mn ions.

4 Workshop/Conference Announcements

4.1 Workshop on Quantum Theory of Solids

QTS-4 Workshop in Aarhus, November 12-15, 1997

Partially supported by Ψ_k-*Network*

TITLE: 'Quantum Theory of Solids 4, QTS-4'
DATES: 12-15 November 1997.
PLACE: Institute of Physics and Astronomy, Aarhus University
DK-8000 Aarhus C, Denmark.
ORGANIZER: Niels E. Christensen (address as above).
E-mail : nec@dfi.aau.dk; Secr.: bhr@dfi.aau.dk
Teleph.: (+45) 8942 3666; Secr.: (+45) 8942 3665
FAX : (+45) 86 12 07 40
Web : **http://www.dfi.aau.dk/~nec/qts4.html**

"QTS-4" is the fourth meeting in Aarhus on quantum theoretical studies of solids. Whereas previous QTS meetings were limited to 1 or 2 days, QTS-4 is planned to extend over 3 1/2 days. One reason is that we have invited a larger number of speakers, but still wish to have room in the programme for discussions. Another reason is that QTS-4 is a kind of status of the Argentinian- Danish collaboration which was initiated through the EU project 'First principles calculations of electronic and vibronic properties of complex materials'.

The subject of the workshop is: "Methods for accurate *ab initio* calculations of physical properties of complex materials". The purpose is to examine and compare state-of-the-art methods for calculating physical and chemical properties of materials from electronic structures; (FP-LMTO, FP-LAPW, plane-wave methods) including O(N) methods. Recent experiences with improved density-functional methods (gradient corrections, 'optimized potentials', GW and SIC) are discussed. Applications include structural, vibronic, as well as optical properties. In particular Raman intensity calculations for high-temperature superconductors and the relation of these to most recent experiments are discussed. Further specific subjects include 'ab initio' calculations of phonons and defects in perovskites, ferroelectricity, and high-pressure structures of compound semiconductors.

INVITED SPEAKERS:

M. Alouani, Strasbourg (France)
C. Ambrosch-Draxl, Graz (Austria)
O.K. Andersen, Stuttgart (Germany)
F. Aryasetiawan, Lund (Sweden) (not yet confirmed)

M. Cardona, Stuttgart (Germany)
G. Fabricius, Madrid (Spain)
B. Johansson, Uppsala (Sweden) (not yet confirmed)
C. Koenig, Rennes (France)
E.A. Kotomin, Riga (Latvia)
A.I. Lichtenstein, Jülich (Germany)
M. Methfessel, Frankfurt/O (Germany) ("maybe")
R.L. Migoni, Rosario (Argentina)
D. Novikov, Evanston (USA)
A. Postnikov, Osnabrück (Germany)
C.O. Rodriguez, La Plata (Argentina)
M. van Schilfgaarde, Stanford (USA)
W.W. Schulz, Cambridge (UK)
H. Skriver, Lyngby (Denmark)
J. Soler, Madrid (Spain)
M. Stachiotti, Rosario (Argentina)
POSTER SESSION (contributed): Please submit abstracts before Oct. 16.

5 Job Announcements

Post-doctoral position
Technische Universität Dresden, Germany

Applications are invited for a post-doctoral position of the TMR network "Interface Magnetism" in the area of electronic structure and transport properties of magnetic multilayers. Experience in the area of electronic structure calculations would be desirable. The project is based on TB-KKR calculations of layered systems including nanostructural defects and particularly aimed at calculating Giant MagnetoResistance. The project is based on a collabortion with the KKR-group of P.H. Dederichs at the Institute for Solid State Physics in Jülich and the candidate is expected to stay part of the time in Jülich. The position is open for all EU nationals (except Germans) and is available beginning January 1, 1998 or later for one year. Please contact:

Dr. Ingrid Mertig
TU Dresden
Institut für Theoretische Physik
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Post-doctoral Position

Condensed Matter Theory

Strasbourg, France

Applications are invited from European Union nationals, excluding French residents, for the above post funded by a European Training and Mobility Award. The TMR network is supported for a duration of 3 years. The appointment will be funded for a period of two years, extendable on mutual agreement. The successful applicant would be expected to work on the development of a RS-TB-LMTO (Real-Space Tight-Binding LMTO) method and application to interface and thin film magnetism, under the supervision of Pr. Dreysse. Some experience in electronic structure calculations is essential. The salary will be on the Research Assistant scale, according to French regulations. Interested candidates should contact Pr. Dreyssé for further information and send a C.V. with names of two references.

Professor H. Dreyssé
Institut de Physique et de Chimie des Matériaux de Strasbourg
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23, rue du Loess
67037 Strasbourg, France
tel: (33) 03 88 10 70 83
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Announcement

ONE OR TWO POSTOCTORAL POSITIONS AVAILABLE NOW

in the ELECTRONIC STRUCTURE group at the International School for Advanced Studies - Scuola Internazionale Superiore di Studi Avanzati (ISAS-SISSA), Trieste, Italy

http://www.sissa.it/cm/CMsector/electronic_str

One or two post-doctoral associate positions are available now in the following fields:

- first-principles studies of SEMICONDUCTOR ALLOYS AND INTERFACES,
- first-principles studies of SURFACES.

The applicants should have a background in Density Functional techniques. Experience on Parallel Computining is welcome. These positions are available from Fall 1997 for one year and renewable for a second year. Net salary is 26.000.000 lire. Please send a copy of your CV to **degironc@sissa.it** or fax it to **(+39) (40) 3787-528**. Please include the name, phone number, and email address of one reference.

6 WEB announcement

Do you want to enjoy the beauty of Fermi surfaces ?

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and go to

http://www.phy.tu-dresden.de/~fermisur/

Fermi surfaces, Fermi velocities and character of the electronic states at the Fermi surfaces of the highlighted metals in the periodic system are at the moment available. The systems are considered in the given crystal structure.

Ingrid Mertig

mertig@ptprs2.phy.tu-dresden.de

7 Abstracts

Theoretical tensile stress in tungsten single crystals by full-potential first-principles calculations

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Abstract

First ab initio electronic structure calculation of theoretical tensile strength is performed by means of full-potential LAPW method. As a specific example, tensile strength of single-crystalline tungsten loaded uniaxially along the (001) and (111) directions is analyzed. Although tungsten is elastically nearly isotropic ($C_{44} \approx C'$), theoretical tensile strength exhibits a marked anisotropy ($\sigma^{th}_{001} = 0.289$ Mbar, $\sigma^{th}_{111} = 0.401$ Mbar). This anisotropy is explained in terms of structural energy diferences between bcc, fcc and simple cubic structures which occur on the calculated deformation paths. Theoretical results compare favorably with experimental value of 0.247±0.036 Mbar obtained for tungsten whiskers grown along the (110) direction.

(Mat. Sci. Eng. A **234-236** (1997), 1075-1078;
Proc. 11th Conf. on the Strength of Materials, Prague, Czech Republic, August 25-29, 1997)
Reprint requests to: mojmir@ipm.cz

Defect and atomic ordering studies in highly deformed and disordered $Fe_{72}Al_{28}$ alloy

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[2]*Department of Low Temperature Physics,*
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Abstract

The results of positron annihilation and Mössbauer spectroscopies in $Fe_{72}Al_{28}$ alloy with different structural morphology obtained by severe deformation and by quenching from a high temperature are presented and theoretically analyzed with the help of first-principles electronic structure calculations. It is shown that the dominant component of 184 ps in the positron lifetime spectrum of a highly disordered sample corresponds to the annihilation in vacancies which prefer to occupy that sublattice in $D0_3$ superstructure containing both aluminium and iron atoms. Important changes in vacancy concentration take place only when an annealing above $D0_3 \leftrightarrow B2$ (823 K) is performed. Temperature treatment below this phase transformation temperature is characterized above all by changes in the density of dislocations.

(Acta Mater. **45** (1997), 2147-2154)
Reprint requests to: mojmir@ipm.cz

Calculation of theoretical strength of solids by linear muffin-tin orbitals (LMTO) method

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Abstract

Ab initio calculation of theoretical strength of Si, Na, W, Cu and Ir cubic crystals under three-axial tension is performed using the linear muffin-tin orbitals (LMTO) method in atomic-sphere approximation (ASA). Computed values are compared with those obtained previously by other methods. The results obtained verify some semi-empirical pair and many-body potential approximations.

(Mat. Sci. Eng. A **234-236** (1997), 370-372;
Proc. 11th Conf. on the Strength of Materials, Prague, Czech Republic, August 25-29, 1997)
Reprint requests to: mojmir@ipm.cz

Ballistic Transport and Electronic Structure

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Abstract

The role of the electronic structure in determining the transport properties of ballistic point contacts is studied. The conductance in the ballistic regime is related to simple geometrical projections of the Fermi surface. The essential physics is first clarified for simple models. For real materials the band structure is taken into account using parameter-free local-spin-density approximation calculations. In magnetic metallic multilayers the electronic structure gives rise to a large difference in conductance between the parallel and anti-parallel configurations. For Co/Cu and Fe/Cr multilayers the dependence of the conductances on the layer thicknesses and the crystal orientations is investigated for the geometries with the current perpendicular, parallel and at an angle to the interface planes. In spite of there being no spin-dependent scattering at defects, the ballistic giant magnetoresistance effects in the perpendicular geometry can be as large as 120% and 230% in Co/Cu and Fe/Cr multilayers, respectively.

(submitted to Physical Review B)
Preprints available from: schep@natlab.research.philips.com

Quasi-Particle Spectra of High Temperature Superconductors

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and O. Jepsen[(c)]
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[(c)] *Max-Planck-Institut für Festkörperforschung,*
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Abstract

We present a semi-phenomenological approach to calculating the quasi-particle spectra of High Temperature Superconductors (HTSC). It is based on a particularly efficient parametrization of the effective electron-electron interaction afforded by the Density Functional Theory for superconductors and a Tight-Binding-Linearized-Muffin-Tin-Orbital (TB-LMTO) scheme for solving the corresponding Kohn-Sham-Bogoliubov-de Gennes equations. We illustrate the method by investigating a number of site and orbital specific, but otherwise phenomenological models of pairing in quantitative detail and discuss the results using BCS-like theory and insights to the HTSC band structures. We compare our results for the gap function on the Fermi surface with those deduced from photoemission experiments on single crystals of $YBa_2Cu_3O_7$. We also compare our predictions for the temperature dependence of the specific heat with measurements and conclude, provisionally, that the dominant pairing interaction operates between electrons of opposite spin, on nearest neighbour Cu sites in $d_{x^2-y^2}$ orbitals.

(Submitted to Phys. Rev. B)
Manuscripts available from: W.M.Temmerman@dl.ac.uk

The Van Hove Scenario and the 'Eight-Band Model' for High T_c Superconductors

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Abstract

We investigate the doping dependence of the superconducting transition temperature, T_c, for the cuprate superconductors on the basis of a semi-phenomenological description of pairing and the ' eight-band model' for their electronic structure in the normal state. We find, in agreement with experiments, that T_c rises and then falls steeply as the number of holes increases from under- to over-doping. We identify the characteristic saddle points of the 'eight-band model' as the cause of this behaviour. Because our description of pairing is semi-phenomenological, we do not shed new light on the physical mechanism of pairing, nevertheless we present new evidence that the above Van Hove-like scenario is an essential feature of superconductivity in these materials.

(Submitted to Phys. Rev. Lett.)

Manuscripts available from: Z.Szotek@dl.ac.uk

Electronic structure and elastic properties of strongly correlated metal oxides from first principles: LSDA+U, SIC-LSDA and EELS study of UO_2 and NiO

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Abstract

We compare experimentally observed electron energy loss spectra (EELS) of uranium dioxide UO_2 and nickel monoxide NiO with the results of *ab-initio* calculations carried out by using a method combining the local spin density approximation and the Hubbard U term (the LSDA+U method). We show that by taking better account of strong Coulomb correlations between electrons in the $5f$ shell of uranium ions in UO_2 and in the $3d$ shell of nickel ions in NiO it is possible to arrive at a better description of electron energy loss spectra, cohesive energies and elastic constants of both oxides compared with local spin density functional theory. For NiO we also compare the LSDA+U results and EELS spectra with a self-interaction corrected LSDA calculation.

(Submitted to Phys. Stat. Sol. / M. Rhule special issue)

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Low-field galvanomagnetic properties of aluminum-based dilute alloys

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Abstract

We report a systematic study of low-field galvanomagnetic properties of aluminum-based dilute alloys with $3d$ and $4sp$ impurities. The low-field magnetoresistivity tensor is determined by exactly solving the linearized Boltzmann equation for the anisotropic vector mean free path, without using any adjustable parameter. Our method of calculation is based on the on-Fermi-sphere approximation which allows us to combine the full anisotropy of the host Fermi surface, obained by the four-orthogonal-plane-wave method, with the impurity scattering phase shifts, evaluated by self-consistent local-density-functional impurity-in-jellium calculations. Our results for the Hall coefficient and the magnetoresistance are in good agreement with the experimental data.

(Accepted by J. Phys.: Condens. Matter)
Preprints from: nstefan@atlas.uoa.gr

Anomalous shift of the 1075 cm^{-1} oxygen-hydrogen defect in silicon

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Abstract

First principles calculations are carried out on i) hydrogen and ii) water molecules trapped near an interstitial oxygen atom in Si. We find that it is possible for these molecules to cause an upward shift in the antisymmetric stretch mode of O_i when H is replaced by D, which could explain the anomalous shift in the 1075 cm^{-1} O-H related local vibrational mode. Both these molecules lead to modes in the 3500-4000 cm^{-1} region but those of the H_2 lie close to those recently detected using Fourier transform infra-red spectroscopy.

(To appear in the Proceedings of the 19th ICDS, Aveiro - Portugal)
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Theory of threading edge and screw dislocations in GaN

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Abstract

The atomic structures, electrical properties and line energies for threading screw and threading edge dislocations of wurtzite GaN are calculated within the local–density approximation. Both dislocations are electrically inactive with a band gap free from deep levels. These results are understood to arise from relaxed core structures which are similar to $(10\bar{1}0)$ surfaces.

(To be published in *Physical Review Letters*)
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The N2 and N4 optical transitions in diamond: a breakdown of the vacancy model

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Abstract

The vacancy model for impurity vacancy defects in semiconductors assumes that the ground and low energy excited states are derivable from the four sp^3 hybrid orbitals on atoms bordering the vacancy. There are many cases where this model works but we describe here a counter-example concerning the lowest excited state of the [V-N_3] defect in diamond. It is shown that a shallow electron trap, localised outside the vacancy, is involved in the first excited state and responsible for the N2 and N4 optical bands associated with the defect.

(To appear in Phys. Rev. B, **56**)
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Theory of gold-hydrogen complexes in silicon

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Abstract

Spin-polarised local density functional cluster calculations are carried out on substitutional Au and Au–H_n complexes in Si. Slater's transition argument and Janak's theorem are combined to discuss the donor-acceptor levels of the defects and the results are compared with experiment. The calculations give a good account of the donor/acceptor levels of Au and the Au–H pair. They confirm that the G1, G2 and G4 levels are due to Au–H. Au–H_3 is found to be an electrically inactive defect and may be identified with the passive defect found experimentally.

(To be published Materials Science Forum, in press)
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Comment on Electronic structure of the N-V center in diamond

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Abstract

It is argued that the model advanced by Lenef and Rand [Phys. Rev. B **53**, 13441, (1996)] for the nitrogen-vacancy center in diamond, exhibiting the 1.945 eV luminescence is incorrect. Lenef and Rand argue that the electronic ground state consists of two electrons occupying a_1 states localised on N and C respectively and are 'decoupled' from the other three electrons occupying gap states. This model is inconsistent with experiments and other theoretical works. The existence of a Jahn-Teller state 46 cm^{-1} above the excited state of the defect is also in conflict with previous experiments.

(To be published Physical Review B)
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Ab initio modelling of defects in non-metallic systems

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Abstract

The simplicity of density functional methods and the increasing availability of computer processing power have enabled *ab initio* modelling techniques to be applied to very large systems. Our own code, AIMPRO, enables the relaxation of clusters as large as 800 atoms while still maintaining full self-consistency and a realistic basis. Moreover, the use of pseudopotentials means that applications to Ge clusters are no more time consuming than those built of C. An important thrust of our work has been the computation of accurate vibrational modes of defects and this has caught the attention of several prominent experimental groups with the consequence that joint studies of rather complicated defects have been successfully made. We outline the procedures used and discuss applications that have been made to a wide variety of non-metallic systems such as vacancy defects in diamond, interstitial centres in silicon and carbon-related defects in III-V materials.

(To be published, Bunsen Society, 1997)
Manuscripts available from: jones@excc.ex.ac.uk

Origin of Giant Magnetoresistance–bulk or interface scattering

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Abstract

Ab initio calculations of giant magnetoresistance (GMR) of Co/Cu (001) multilayers are presented. Starting from density functional theory the electronic structure of the multilayer and the scattering at impurities are described by means of a new Greens function method. It will be shown that due to the existence of quantum well and interface states in multilayers GMR is strongly affected by scattering centers at the interface. Results for a multilayer with a Cu thickness corresponding to the first antiferromagnetic maximum of the interlayer exchange coupling (IEC) are discussed in detail.

(Submitted to Phys. Rev. Lett.)
Manuscripts available from: mertig@PTPRS2.phy.tu-dresden.de

Self-consistent interface properties of d and s wave superconductors

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Abstract

We develop a method to solve the Bogoliubov de Gennes equation for superconductors self-consistently, using the recursion method. The method allows the pairing interaction to be either local or non-local corresponding to s and d-wave superconductivity, respectively. Using this method we examine the properties of various S-N and S-S interfaces. In particular we calculate the spatially varying density of states and order parameter for the following geometries (i) s-wave superconductor to normal metal, (ii) d-wave superconductor to normal metal, (iii) d-wave superconductor to s-wave superconductor. We show that the density of states at the interface has a complex structure including the effects of normal surface Friedel oscillations, the spatially varying gap and Andeev states within the gap, and the subtle effects associated with the interplay of the gap and the normal van Hove peaks in the density of states. In the case of bulk d-wave superconductors the surface leads to mixing of different order parameter symmetries near the interface and substantial local filling in of the gap.

(Submitted to Phys. Rev. B)
Manuscripts available from: james@annett.phy.bris.ac.uk
(The article is also available on the preprint server-(cond-mat 9708086))

Molecular Dynamics description of Silver adatom diffusion on Ag(100) and Ag(111) surfaces

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Abstract

The self-diffusion processes of single adatoms on Ag(100) and Ag(111) surfaces have been studied using molecular-dynamics simulations and a many- body potential derived in the framework of the second-moment approximation to the tight-binding model. Our results for the (100) surface indicate that, although the migration energy for hopping is lower than that of the exchange mechanism, the exchange diffusion is higher than hopping diffusion for temperatures above 600 K. The migration energy for the hopping mechanism is in very good agreement with the experiment and the results of ab-initio calculations. We also find that for the Ag(111) face the dominant mechanism is the hopping, which exhibit Arrhenius behaviour with two distinct temperature ranges, corresponding to two different migration energies. The diffusion in the high temperature region is mainly due to correlated jumps requiring an activation energy which is in excellent agreement with the experimental data. In addition the temperature dependence of the mean-square-displacements and the relaxations of both surface atoms and adatoms are presented and compared with previous studies.

(To be published in Computational Materials Science (1997))
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This work was supported by the Ψ_k-Network.

Effects of nonstoichiometry in $A_{3-x}C_{60}$ (A= K, Rb)

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Abstract

We perform self-consistent Hartree calculations for a model of $A_{3-x}C_{60}$ (A= K, Rb) to study the effects of the presence of vacancies. We find that the strong vacancy potential is very efficiently screened and the density of states is only weakly influenced by the presence of vacancies.

(J. Electrochem. Soc.)
Manuscripts available from: gunnar@and.mpi-stuttgart.mpg.de

First Principles Calculation of Cu(100)c(2×2) 3*d* Surface Alloys Formation Energy: Improvement by Generalized Gradient Approximation

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Abstract

The energetics of Cu(100)c(2×2)3d 2D surface alloys are reinvestigated by use of the generalized gradient approximation (GGA). The calculations improve former results based on the local-spin-density approximation, in agreement with experiments. But trend analyses show that the success is principally due to an improved lattice constant of Cu substrate calculated by GGA. The mechanisms of the energetics are discussed.

(Physica B **237-238**, 359 (1997).)
Manuscripts available from: bluegel@iff064.iff.kfa-juelich.de

Electronic Structure of two–dimensional magnetic alloys: c(2×2) Mn on Cu(100) and Ni(100)

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Abstract

Half a monolayer of Mn deposited above 270 K on the (100) surfaces of Cu and Ni form ordered surface alloys of c(2×2) structure. Their electronic structure is studied in a combined experimental and theoretical work. The experimental approach, combining angle-resolved photoemission and inverse photoemission, characterizes both systems as ideal cases of well-ordered magnetic surface alloys and permits a complete measurement of quantum numbers. A large atomic-like splitting between majority and minority Mn states is measured: 5.5 eV for the Cu-based system and 5.25 eV for the Ni-based system. The large splittings are direct evidence that Mn develops a high local magnetic moment in these systems. Our first-principles band-structure calculations of $c(2\times2)$ CuMn/Cu(100) and $c(2\times2)$ NiMn/Ni-(100) corroborate this finding and give values of 3.75 μ_B and 3.5 μ_B, respectively, for the Mn moments. We find that the measured splittings are even larger than the ones calculated from first principles. The difference amounts to 2.7 eV and 1.7 eV for the Cu-based and Ni-based system, respectively. We conclude that the splitting measured in photoemission and inverse photoemission is increased by a Coulomb energy U due to the enhanced localization of the Mn d states in the surface alloy. This high localization can be quantified by the small band dispersion of 110±60 meV measured for the Mn minority d band in the Cu-based system.

(Phys. Rev. B **55**, 5404 (1997).)
Manuscripts available from: bluegel@iff064.iff.kfa-juelich.de

Correlation satellite driven by reduced dimensionality

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Abstract

Photoelectron spectra of the ordered two-dimensional surface alloy $c(2 \times 2)$ CuMn/Cu-(100) show distinct valence band spectral features at higher binding energy, which are most pronounced for the optimal CuMn $c(2 \times 2)$ superstructure. Our analysis comprising resonant photoemission, core-level photoemission, inverse photoemission, first-principles local-density calculations, and configuration-interaction cluster-model calculations shows that the data represent the first observation of a valence-band satellite caused by the increased electron correlation in an ultrathin metal film.

(Accepted at Europhys. Lett.)

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Structure, Growth and Magnetism of Mn on Cu(110)

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Abstract

We found a new two-dimensional, ordered surface alloy: Cu(110)-c(2 × 2)-Mn. The structure and composition of this surface compound were determined by quantitative low energy electron diffraction analysis, which shows a large buckling in the surface alloy layer. The Mn atoms buckle outwards and the Cu atoms inwards with a total buckling amplitude of 0.22Å (17.2% of the ideal interlayer distance of Cu(110)). The results are compared to *ab initio* total energy and force calculations. The theoretically determined buckling of 16.3% reproduces the experimental situation. The calculations predict a large magnetic moment for Mn of $M = 3.82\mu_B$. A hypothetical nonmagnetic Cu(110)-c(2 × 2)-Mn surface alloy shows no buckling (< 1%), proving that the buckling is due to the magneto-volume effect of Mn. Investigation of the growth shows, that for substrate temperatures above 180 K, deposition of submonolayer amount of Mn leads to the formation of a c(2 × 2) superstructure. A well-ordered structure at 0.5 monolayer (ML) was observed in the temperature range between 270 K and 350 K. For films above 1 ML, a 16 × 1 superstructure was observed giving evidence for a buckled, Mn rich top layer. The influence of Mn on the chemical reactivity of the Cu(110) surface with O_2 is discussed. We also investigated the work function change upon surface alloy formation. The *ab initio* calculations predict a work function lowering of about 0.5 eV and we identified the magnetism of Mn as basic origin of the work function change. The results are compared to the Cu(100)-c(2 × 2)-Mn surface alloy.

(Accepted at Phys. Rev. B.)
Manuscripts available from: bluegel@iff064.iff.kfa-juelich.de

Acknowledgement includes: Program Training and Mobility of Researchers 'Interface Magnetism' (Contract: FMRX-CT96-0089) of the European Union.

Total Energy spectra of complete sets of magnetic states for Fe films on Cu(100)

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Abstract

Based on a total energy search among the complete set of 2^{n-1} collinear spin-states for n ($n = 1, \cdots, 6$) monolayers (ML) of fcc-Fe on Cu(100) by use of the generalized gradient approximation (GGA) combined with the full-potential linearized augmented plane-wave (FLAPW) method, we find ferromagnetism for 1, 2, and 3ML fcc-Fe/Cu(100), bilayer antiferromagnetism for even number of layers (4ML and 6ML), and the coexistence of several spin-states for an odd number of layers (5ML). The results are consistent with the experimental situation and a possible spin-spiral ground-state of fcc-Fe. Effects of band-narrowing, relaxations, interface mixing and surface steps are addressed.

(Phys. Rev. Lett. **57**, 507 (1997).)
Manuscripts available from: bluegel@iff064.iff.kfa-juelich.de

Chemical identification of atoms at multicomponent surfaces on an atomistic scale: $CoSi_2(100)$

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Abstract

We found a $(\sqrt{2}\times\sqrt{2})$R45° surface reconstruction on the plateau of three-dimensional Cobalt silicide islands epitaxially grown on Si(100), which has an inhomogeneous occupation of the lattice sites. We combined voltage dependent scanning tunneling microscopy (STM) with *ab initio* electronic structure calculations and resolve the chemical identity of the surface atoms. We found that Co atoms are imaged for bias voltages probing the unoccupied states above +0.5 eV and vice versa Si atoms are imaged below +0.5 eV, counterintuitive to the simple interpretation based on the electronic structure of bulk Co and Si.

(Phys. Rev. B. **55**, R13444 (1997).)
Manuscripts available from: bluegel@iff064.iff.kfa-juelich.de

Dichroism and Spin Information in Soft X-ray Emission

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Abstract

We demonstrate that soft x-ray emission excited at the L_3 threshold of Fe metal maps the spin-resolved local partial density of occupied states. In contrast to earlier predictions, the spin resolution is achieved primarily by selective excitation. Circular magnetic dichroism in the x-ray emission process is also shown to have an influence on the final state spin contribution to the spectra. The importance of taking reflection, self-absorption of the incoming and outgoing radiation and the Auger decay channels into consideration in the data analysis is emphasized.

(Accepted at Solid State Communications)
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90 Degree Magneto-Optical Kerr Rotation: Criteria of Existence and Features

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Abstract

An analysis is made of the general properties required for a magnetic material to exhibit 90° polar Kerr rotation. The approach is phenomenological invoking general properties of the dielectric tensor relating to the energy absorption. It is shown that the effect exists only if the absorptive part of the dielectric tensor is equal to zero. We identify ranges of Re $\varepsilon_{xx}(\omega)$ and Im $\varepsilon_{xx}(\omega)$ where 90° rotation is realized, and study the Kerr ellipticity, the reflectivity, and the magneto-optical signal in these regions.

(Physical Review **B56**, N 18 to be published)
Manuscripts available from: uspenski@td.lpi.ac.ru

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Cation-Aided Joining of Surfaces of β-Si_3N_4. Structural and Electronic Aspects

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Abstract

An atomic scale approach has been applied to both physical-and electronic-structure examination of stable surfaces of β-Si_3N_4. Sterical constraints are shown to prevent the (001) surface from effective chemical reaction with the interface. The theoretical surface-to-surface bonding is investigated using a periodical tight-binding approach. Based on the interpretation of density of states, the balance of the number of states and electrons is performed for stoichiometric Si_3N_4, ideal N-terminated (110) surfaces, oxygen overlayered (110) slabs, and for the metal monolayer, with which slabs are brought into contact. The stable electronic configuration, reached when the cation binds to the interface, represents the electronic reason behind the diffusion of the additive and/or impurity atoms toward grain boundaries. The different bonding propensity of (001) and (110) surface implies that effective bonding of planes parallel to the **c**-direction to the interphase restrains the crystal from the growth in the lateral direction. Conversely, geometry constrained bonding of (001) planes allows the crystal growth producing the rod-shaped β-grains.

(Accepted in J. Amer. Ceram. Soc.)
Manuscript available from: lubomir.benco@unifr.ch

Work performed at Institute of Inorganic Chemistry was supported by the U.S. - Slovak Scientific and Technological Program No. 94039 and Slovak Grant Agency VEGA (grant No. 1172).

Addition spectra and shell filling in semiconductor quantum dots: Role of electron-electron interaction

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Abstract

The addition spectra of semiconductor quantum dots in the presence of magnetic field are studied through a theoretical scheme that allows an accurate treatment of the single-particle states and electron-electron interaction up to large numbers of electrons. The calculated addition spectra exhibit the typical structures of Hund-like shell filling, and account for very recent experimental findings. A full three-dimensional description of Coulomb interaction is found to be essential for predicting the conductance characteristics of these semiconductor macroatoms.

(Submitted to Phys. Rev. Lett.)
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Structure and dynamics of Rh surfaces

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Abstract

The lattice relaxation, surface phonon spectrum, surface energy as well as work function for Rh(100) and Rh(110) surfaces are calculated by using density-functional theory and employing the full-potential linearized augmented plane wave method. Both the local density approximation and the generalized gradient approximation to the exchange-correlation functional are considered. The force constants are obtained from the directly calculated atomic forces and the temperature dependence of the surface relaxation is evaluated by minimizing the free energy of the system. The anharmonic effects are taken into account within the quasiharmonic approximation. The importance of contributions from different phonons to the surface relaxation is analyzed.

to the surface relaxation is analyzed.

PACS numbers: 68.35.Bs, 63.20.Ry, 75.30.Pd.

(submitted to Phys. Rev. B)
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Novel diffusion mechanism on the GaAs (001) surface: the role of adatom–dimer interaction

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Abstract

Employing first principles total energy calculations we have studied the behavior of Ga and Al adatoms on the GaAs(001)–$\beta 2$ surface. The adsorption site and two relevant diffusion channels are identified. The channels are characterized by different adatom–surface dimer interaction. Both affect in a novel way the adatom migration: in one channel the diffusing adatom jumps across the surface dimers and leaves the dimer bonds intact, in the other one the surface dimer bonds are broken. The two channels are taken into account to derive effective adatom diffusion barriers. From the diffusion barriers we conclude a strong diffusion anisotropy for both Al and Ga adatoms with the direction of fastest diffusion parallel to the surface dimers. In agreement with experimental observations we find higher diffusion barriers for Al than for Ga.

(submitted to Phys. Rev.)
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Study of Strain and Temperature Dependence of Metal Epitaxy

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Abstract

Metallic films are important in catalysis, magneto-optic storage media, and interconnects in microelectronics, and it is crucial to predict and control their morphologies. The evolution of a growing crystal is determined by the behavior of each individual atom, but technologically relevant structures have to be described on a time scale of the order of (at least) tenths of a second and on a length scale of nanometers. An adequate theory of growth should describe the atomistic level on very short time scales (femtoseconds), the formation of small islands (microseconds), as well as the evolution of mesoscopic and macroscopic structures (tenths of seconds).

The development of efficient algorithms combined with the availability of cheaper and faster computers has turned density functional theory (DFT) into a reliable and feasible tool to study the microscopic aspects of growth phenomena (and many other complex processes in materials science, condensed matter physics, and chemistry). In this paper some DFT results for diffusion properties on metallic surfaces are presented. Particularly, we will discuss the current understanding of the influences of strain on the diffusion (energy barrier and prefactor) of a single adatom on a substrate.

A DFT total energy calculation by its nature is primarily a *static* calculation. An accurate way to describe the spatial and temporal development of a growing crystal is given by kinetic Monte Carlo (KMC). We will describe the method and its combination with microscopic parameters obtained from *ab initio* calculations. It is shown that realistic *ab initio* kinetic Monte Carlo simulations are able to predict an evolving mesoscopic structure on the basis of microscopic details.

In: "Morphological Organisation during Epitaxial Growth and Removal", Eds. Z. Zhang, M. Lagally. World Scientific, Singapore 1998.
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Ab initio studies of H_2O adsorption on the TiO_2 (110) rutile surface

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Abstract

The geometrical, electronic and energetic properties of the dissociative (1×2) adsorption of H_2O on the rutile TiO_2 (110) surface are calculated by applying the Full Potential Linearized Plane Wave method within the local density approximation. Two possible (1×2) adsorption geometries are investigated. We find a general reduction of the bond lengths at the surface and an inwards relaxation of the surface plane. Whereas the O–H bond lengths are very similar for both geometries and comparable to that of the free H_2O molecule, the corresponding Ti–O bond lengths differ by 0.25 Å, expressing the amphoteric character of the OH groups. Concerning their ground state energies, both adsorption geometries are practically equal. Additional OH surface peaks appear in the density of states, lowered by about -2 eV from the main parts of the O-*s* and -*p* bands of the clean surface. Upon adsorption, the lowest Ti *d*-like states of the conduction band of the clean surface become occupied. Compared to the clean surface, the work function is strongly reduced by about 3 eV.

(submitted to Surface Science)
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Ab initio study of the CO adsorption on NiAl (110) and Pt (100)

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Abstract

We investigate the interaction of CO with the NiAl (110) surface adsorbed in a p (2×1) geometry by means of the Full-potential Linearized Augmented Plane Wave (FLAPW) method. For the fully relaxed surface interacting with the adsorbate, the driving bonding mechanisms on different adsorption sites are discussed. For comparison, we also present results for the c (2×2) CO adsorption on the Pt (100) surface. Calculated charge transfers and surface core level shifts are a sensitive probe of the adsorption geometry.

(Submitted to Surf. Sci.)
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* Work supported by the Austrian Science Foundation, Project No. P11508-PHY, by the City of Vienna (Jubiläumsstiftung), and by the Center for Computational Materials Science.

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8 HIGHLIGHT OF THE MONTH

Forces and Lattice Relaxations Calculated by a Full-Potential KKR-Green's Function Method

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Abstract

In this review we demonstrate that a recently developed full-potential KKR-Green's function method allows an efficient calculation of forces and lattice relaxations in transition metals. The forces can be readily evaluated by the ionic Hellmann-Feynman theorem, while the Green's functions for shifted positions can be obtained by angular momentum transformations. As applications we calculate the lattice relaxations around impurities in Cu and Al. Moreover, we show that the method allows to determine the phonon frequencies of transition metals from a single self-consistent calculation of the coupling constants in real space and present results for ferromagnetic Ni.

8.1 Introduction

The KKR method of electronic structure calculations, introduced by Korringa [1] and Kohn and Rostoker [2], is well known for its mathematical rigorousness and elegance. By taking advantage of multiple scattering theory, the method clearly separates the scattering properties of the single potentials from the geometrical structure of the system. Moreover, the size of the resulting algebraic equations is optimally small. On the other hand, the KKR theory is subtle and complex, so that even the evaluation of the free-space structure constants represents a problem in itself. Moreover, its numerical implementation for band-structure calculation is neither easy nor efficient. This has lead to the fact that the linear methods, like LMTO, ASW, and LAPW, received over the years considerably more attention.

The Green's function (GF) version of the KKR method was introduced by Beeby and others [3, 4, 5] and has later-on been used by the authors in extensive calculations for impurities in metals [6, 7, 8]. The GF version is particularly useful for density-functional calculations and avoids part of the disadvantages connected with the band-structure problem. For instance, the integration over all occupied states can be efficiently performed by a contour integration in the complex energy plane, so that for ground state calculations the time consuming evaluation of the eigenvalues is avoided. Moreover, the availability of the Green's function opens up the way to applications like linear-response problems, disorder in alloys, e.g., within the CPA, and electronic transport in solids [9].

Unfortunately, most KKR calculations up to now have been restricted to spherical potentials of muffin-tin or ASA form. This is a serious limitation of the theory since the structural optimization of a system requires a full-potential description. In this highlight we will review our recent work on extending the KKR-GF method into a full-potential scheme and then show that this allows an accurate and efficient calculation of forces and lattice relaxations. The applicability of full-potential KKR schemes has been widely discussed in the literature [10, 11, 12, 13, 14, 15, 16] and in the view of most authors, this is simply a question of the convergence of the angular momentum expansion which may differ in different versions of the full-potential KKR theory. In view of our experience with the calculated results described below the convergence of the full-potential KKR-GF method described in section 8.2 seems not to be a problem in practical calculations. In contrast to most other methods, the force calculation is very easy within the KKR formalism, while the description of lattice relaxation is more complicated. One should also note that the KKR theory is very different from methods based on localized or mixed orbitals. For instance, the overlap between orbitals on neighboring sites does not enter at all nor has an interstitial region to be introduced. Instead, the solutions in each cell are constructed by angular momentum expansions, and the proper boundary conditions are satisfied by the solution of the Dyson equation.

The outline of the paper is as follows: First we shortly describe the full-potential extension of the KKR formalism in Sec. 8.2 and sketch the calculation of forces and lattice displacements in Sec. 8.3. We then present results for the lattice relaxations around impurities in metals (Sec. 8.4) and demonstrate that the same method can also be used to calculate phonons in real space (Sec. 8.5). Finally we conclude this highlight with a short summary and an outlook on further developments of the KKR method.

8.2 Full-Potential KKR-Green's Function Method

In the sense of multiple-scattering theory, the whole space is divided into non-overlapping and space-filling cells around each atomic site $\mathbf{R}^n$. The crystal Green's function, G, is then written in a cell-centered double expansion around the positions $\mathbf{R}^n$ and $\mathbf{R}^{n'}$ in different cells as [7]

$$G(\mathbf{r}+\mathbf{R}^n,\mathbf{r}'+\mathbf{R}^{n'};E) = \delta_{nn'}G_s^n(\mathbf{r},\mathbf{r}';E) + \sum_{L,L'} R_L^n(\mathbf{r};E)G_{LL'}^{nn'}(E)R_{L'}^{n'}(\mathbf{r}';E) \ , \qquad (2)$$

where $G_s^n(\mathbf{r},\mathbf{r}';E)$ represents the Green's function for a single potential confined inside the cell n in otherwise free space. In the above equation, the position vectors $\mathbf{r}$ and $\mathbf{r}'$ are restricted to the Wigner-Seitz cell and the index L denotes the angular momentum quantum numbers (ℓ,m).

The partial wave $R_L^n(\mathbf{r};E)$ is defined to be the solution of the Lippmann-Schwinger equation for a spherical wave $j_\ell(r\sqrt{E})Y_L(\hat{\mathbf{r}})$ incident on the potential $V^n(\mathbf{r})$ at the site $\mathbf{R}^n$

$$R_L^n(\mathbf{r};E) = j_\ell(r\sqrt{E})Y_L(\hat{\mathbf{r}}) + \int d\mathbf{r}'\, g(\mathbf{r},\mathbf{r}';E)V^n(\mathbf{r}')R_L^n(\mathbf{r};E)\ . \tag{3}$$

Here r is the length of the vector $\mathbf{r}$ and j_ℓ and Y_L are the spherical Bessel functions and the spherical harmonics, respectively, while $g(\mathbf{r},\mathbf{r}';E)$ is the free-electron Green's function being analytically known. In our full-potential KKR-GF method, the potentials and the partial waves are expanded in real spherical harmonics as

$$V^n(\mathbf{r}) = \sum_L V_L^n(r)Y_L(\hat{\mathbf{r}}) \tag{4}$$

and

$$R_L^n(\mathbf{r};E) = \sum_{L'} R_{L,L'}^n(r;E)Y_L(\hat{\mathbf{r}})\ . \tag{5}$$

The non-spherical part of the potential, $\Delta V^n(\mathbf{r}) = V^n(\mathbf{r}) - V_0^n(r)$, couples the angular momentum channels and leads to a system of coupled radial equations for the functions $R_{L,L'}^n$. These equations can be solved quite efficiently by the following procedure: First, calculate the solutions $R_\ell(r;E)$ corresponding to the spherical part of the potential, $V_0^n(r)$, and then calculate the full solutions iteratively by using an integral equation with $\Delta V^n(\mathbf{r})$ as perturbative potential. The corresponding Born series converges rapidly, and in most cases the second Born approximation is already sufficient for an accurate solution to the coupled radial equations [17].

When solving the radial and Poisson equations, the shape of the different cells is described by the shape functions $\Theta^n(\mathbf{r})$, being equal to 1 inside the cell n and vanishing everywhere else. These functions are also expanded in real spherical harmonics as

$$\Theta^n(\mathbf{r}) = \sum_L \Theta_L^n(r)Y_L(\hat{\mathbf{r}})\ . \tag{6}$$

The (possibly) distorted lattice is described in this work by keeping the same space division as in the undistorted lattice, but the shape functions are expanded around the shifted atomic positions. This procedure is working well for small displacements. For larger displacements, the use of the Wigner-Seitz construction in the distorted geometry would be a better choice to obtain the cells because it optimizes the angular momentum convergence of the wave functions.

While the ℓ-convergence of the expansion (6) is rather slow, the overall convergence is determined by the ℓ-cut-off of the wave functions $R_{L,L'}^n$ of Eq. (5). If here angular momenta up to $\ell_{\max}$ are taken into account, then the expansion of the charge density and the potential (Eq. 4) includes only contributions up to $2\ell_{\max}$, while the evaluation of the Coulomb and exchange-correlation terms in the potential energy requires ℓ-terms up to $4\ell_{\max}$. Thus only shape functions $\Theta_L^n(r)$ up to $4\ell_{\max}$ are needed in the calculations.

The multiple scattering is contained in the second term of Eq. (2) through the so-called structural Green's function $G_{LL'}^{nn'}(E)$. This can be obtained by the Dyson equation

$$G_{LL'}^{nn'}(E) = G_{LL'}^{0,nn'}(E) + \sum_{n''L''L'''} G_{LL''}^{0,nn''}(E)\Delta t_{L''L'''}^{n''}(E)G_{L'''L'}^{n''n'}(E)\ , \tag{7}$$

which can be written in matrix notation as

$$\mathcal{G} = \mathcal{G}^0 + \mathcal{G}^0\Delta\mathrm{t}\mathcal{G}\ , \tag{8}$$

where $\mathcal{G}^0$ is the structural Green's function of the reference system and $\Delta\mathsf{t} = \mathsf{t} - \mathsf{t}^0$ is the difference between the site-diagonal t-matrices of the reference and the actual systems. The latter one is given by

$$t^n_{LL'}(E) = \int d\mathbf{r}\, j_\ell(r\sqrt{E}) Y_L(\hat{\mathbf{r}}) V^n(\mathbf{r}) R^n_{L'}(\mathbf{r}; E) \ . \tag{9}$$

In the Dyson equation the non-spherical character of the full potentials $V^n(\mathbf{r})$ enters only in the non-diagonal elements $t^n_{LL'}$ of the t-matrix. From numerical point of view, they present no complication and no increase in computer time occurs in the solution of the Dyson equation. Thus only the single-site scattering problem, in particular the solution of the coupled radial equations, becomes more complicated. Therefore, the additional numerical effort needed for the full-potential KKR calculations compared to the muffin-tin KKR calculations scales only linearly with the number of non-equivalent potentials.

The electron density in cell n can be calculated from the imaginary parts of the site-diagonal elements of the Green's function

$$n^n(\mathbf{r}) = -\frac{1}{\pi} \int_{E_B}^{E_F} dE\, \mathrm{Im}\{ G(\mathbf{r} + \mathbf{R}^n, \mathbf{r} + \mathbf{R}^n; E)\} \ . \tag{10}$$

Here, the energy integral extends over all occupied states from a suitably chosen energy, E_B, between the valence band and the core states up to the Fermi energy, E_F. By using analytical properties of the Green's function, one can replace the energy integration in Eq. (10) by a contour integral in the complex energy plane, which can be evaluated with much fewer energy points. In this case, the full structure of the Green's function close to E_F is important, so that the energy mesh has to be more dense in this region. A recent extension to finite temperatures, using a Fermi-Dirac distribution for the occupation function, has the additional advantage that only complex energies are needed. The closest one to the real axis is given by $E_F + i\pi kT$, the value of the first Matsubara energy [18].

8.3 Calculation of Forces and Lattice Relaxations

From the self-consistent charge densities and potentials, one can calculate the forces on the atoms by using an ionic version of the Hellmann-Feynman theorem [19]

$$\mathbf{F}^n = Z^n \left. \frac{\partial V^n_C(\mathbf{r})}{\partial \mathbf{r}} \right|_{\mathbf{r}=\mathbf{0}} - \int d\mathbf{r}\, \rho^n_c(r) \frac{\partial V^n_{\mathrm{eff}}(\mathbf{r})}{\partial \mathbf{r}} \ , \tag{11}$$

where $V^n_C(\mathbf{r})$ is the Coulomb part of the effective one-electron potential, $V^n_{\mathrm{eff}}(\mathbf{r})$, in cell n due to all electrons and all other nuclei outside this cell. Clearly, the first term is the force on the nucleus, evaluated with spherical core charge densities, $\rho^n_c(r)$, and the second term is the force on the core electrons. It is obvious that the force on the nucleus is to a large extent compensated by a nearly equal but opposite force on the core electrons. In deriving the ionic force formula (11), it is only assumed that the unperturbed core density is spherically symmetric. Therefore, the force formula is equally valid if one makes the frozen core approximation or if the core states are allowed to relax retaining the spherical symmetry as it is usually done in all-electron calculations. Within the full-potential KKR-GF method, the force calculation does not require an additional effort, since the force is readily calculated from the $\ell = 1$ components of the

potentials and since all potential components are anyhow evaluated up to $2\ell_{\max}$ self-consistently. As a further advantage of the KKR theory, Pulay corrections to the force formula (11) arising from the restriction to a finite basis set vanish within the formalism so that in the full-potential description the evaluation of the force is straightforward.

Compared to this the evaluation of lattice relaxations becomes more complicated. This is due to the fact that one needs two angular momentum expansions, one for the unshifted ("ideal") position and one for the new shifted position. While the Green's function for such "interstitial" positions can also be evaluated by Brillouin zone integration, we use here a simpler method introduced by Lodder [20] which is, however, limited to small displacements. The structural Green's function $\mathcal{G}^0 = \{G^{0,nn'}_{LL'}\}$ of the host transformed to the shifted positions $\mathbf{R}^n + \mathbf{s}^n$ and $\mathbf{R}^{n'} + \mathbf{s}^{n'}$ is given by

$$\tilde{\mathcal{G}}^0 = \mathcal{U}\mathcal{G}^0\mathcal{U}^{-1} \ , \tag{12}$$

where the transformation matrix $\mathcal{U}$ is local in the site index and given by

$$U_{LL'}(\mathbf{s}^n) = 4\pi \sum_{L''} i^{\ell''-\ell'+\ell} C_{LL'L''} j_{\ell''}(s\sqrt{E}) Y_{L''}(\hat{\mathbf{s}}^n) \ , \tag{13}$$

where $C_{LL'L''}$ are the Gaunt coefficients and s is the length of the displacement vector $\mathbf{s}^n$. From the behavior of the Bessel functions for small arguments it can be seen, that for small displacements contributions arising from $\ell'' > 1$ can be neglected and that the matrix $\mathcal{U}$ couples angular momentum quantum numbers ℓ with $\ell \pm 1$, so one has to increase the angular momentum cut-off of the radial wave functions, e.g., from $\ell_{\max} = 3$ to $\ell_{\max} = 4$. An analogous expression holds for the unshifted t-matrix ,t^0, of the host in the shifted ℓ-representation, i.e., $\tilde{\mathrm{t}}^0 = \mathcal{U}\mathrm{t}^0\mathcal{U}^{-1}$, so that in the shifted representation the Dyson equation takes the form

$$\mathcal{G} = \tilde{\mathcal{G}}^0 + \tilde{\mathcal{G}}^0(\mathrm{t} - \tilde{\mathrm{t}}^0)\mathcal{G} \ , \tag{14}$$

which is quite analogous to Eq. (8) except for the complications arising from the $\mathcal{U}$-transformation and the resulting increase in $\ell_{\max}$.

In order to accelerate the determination of the equilibrium atomic positions from the *ab initio* forces, we use a lattice statics simulation based on the Kanzaki method [21]. In the harmonic approximation the displacement pattern $\mathbf{s}^{n'}$ is related to the force distribution, $\mathbf{F}^n$, by $\mathbf{F}^n = \sum_{n'} \Phi^{nn'}\mathbf{s}^{n'}$, where Φ denotes the coupling constant matrix of the defect system. By splitting this up, $\Phi = \Phi_0 + \Delta\Phi$, into the coupling constant matrix Φ_0 of the ideal crystal and the changes $\Delta\Phi$ induced by the defect, the Kanzaki forces are given by

$$\mathbf{F}^n_K = \sum_{n'} \Phi^{nn'}_0 \mathbf{s}^{n'} = \mathbf{F}^n - \sum_{n'} (\Delta\Phi)^{nn'}\mathbf{s}^{n'} \ , \tag{15}$$

In the simulation we use for the Born-von Karman parameters, Φ_0, values fitted to experimental phonon dispersion curves (they might as well be calculated; see below). The changes $\Delta\Phi$ are restricted to the first nearest neighbors of the defect and determined from *ab initio* calculations where only these first nearest neighbors are moved. The force pattern $\mathbf{F}^n_K$ is then applied on a hypothetical ideal lattice described by coupling constants Φ_0 and the displacements of all atoms are determined self-consistently. The resulting $\mathbf{s}^n$ are used as input in the next *ab initio* study and the whole procedure is repeated until all forces $\mathbf{F}^n$ are sufficiently small. Usually, one or

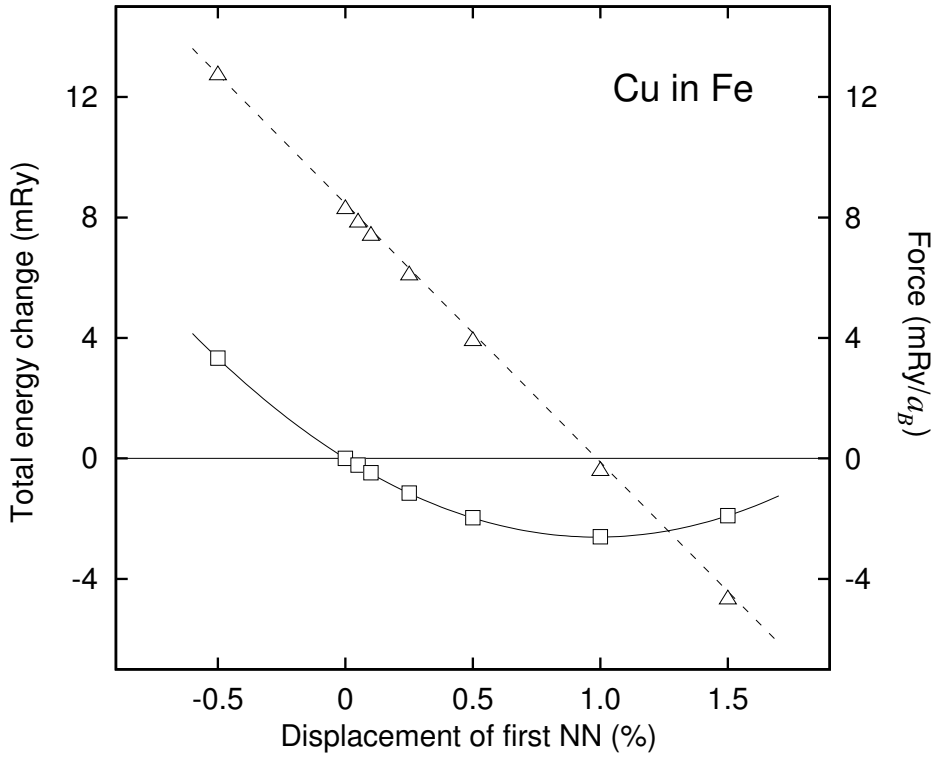

Figure 1: Total energy variation and radial force on a first nearest neighbor (NN) atom as a function of the change of the NN distance for a Cu impurity in bcc Fe. The dashed line shows the resulting force as obtained from a parabolic fit (solid line) to the calculated total energies (squares). The triangles are the *ab initio* forces calculated from the Hellmann-Feynman theorem (11).

two such iterations are enough. The Kanzaki forces $\mathbf{F}_K^n$ determined in this way can be used to determine the volume change ΔV induced by the defect, which can be experimentally obtained from lattice parameter measurements. The volume change is given in the Kanzaki model as

$$\Delta V = \frac{1}{3K} \sum_n \mathbf{F}_K^n \cdot \mathbf{R}^n \ , \tag{16}$$

where K is the bulk modulus of the ideal crystal.

To demonstrate the accuracy of the force calculation we discuss the relaxation of the first nearest neighbors for a Cu impurity in bcc Fe. Fig. 1 shows the variation of the total energy, if only the first nearest neighbor (NN) atoms are shifted. The dashed line shows the resulting force as obtained from a parabolic fit (solid line) to the calculated total energy values (squares) which compares very well with the *ab initio* forces (triangles) from the Hellmann-Feynman theorem (11). In both cases an outward relaxation of 1 % of the NN distance is obtained. The slope of the force curves can be used to determine the change $\Delta\Phi$ of the coupling parameters in Eq. (15).

8.4 Lattice Displacements around Impurities

We have performed calculations of lattice displacements around impurities in Al and Cu. The calculations were done using a cluster of 79 perturbed potential, so that an impurity and the 5

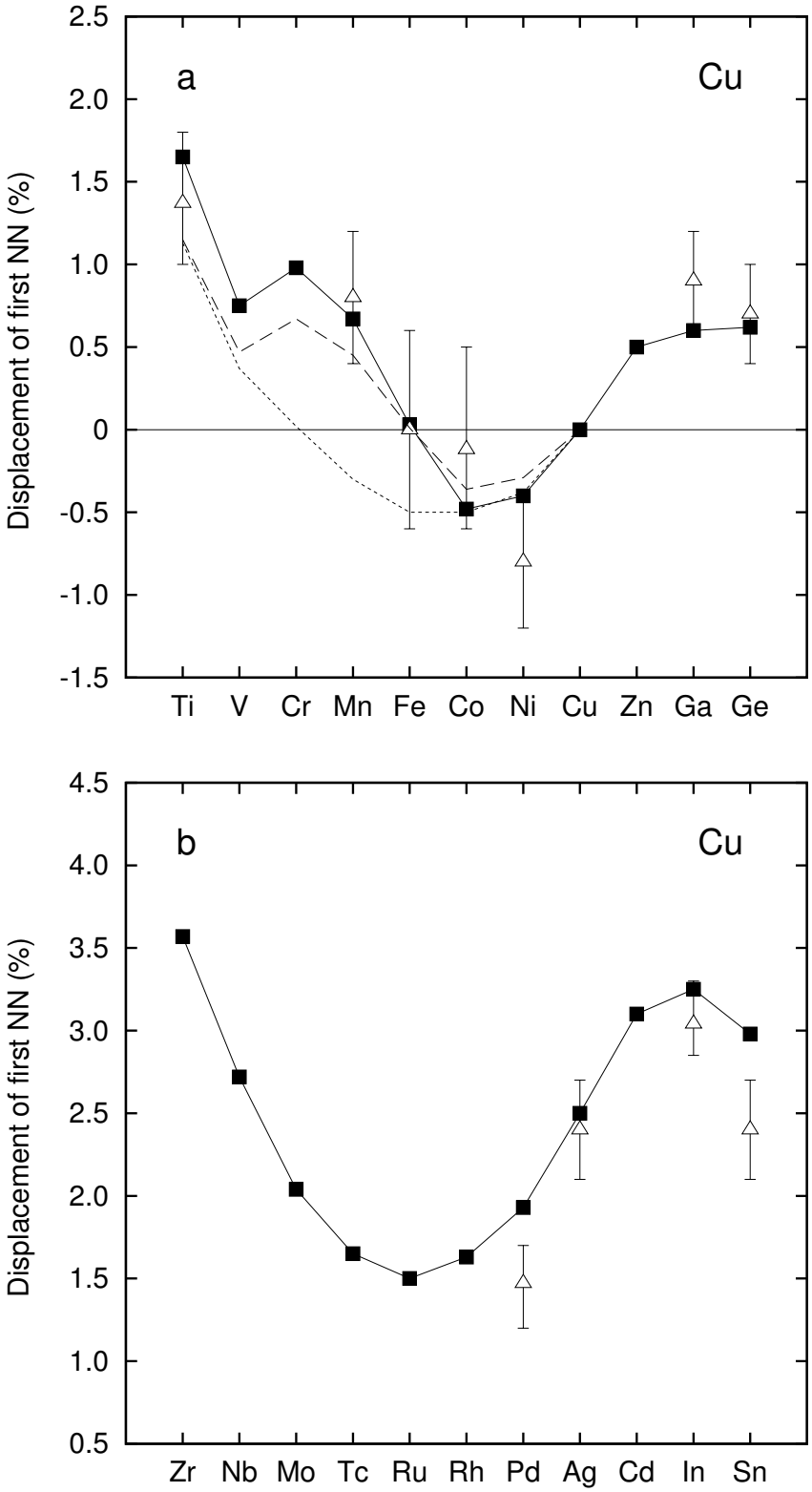

Figure 2: Calculated (solid squares) and experimental (open triangles with error bars) first NN shifts around 3d and 4sp (a) and around 4d and 5sp (b) substitutional impurities in Cu. The experimental values are the EXAFS data from Ref. [22]. Dashed and dotted lines are the results of calculations allowing only first NN relaxation for spin-polarized and non-spin-polarized cases, respectively.

NN fcc shells around the impurity were treated self-consistently. The theoretical values for the lattice constants obtained by a total energy minimization were used in both cases. The values 7.55 a.u. and 6.71 a.u. for Al and Cu, respectively, are somewhat smaller than the corresponding experimental values 7.65 and 6.82 a.u., representing a known failure of the LDA.

Fig. 2 shows the calculations of the first NN relaxations for a large number of impurities in Cu, i.e., for impurities of the 3d and 4sp series in Fig. 2a and of the 4d and 5sp series in Fig. 2b. The calculated values as given by the solid squares are in excellent agreement with EXAFS data of Scheuer and Lengeler [22]. The dashed line refers to a calculation where only the n.n. atoms were allowed to relax and all other atoms were fixed. This always underestimates the relaxation of the nearest neighbors.

As can be seen from Fig. 2, most impurities dilate the Cu lattice. Due to their large atomic sizes, this is particularly true for the 4d and 5sp impurities. The parabolic behavior across the 4d series is quite analogous to the behavior of the lattice constants of the 4d metals and can be explained by simple band filling arguments. In the 3d series the behavior is more complicated due to magnetism. While a non-spin-polarized calculation (dotted line) leads also to a parabolic trend, the large local moments of the Cr, Mn, and Fe impurities give rise to a pronounced magneto-volume expansion which also shows up in the EXAFS experiments. While the local moments strongly affect the lattice relaxations, the opposite is not true: The local moments are quite stable in Cu and are barely affected by the relaxation. For instance, the Mn-moment increases slightly from 3.39 μ_B to 3.42 μ_B when changing from the unrelaxed configuration to the relaxed one.

The calculations show that for the early 3d and 4d impurities in Cu a realistic description of the relaxations is only obtained, if the semicore s- and p-states are treated as valence states. For instance, for Ti or Zr impurity the first NN relaxations would be 50 or 60 % too large, if the semicore states were treated as core states. Therefore, we use in our calculations an extra long energy contour which includes both the valence states as well as the semicore states of the impurity. Thus contrary to the linearized methods the full-potential treatment of the semicore states is conceptually very easy within the KKR formalism.

As another example of our results, Fig. 3 shows the calculated volume changes per impurity in dilute Al-alloys with 3d and 4sp impurities [23]. With the exception of Mn the results are in a very good agreement with the lattice-parameter measurements [24]. Typically the 3d impurities show a large inward relaxation of the first nearest neighbors. For instance, for an Fe impurity the first nearest neighbors relax inward by 4 %, whereas in Cu the corresponding relaxation is practically zero. As a consequence in Al the local moments of the 3d impurities are strongly affected by the relaxations. In particular, the Fe-moment, calculated to be 1.5 μ_B in the ideal lattice geometry, vanishes in the relaxed configuration which is in agreement with susceptibility studies.

8.5 Calculation of Phonons

Many of the macroscopic properties of a crystalline material (e.g., specific heat, velocity of sound, thermal expansion) are determined by its vibrational degrees of freedom. The knowledge

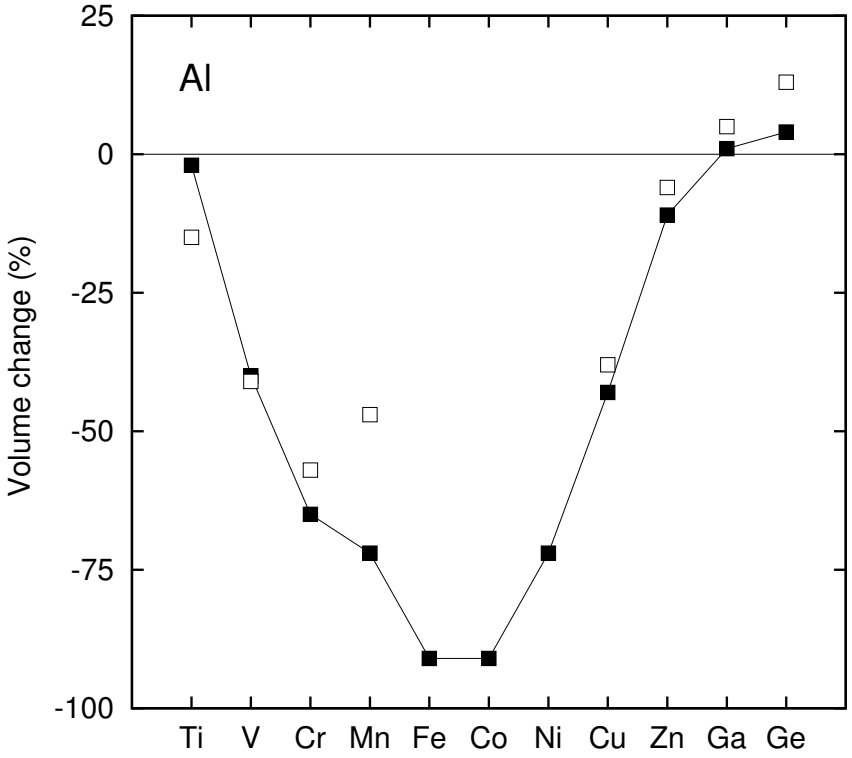

Figure 3: Calculated relative volume change per impurity in Al-based dilute alloys with 3d and 4sp impurities (filled squares). The results of lattice-parameter measurements [24] are also shown (empty squares).

of the phonon spectrum enables the calculation of the free energy within the quasi-harmonic approximation. The instabilities and peculiarities of the phonon spectrum may be used to understand phase transformations. Traditionally, phonon dispersion curves are calculated by lattice dynamics employing, e.g., the harmonic approximation, where one has to use some kind of analytical models for forces between atoms to obtain the force constant matrix. The first *ab initio* calculations for phonons in transition metals were performed using the frozen-phonon technique, where a (high symmetry) commensurable phonon **k**-vector is chosen and the atomic positions inside the corresponding supercell are displaced accordingly. From the calculated total energies one can then deduce the frequency of the chosen phonon mode. A recently introduced variational linear-response method avoids the use of supercells and can be applied to any phonon wave-vector [25]. For each **k**-value a new self-consistent calculation is required. In this highlight we show that the full-potential KKR-GF method can be used to determine the whole phonon spectrum of a transition metal from a single self-consistent calculation in real space.

We start from the definition of the Born-von Karman parameters Φ_{ij}^{mn} in the harmonic theory. By displacing the atom n in the direction j by the amount s_j^n, the force on the atom m in the direction i is given by $F_i^m = -\Phi_{ij}^{mn} s_j^n$. Thus we displace one atom by a small amount s_j^n in the ideal lattice and then calculate the forces F_i^m on all neighboring atoms self-consistently. This represents a typical "impurity problem" which can be handled by the above described full-potential KKR-GF method. For a cubic crystal with one atom per unit cell, only a single self-consistent calculation with a shift, e.g., in (100) direction, is sufficient to determine all non-equivalent Born-von Karman parameters Φ_{ij}^{mn}. The phonon frequencies for all **k**-vectors in the

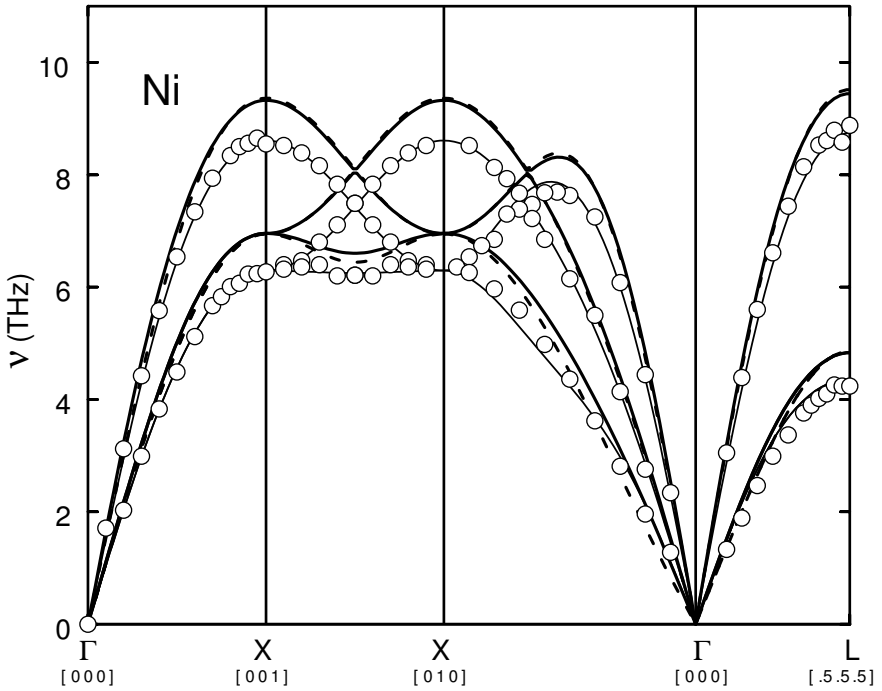

Figure 4: Calculated (solid line) and experimental (open circles) phonon spectra of fcc Ni. The phonon spectra obtained from a non-spin-polarized calculation is also shown (dashed line). The thin solid line is a Born-von Karman fit to the experimental points.

Brillouin zone can then be easily obtained by a Fourier transform. In the calculations below for fcc metals we use a displacement of 0.5 % of the NN distance and calculate the forces on 5 fcc shells of neighboring atoms. The effects of third order anharmonicities are corrected by symmetry considerations.

In Fig. 4 the calculated phonon dispersion curves of fcc nickel are compared with the experimental ones determined by neutron scattering. The overall agreement of the shape of the spectra is very good. The calculated phonon frequencies are slightly higher than the experimental ones, but this is probably due to the LDA used in the electronic structure calculation. The standard trend of the LDA is to make the lattice stiffer by reducing lattice constants by a few percent and by overestimating elastic constants. The calculated *ab initio* lattice constant for fcc Ni (6.52 a.u.) was used in the phonon calculations instead of the experimental value (6.65 a.u.). In Fig. 4 also the result of a non-spin-polarized calculation is shown by the dashed line. It is seen that the magnetism of Ni has not any remarkable effect on the phonon spectra, which one might anticipate since Ni does not undergo any structural phase transition between the low-temperature ferromagnetic and the high-temperature paramagnetic phases nor do the lattice parameter and the elastic constants show any significant anomalies near the Curie temperature. We have also calculated the phonon spectra of fcc Cu for different lattice parameter values and used these to determine the vibrational contribution to the lattice free energy. The lattice constant as a function of temperature was determined from these calculations and the thermal expansion coefficient of Cu was found to be $\alpha_{\rm theor} = 16 \cdot 10^{-6}$ K^{-1}, which is in surprisingly good agreement with the experimental value $\alpha_{\rm exp} = 17 \cdot 10^{-6}$ K^{-1}.

8.6 Summary and Outlook

We have demonstrated in this highlight that the full-potential KKR-Green's function method allows an efficient evaluation of forces and enables the study of lattice relaxations around impurities in metals, especially in transition metals where methods based on plane waves are difficult to apply. The forces are calculated by the ionic Hellmann-Feynman theorem and the Green's functions for shifted atomic positions are obtained by angular momentum transformations. It is also demonstrated how the whole phonon spectrum of a transition metal can be determined from a single self-consistent calculation in real space.

We have applied the full-potential KKR-GF method to predict impurity induced lattice relaxations around a large number of impurities in Cu (3d, 4sp, 4d, and 5sp impurities) and in Al (3d and 4sp impurities). The results for first NN displacements and macroscopic volume changes induced by impurities are in good agreement with the available experimental data from EXAFS and lattice parameter measurements. We have also used the method to calculate the phonon spectrum of ferromagnetic Ni and the thermal expansion coefficient of Cu. Both of these results are found to agree very well with experiments, the latter one even surprisingly well.

In the future we plan to implement force and relaxation calculations into our existing KKR-GF computer programs for surfaces, layered systems, and surface impurities. We believe that this can be done straightforwardly. We also want to point out that the described force and relaxation calculations can be used in connection with our recently developed version of the tight-binding KKR method [26, 27]. We are also developing a new method which allows to calculate bigger relaxations than the present version by reducing the angular momentum cut-off needed in the Dyson equation (14) at shifted positions. These improvements would make the KKR-GF formalism a very powerful tool to study the properties of materials from first principles.

Acknowledgments

This work has been supported by a bilateral German–Greek cooperation grant and was partially funded by the TMR Network "Ab initio calculations of magnetic properties of surfaces, interfaces and multilayers" (Contract: ERBFMRXCT960089). It has also benefited from collaborations within the HCM Network "Ab initio (from electronic structure) calculation of complex processes in materials" (Contract: ERBCHRXCT930369).

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