6 News from UK's CCP9 Programme

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

6.1 Report on the KKR Workshop

KKR Workshop: new developments, applications and collaborations

October 20-22, 2006

Burwalls (University of Bristol, Bristol UK)

Summary

The KKR method is a well established versatile tool for calculating the electronic structure of a wide variety of systems, including periodic solids, disordered alloys, surfaces and nanostructures. In addition it supplies a very efficient and powerful basis for the calculation of spectroscopic properties and the inclusion of many-body effects. The Bristol workshop brought together the leading experts in KKR calculations throughout Europe, together with their students and postdocs and selected invited participants from USA and Japan. Recent advances in the theory of disordered alloys, such as the non-local coherent potential approximation (NLCPA), local self-interaction correction (L-SIC), and first principles many-body dynamical mean field theory (DMFT), were among the topics discussed. Physical problems ranging from magnetism in bulk materials to, electronic structure of surfaces, electronic transport, and ab initio modelling of spectroscopy in pump-probe experiments and scanning tunnelling microscopy.

Scientific content of the workshop

The workshop was opened by H. Ebert, who described the recent development of KKR LDA+U and KKR DMFT methods for first principles many-body theory. He showed that the LDA+U formalism was possible with a relatively straightforward extension of the KKR t-matrices, to include Hubbard U and exchange integrals J on a single site. The KKR DMFT formalism is more complex to implement, involving construction of a suitable single-site self-energy, evaluated within the FLEX approximation. Different schemes of analytic continuation of complex frequencies were discussed. Results were presented for spin and orbital magnetic moments in pure fe Ni Co and for Fe-Ni alloys. Spin dependent photoemission spectra for Ni(001) were also presented.

J. Staunton showed how the disordered local moment model (DLM) can be used to describe the onset of magnetic order and the paramagnetic susceptibility of metals. Using the local Self Interaction Correction (L-SIC) to remove the spurious self energy associated with electrons interacting with themselves that the LSDA includes, she demonstrated how the magnetism of metals with strongly correlated electrons can also be treated using the DLM picture. Prof. Staunton illustrated the approach by describing a study of heavy rare earth systems which was interpreted in terms of an ab-initio RKKY interaction.

A. Ernst presented a the use of multiple scattering theory for spintronics applications. He showed that convergence problems with the KKR method at high angular momenta values, can be corrected by use of the Lloyd formula for the density of states. Corrections to the atomic sphere approximation were compared, including the atomic sphere approximation (ASA), muffin tin approximation (MTA) and full-charge approximation (FCDA). Calculation of exchange integrals and spin-waves were discussed, and results were shown for the magnetization and critical temperature in RPA or mean field theory for Co and Fe. The extension of self-interaction-correction (SIC) to multiple scattering theory was also described. Calculated critical temperatures for transition metal oxides were presented, as obtained with the SIC-KKR method.

I. Abrikosov presented the talk of L. Vitos who was unable to attend the workshop. He described the construction of exact muffin-tin orbitals, and their implementation in a CPA formalism for alloys, EMTO-CPA. Results were presented for the elastic constants of the Hume-Rothery alloys Ag-Zn and for the Fe-Si alloys, occurring in the Earth's core. Finally the problems presented by two center expansions of the slope matrix, in cases with semi-core states, and it was shown that a two centre expansion with extra screening spheres was able to give accurate results for a wide range of energies around the muffin tin zero.

The next session focused on the non-local coherent potential approximation, NLCPA, for alloys. This is a cluster generalisation of the CPA which includes nonlocal correlations and hence a description of short-range order. Dr. Rowlands presented an overview of the idea and formalism of the NLCPA both within a tight-binding framework and the KKR framework. The new fully charge self-consistent (SCF)-KKR-NLCPA was also presented. This enables the total energy to be investigated as a function of short-range order, whilst systematically taking into account the charge-correlation (Madelung) contribution missing in the CPA. Dr. Tulip described the formalism for calculating the spectral function within the KKR-NLCPA. The coarse-grained

spectral function was calculated by averaging the spectral functions for each NLCPA tile in the Brillouin zone. A new formalism was presented in which the Green function is split into cluster diagonal and cluster off diagonal components which allows the calculation of the spectral function at specific k-space points. G. Batt described a new method for obtaining a fully continuous spectral function within the NLCPA formalism. The key idea is to use different sets of boundary conditions and to combine these together to yield a continuous spectral function and Fermi surface, unlike in standard NLCPA where both have unphysical discontinuities for small cluser sizes. Dr. D. Ködderitzsch described the KKR-NLCPA formalism in a fully relativistic implementation. This requires an efficient implementation which uses symmetry in the **k**-space coarse-graining procedure and several means to stabilise the NLCPA-algorithm. Results were presented for an application to the alloy system Fe_xPt_{1-x}.

The following session concerned magnetism. J. Minar described how the fully relativistic spin-polarized KKR method has been used to study the magnetic and spectroscopic properties for supported alloy clusters. For a variety of clusters supported on various transition metal substrates it was shown that the magnetic properties depend on several different parameters such as substrate type, cluster size as well as shape etc. The magnetic coupling constants of these clusters were calculated and used within a Monte Carlo simulations in order to obtain temperature-dependent magnetic properties. S. Mankovsy presented the results of a theoretical study of the high-field magnetic susceptibility of ferromagnetic metals and alloys. The approach presented has been applied for calculations of the high-field susceptibility of some 3d transition metals (Fe, Ni, Co) and alloys (Fe_xCo_{1-x}, Ni_xCu_{1-x}) and all results of the calculations are found to be in good agreement with experiment. In the case of Fe_xCo_{1-x} alloy the high-field susceptibility was found to be strongly dependent on the ordering of the system. S. Lounis described non-collinear magnetism in 3d nanostructures on surfaces, focusing on small magnetic $3d$ clusters on Ni(001), on *fcc* Fe/Cu(001) and on Ni(111). The size and orientation of the local moments and the stabilization of noncollinear magnetic solutions were investigated. While clusters of Fe, Co, Ni atoms are magnetically collinear, noncollinear magnetic coupling is expected for Cr and Mn clusters on surfaces of elemental ferromagnets. P. Mavropoulos described two methods to obtain temperature dependence of magnetization in half metals. The first approach, uses the full-potential linearized augmented plane-wave (FLAPW) method to deduce magnon dispersion relations. Fourier transformation yields the real-space exchange constants J_{ij} of the Heisenberg model from which we find the thermodynamic quantities, such as the magnetization curve $M(T)$, susceptibility and the Curie temperature T_C. The second approach is based on the disordered local moment (DLM) state $NiMn^{\uparrow}_{1-x}Mn^{\downarrow}_{x}Sb$, with $0 \leq x \leq 0.5$ representing the temperature range $0 \leq T \leq T_C$. This approach describes the system at $T > 0$ in a mean-field manner. The DLM calculations are done within the coherent-potential approximation (CPA) and the Korringa-Kohn-Rostoker full-potential Green function method. In the last talk of this section, R. Zeller described the use of the Lloyd formula, which is an important concept in the KKR Green function method since it directly gives the integrated density of states with a considerably better angular-momentum convergence than the the truncated multiple-scattering representation of the Green function. It was shown how the disadvantages of the Lloyd formula can be avoided in numerical applications and that rather accurate results can be obtained. It is also shown how the charge density obtained from the Green function can be made consistent

with the Fermi level obtained from the Lloyd formula. These points were illustrated by calculations for the temperature dependence of the Fermi level in GaN. and the magnetic moments of zincblende CrAs, dilute $Mn_xGa_{1-x}N$ and a FeSi multilayer.

I. Abrikosov presented theoretical models used for the calculation of core-level shifts, including the initial state, transition state, and final state. He discussed core level shifts and Auger energy shifts in the bulk of random alloys, and disorder broadening of core spectral lines. He concluded with comments on implications for the structure of thin film alloys, such as $Ag_{50}Pd_{50}$ alloy deposited at Ru (0001).

E. Bruno described the ab initio theory and simulation of metallic alloys within the Charge Excesses Functional Theory - Monte Carlo Method (CEF-MC). He showed that CEF theory constitutes a simple, very realistic model for the energetics of metallic alloys. It is a coarse grained density functional in which the full density is replaced by effective charges q_i. The resulting CEF defines an configurational Hamiltonian that includes an infinite series of many-body interactions, for which no truncation for the range of interactions is necessary. A Monte Carlo-CEF algorithm allows for the study of order-disorder phase transitions. Order-disorder phase transitions have been reviewed as charge-monodisperse to charge-polydisperse transitions

The session on self-interaction corrected DFT (SIC) featured talks by W. Temmerman, Z.Szotek, M. Lüders and M. Toyoda. W. Temmerman showed that SIC-LSD leads to an useful definition of valency in strongly interacting systems, important examples of which include systems such as Ce, $LiNiO_2$, Fe in MgO, the rare earths, the actinides and Mn in GaAs and ZnO. Z. Szotek discussed energy gaps calculated using self-interaction corrected local spin density (LSD) approximation. Results were presented for rare gas solids, transition metal oxides, parent compounds of high temperature superconductors, and Europium chalcogenides. The SIC-LSD energy gaps were compared to such approaches as LSD, GGA, LDA+U and hybrid functionals. M. Lüders described the two ways in which SIC can be implemented in the KKR method, namely local SIC and Pseudo-SIC. In particular the differences and similarities in the technical implementation, as well as in the philosophy of the applications were described. M. Toyoda investigated the electronic structure and magnetic properties of dilute magnetic semiconductors from first principles calculations in the self-interaction-corrected local density approximation (LDA+SIC). The implementation is based on the pseudopotential-like self-interaction correction scheme (pseudo-SIC) implemented with the Korringa-Kohn-Rostocker coherent potential approximation (KKR-CPA) method. It was found that the electronic structures calculated by LDA+SIC are in better agreement with the photoemission spectrum than those calculated by LDA. Magnetic properties and Curie temperature calculated by LDA+SIC also differ from those calculated by LDA. For example, showing enhancement of ferromagnetic interactions between the Mn ions in GaMnAs.

The Sunday morning session focussed on surfaces and nanostructures. I. Mertig started the session with the theory of tunnel magnetoresistance in Fe/MgO/Fe multiyayers with amorphous Fe electrodes. She described the structure models and electronic structure of amorphous Fe, and then TMR tunnelling with such amorphous electrodes, compared to free electron leads. It was found that a crystalline Fe layer is important to act as a spin filter and so giant TMR arises from by the matching of crystalline Fe to the MgO barrier. W. Hofer described calculations of high resolution tunnelling microscopy within a multiple scattering formalism. Multiple electron

scattering in a vacuum barrier are obtained using real-space single-electron wavefunctions for the separate surfaces. The Green functions in the vacuum barriers are calculated to first order, showing that the zero-order current is equal to the usual Bardeen approach only in the limit of zero bias and derive the modifications in the finite bias regime. A first-principles formulation for the energy of interaction between the two surfaces, is shown to be proportional to the tunnelling current and hence the tunnelling current can in principle be computed to any order in the Dyson expansion. L. Szunyogh discussed the size dependent Kondo effect under the assumption that a magnetic impurity near a surface experiences spin-orbit induced anisotropy. The splitting of the states of a Kondo impurity interacting with the neighboring host atoms is calculated via perturbation technique up to second order in the exchange coupling strength. Two models were investigated, namely, an impurity subject to a host-induced magnetic anisotropy and an impurity with a strong local spin-orbit coupling. It is found that in both cases the anisotropy energy is an oscillating function of the distance d from the surface. The numerical estimates suggests that second mechanism gives rise to an anisotropy energy of the desired order of magnitude whereas the first mechanism seems to be of negligible importance. S. Shallcross described the calculation of exchange integrals using an expansion based upon the Lloyd formula. In particular the RKKY interaction between moments was derived in an asymptotic expansion from this formula.

In the final session of the workshop, A. Vernes described calculation of pump-probe spectroscopy within KKR. The Kubo response theory was extended by linearizing the density of both pump- and probe-excited states and neglecting the overlap between the femtosecond pulses. The time-dependent zeroth order dynamic conductivity as obtained by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker method for fcc Ni(100) predicts a relatively fast demagnetization process over 100 fs after the impact of the probe pulse, which is in a reasonably good agreement with available experimental data. Calculations for Co/Pt(111) and Pt/Co/Pt(111) systems show that the layer-resolved demagnetization time is not necessarily dominated by the demagnetization time of the ferromagnetic Co layer. B. Lazarovitis described calculations of artificial atomic structures, notably quantum corrals. In particular the density of states was obtained for magnetic adatoms, leading to spin-dependent properties, and surface states which can be compared with the STM dI/dV experiments. M. Stocks described the LSMS multiple scattering code, and its applications to various nanostructures as carried out at Oak Rigde National Laboratory USA. Finally, L. Udvardi described the calculation of relativistic exchange interactions. A relativistic extension of the magnetic torque theorem was combined with the Lloyd formula for the band energy to obtain closed formulas for various derivatives of the free energy. The resulting magnetic anisotropy energies were presented for thin Fe layers on a range of non-magnetic transition metal surfaces. The Dzyaloshinsky-Moria interaction was shown to dominate and to play the key role in domain formation.

Assessment of results and impact

The KKR method is a versatile computational tool for a wide range of electronic structure problems. The workshop has highlighted this versatility in terms of the range of applications, such as nanostructures, spectroscopy, surface physics, and spintronics. The KKR method is

also evolving and expanding its applicability with a number of exciting new developments, such as the improved cluster methods for alloys (NLCPA and CEF-QMC), and with novel many-body methods for strongly interacting systems (eg L-SIC and DMFT). The workshop provided a timely opportunity for all the main practitioners of the KKR method in Europe to meet and exchange ideas on these new devlopments. The workshop was attended by a number of PhD students, postdocs and young researchers, who will continue these developments in the future.

Final Program of Meeting

October 20

Arrival, Registration and Welcoming reception

October 21

8:30-9:00	H.Ebert	*Combination of the LDA+U and DMFT methods with KKR*
9:00-9:30	J.B.Staunton	*DLM and the LSIC: a description of magnetism in the heavy rare earths*
9:30-10:00	A.Ernst	*GW with KKR*
10:00-10:30	I. Abrikosov	*The Exact Muffin-Tin Orbitals-CPA method and applications*
10:30-11:00	Coffee	
11:00-11:30	D.A. Rowlands	*KKR nonlocal coherent-potential approximation*
11:30-11:50	P.R. Tulip	*First-principles spectral function in the KKR-NLCPA*
11:50-12:10	G.Batt	*Fermi Surfaces of Random Alloys*
12:10-12:30	D.Ködderitzsch	*An efficient and fully relativistic implementation of the KKR-NLCPA making use of symmetry*
12:30-1:30	Lunch	
1:30-1:50	J. Minar	*Magnetic properties of deposited transition metal clusters*
1:50-2:10	S.Mankovskyy	*Spin and orbital high-field susceptibility of ferromagnetic alloys calculated using the SPR-KKR*
2:10-2:30	S.Lounis	*Non-Collinear KKR Green function method: application to 3D nanostructures on surfaces*
2:30-2:50	P.Mavropolous	*Temperature dependence of spin polarization of half-metallic ferromagnets: an approach from first principles*
2:50-3:10	break	

3:00-3:20	R.Zeller	*The Lloyd Formula*
3:20-3:40	I.Abrikosov	*Core Level Spectroscopy in Alloys*
3:40-4:00	E.Bruno	*First-principles calculations of phase diagrams in metallic alloys through the CEF-QMC method*
4:00-4:30	Tea	
4:30-5:00	W.E.Temmerman	*Valency and SIC*
5:00-5:30	Z.Szotek	*Gaps and SIC*
5:30-6:00	M.Lüders	*Local SIC*
6:00-6:30	M. Toyoda	*Electronic Structure of Dilute Magnetic Semiconductors from LDA+SIC calculations*

October 22

9:00-9:30	I. Mertig	*Tunnelling magnetoresistance with amorphous electrodes for Fe/MgO/Fe*
9:30-10:00	W. Hofer	*Simulation of high-resolution tunnelling microscopy and spectroscopy*
10:00-10:30	L. Szunyogh	*Kondo Impurity near a Surface*
10:30-10:50	S. Shallcross	*Asypmtotics of exchange integrals*
10:50-11:20	Coffee	
11:20-11:40	A.Vernes	*Ab initio description of pump-probe spectroscopy using KKR*
11:40-12:00	B. Lazarovits	*Quantum Corrals*
12:00-12:30	G.M.Stocks	*LSMS*
12:30-1:00	L.Udvardi	*Relativistic Exchange Interactions*

Posters

H. Akai and M. Ogura	*Magnetic properties of* $La_{1-x}Ca_xMnO_3$
M. Ogura	*Magnetic properties of 3d pyrite-type mixed crystals calculated by the full potential KKR-CPA method*
S. Gallego	*Magnetic anisotropy of free and capped Co/Ru(0001): strain and stacking*
P. Mavropoulos	*Complex magnetic order and transition to half metallicity in Mn-doped* Fe_3Si

Abstracts

The DLM and the LSIC: a description of magnetism in strongly correlated electron systems.

I.Hughes[1], M.Däne[2], A.Ernst[3], W. Hergert[2], M.Lüders[4], J.Poulter[5], J.B.Staunton[1], Z. Szotek[4], W.M.Temmerman[4]

[1] Department of Physics, University of Warwick, U.K.
[2] Martin-Luther-Universität, Halle, Germany.
[3] Max Planck Institut für Mikrostrukturphysik, Halle, Germany.
[4]Daresbury Laboratory, U.K.
[5]Mahidol University, Thailand

We investigate the onset of magnetic order in materials using a 'first-principles' theory of finite temperature metallic magnetism, in which the effects of strong electron correlations are included. Thermally induced spin fluctuations are treated within a mean-field 'disordered local moment' (DLM) picture [1] and the Self-Interaction Correction (SIC) is used to remove the spurious energy associated with electrons interacting with themselves that is inherent in the LSDA [2]. The scheme is implemented using the Korringa-Kohn-Rostoker (KKR) multiple scattering method. We describe our first applications to rare earth systems and interpret our results in the language of a first-principles RKKY interaction.

1. B.L. Gyorffy et al., J. Phys. F 15, 1337 (1985); J.B. Staunton et al., J. Phys. F 15, 1387 (1985).
2. M. Lüders et al., Phys. Rev. B 71, 205109 (2005).

Multiple-scattering theory for spintronics

Arthur Ernst
Max-Planck-Institut für Mikrostrukturphysik, Halle

Multiple-scattering theory (MTS) is a very efficient technique for calculating the electronic properties of an assembly of atoms. Combined with the coherent potential approximation this method can be used as well for studying the electronic structure of alloys and pseudo-alloys. The MTS provides the explicit Green function, which can be used in many applications such as spectroscopy, transport and many-body physics. The main application field of the multiple-scattering theory are mostly metallic systems because semiconductors and insulators can not be correctly described in frame of the local density approximation (LDA). However, even within the LDA the most of existing implementations of the MST can not reproduce results obtained by conventional basis methods. The main reason are the construction and some convergence

properties of the Green function. Here I shall present an approach based on the LDA, which provides an absolutely converged Green function and can be used for half-metals, semiconductors and insulators.

The Exact Muffin-Tin Orbitals-CPA Method and Applications

Levente Vitos[1], Igor Abrikosov[2]

[1] *Applied Materials Physics, Department of Materials Science and Engineering, Royal Institute of Technology Stockholm*

[2] *Department of Physics, Chemistry, and Biology, University of Linkping, Sweden*

The implementation of the Exact Muffin-Tin Orbitals (EMTO) theory [1] within the Spherical Cell Approximation [2] is reviewed. We present an investigation of the energy dependence of the EMTO slope matrix in the complex energy plane [3]. We show that a two-center Taylor type expansion of the slope matrix accurately reproduces the exact values within a complex energy range covering the usual energy window used in electronic structure calculations.

Combined with the Coherent Potential Approximation (CPA), the EMTO method [4,5] is found to be suitable for an accurate determination of the electronic structure and total energy of completely random alloys with a substitutional disorder on any kinds of underlying crystal structure. The accuracy of the EMTO-CPA method is demonstrated in the case of cubic and hexagonal Hume-Rothery alloys (CuZn and AgZn) [4,6]. As applications, our recent results obtained for Fe-Si [7] and Fe-Mg [8] are discussed.

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[3] A. E. Kissavos, L. Vitos, and I. A. Abrikosov, to be published.
[4] L. Vitos, I. A. Abrikosov, and B. Johansson, Phys. Rev. Lett. 87, 156401 (2001).
[5] L. Vitos, I. A. Abrikosov, and B. Johansson, in Complex Inorganic Solids, Structural, Stability, And Magnetic Properties Of Alloys, eds. A. Meike, P. E. A. Turchi, A. Gonis, K. Rajan, pg. 339 (14), Springer Verlag (2005).
[6] B. Magyari-Kope, L. Vitos, and G. Grimvall, Phys. Rev. B, 70, 052102(4) (2004).
[7] L. Dubrovinsky, et al., Nature, 422, 58-61 (2003).
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Korringa-Kohn-Rostoker nonlocal coherent potential approximation

D. A. Rowlands

University of Bristol, Dept. of Physics, Brisotl, BS8 1TL, UK

The nonlocal coherent-potential approximation (NLCPA) [1] has emerged as a viable method for generalising the CPA by systematically including nonlocal fluctuations about the single-site

mean field. In this talk the idea and formalism of the NLCPA is first introduced within the tight-binding framework, and then the KKR-NLCPA [2] formalism described by analogy. Results are presented for the CuNi system, illustrating the effects of nonlocal scattering correlations and short-range order. The combination of the KKR-NLCPA with density functional theory is then described, which results in a fully charge self-consistent (SCF)-KKR-NLCPA method [3] which systematically takes into account the charge-correlation (Madelung) contribution to the total energy missing in the conventional KKR-CPA. An application to the CuZn system is presented, demonstrating how the total energy varies as a function of short-range order.

[1] M. Jarrell and H. R. Krishnamurthy, PRB 63, 125102 (2001)
[2] D. A. Rowlands, J. B. Staunton, B. L. Gyorffy, PRB 67, 115109 (2003)
[3] D. A. Rowlands, A. Ernst, B. L. Gyorffy, J. B. Staunton, PRB 73, 165122 (2006)

Fermi Surfaces of Random alloys

G. M. Batt
University of Bristol, Dept. of Physics, Brisotl, BS8 1TL, UK

The effects of short range ordering (SRO) on the Fermi Surface for random metallic alloys was discussed. Specifically how well the nonlocal coherent-potential approximation (NLCPA) [1], predicts deviations from the Fermi Surfaces predicted by the single-site mean field theory of the CPA. It was shown that when the difference is large between the two, one sees in the NLCPA the introduction of discontinuities in **k**-space across coarse grained tile boundaries. The consequence is that the resulting Fermi Surface becomes unphysical leading to large fluctuations in the density of states (DOS). A solution [2] to overcome this problem was introduced that uses all possible boundary conditions when transforming between lattice and cluster quantities in the NLCPA. Where all discontinuities are removed, resulting in an improved DOS over the standard implementation of the NLCPA. The procedure was demonstrated for the 2D tight-binding analogue of CuPd. Giving dramatic **k**-dependent corrections to both the topology and lifetime of the Fermi Surface over the CPA. Also preliminary results for the KKR-NLCPA [3] prediction for the Fermi Surface of CuPd were given. Where a small change was observed over the KKR-CPA. It is expected that a combination of the new method [2] with [3] will improve further on this prediction.

[1] M. Jarrell and H. R. Krishnamurthy, PRB 63, 125102 (2001)
[2] G. M. Batt and D. A. Rowlands J. Phys.: Condens. Matter 18 (2006) 11031-11046
[3] P. R. Tulip, J. B. Staunton,D. A. Rowlands, B. L. Gyrffy, E. Bruno, and B. Ginatempo,Phys. Rev. B 73, 205109 (2006)

An efficent and fully relativistic implementation of the KKR-NLCPA

D. Ködderitzsch
Ludwig-Maximilians-Universität München, Department Chemie und Biochemie, Physikalische Chemie

The KKR-method allows to elegantly make use of the coherent-potential approximation (CPA). As a single-site mean field theory the CPA is not able to describe short-range order (e.g. clustering) effects. Recently the so called non-local CPA (NLCPA) has been devised which remedies these shortcomings. We present our fully relativistic extensions of this formalism. Also we devise an efficient implementation which uses symmetry in the **k**-space coarse graining procedure and several means to stabilise the NLCPA-algorithm. Finally we present an application to the alloy system Fe_xPt_{1-x}.

Spin and orbital high field susceptibility of ferromagnetic alloys calculated using the SPRKKR

S. Mankovsky and H. Ebert

Chemie und Biochemie, Universität München, Butenandtstr. 5-13, D-81377 München, Germany

The results of a theoretical study of the high-field magnetic susceptibility of ferromagnetic metals and alloys are presented. The theoretical description of the magnetic susceptibility is based on a combination of a linear response approach and the fully relativistic Green's function formalism. This approach accounts not only for the Pauli spin and Van Vleck orbital susceptibilities, but also for the spin-orbit induced susceptibility as well as the Landau susceptibility. Calculation of the Landau susceptibility shows that it can give an substantial contribution to the total susceptibility. It can be positive in the case of transition metals and alloys and should be taken into account to get a proper value of magnetic susceptibility.

In contrast to the paramagnetic state, the spin-charge response function as well as contributions to the susceptibility related to a Fermi level shift become important in ferromagnetic materials. Therefore, these effects have been also taken into account in the present calculations of the magnetic susceptibility.

The approach presented has been applied for calculations of the high-field susceptibility of some 3d transition metals (Fe, Ni, Co) and alloys (Fe_xCo_{1-x}, Ni_xCu_{1-x}). All results of the calculations are found in good agreement with experiment. It is shown that the orbital Van Vleck and Landau contributions are dominating terms in the magnetic susceptibility of the pure Fe, Ni and Co ferromagnetic metals.

In the case of Fe_xCo_{1-x} alloy the high-field susceptibility was found to be strongly dependent on the ordering of the system. In the Fe reach limit the results of our calculations obtained for disordered alloy are in a very good agreement with experiment, while in the middle region of concentrations the experimental results are in a better agreement with those calculated for partially ordered alloys.

Non-collinear KKR Green function method: Application to 3d nanostructures on surfaces

Samir Lounis (s.lounis@fz-juelich.de), Phivos Mavropoulos, Rudolf Zeller, Peter H. Dederichs, Stefan Blügel

Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany

Rather few *ab-initio* calculations for small magnetic nanostructures exists, especially when noncollinear magnetic configurations have to be considered. Motivated by this, we developed and extended the Jülich version of the full-potential scalar–relativistic Korringa-Kohn-Rostoker (KKR) Green–function code to treat noncollinear magnetic nanostructures on surfaces. We focus on small magnetic $3d$ clusters on Ni(001), on *fcc* Fe/Cu(001) and on Ni(111). The size and orientation of the local moments and moreover the stabilization of noncollinear magnetic solutions are investigated. While clusters of Fe, Co, Ni atoms are magnetically collinear, noncollinear magnetic coupling is expected for Cr and Mn clusters on surfaces of elemental ferromagnets.

The origin of the noncollinear states is frustration, *i.e.* the competition of the antiferromagnetic exchange coupling among the Cr or Mn atoms with the antiferromagnetic (for Cr) or ferromagnetic (for Mn) exchange coupling between the impurities and the substrate.

For the Ni(001) surface, we find that Cr and Mn first-neighbouring dimers show noncollinear behavior nearly degenerate with the most stable collinear configuration. Increasing the distance between the dimer atoms leads to a similar behavior as the single impurities. In the case of fcc Fe surface, the exchange interaction with the impurities is stronger leading then to the formation of new complex magnetic configurations. The Cr-trimer, tetramer and pentamer all prefer noncollinear magnetic coupling. For the Ni(111) surface, we examine the Cr- and Mn-trimer, which shows the well-known triangular frustration.

Finally, we compare some of the ab-initio results to the results for a classical Heisenberg model, where the exchange constants were fitted to total energy results of the collinear state; the agreement is sometimes surprisingly good, in other cases it fails, however.

Temperature dependence of spin polarization in half-metallic ferromagnets: an approach from first principles

Ph. Mavropoulos, M. Ležaić, and S. Blügel

Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany

Half-metallic ferromagnets (HMF) are ferromagnetic metallic compounds showing, under ideal circumstances, e.g. at zero temperature and no structural disorder, a spin-polarization P of 100% at the Fermi level. This makes them, at least hypothetically, ideal candidates for spintronics applications. However, it is also clear that the ideal half-metallic property ($P = 100\%$) may not be realized at elevated temperatures. Combining first-principles calculations with a Monte Carlo method, we investigate the effect of temperature on the band gap of HMF. In particular, we examine HMF which contain more than one magnetic atom in the unit cell, and describe the mechanisms which lead to the drop of polarization at elevated temperatures.

We follow two approaches to the excited magnetic states. The first approach is based on the full-potential linearized augmented plane-wave (FLAPW) method[1]. Using the force theorem we deduce magnon dispersion relations. A subsequent Fourier transformation yields the real-space exchange constants J_{ij} of the Heisenberg model. We use a Monte Carlo method to find

the thermodynamic quantities, such as the magnetization curve $M(T)$, susceptibility and the Curie temperature T_C. The second approach is based on the disordered local moment (DLM) state $NiMn^{\uparrow}_{1-x}Mn^{\downarrow}_{x}Sb$, with $0 \leq x \leq 0.5$ representing the temperature range $0 \leq T \leq T_C$. This approach describes the system at $T > 0$ in a mean-field manner. The DLM calculations are done within the coherent-potential approximation (CPA) and the Korringa-Kohn-Rostoker full-potential Green function method[2].

For multicomponent HMF we cannot confirm the proposed model which assumes that $P(T) \sim M(T)$ [3]. The presence of different magnetic components can, in principle, lead to a drop of the spin-polarization at $T > 0$ which is much faster than the drop of magnetization in these systems. This effect can be quite drastic and depends largely on the strength of the hybridization among the components [4]. We describe the effects of temperature in more detail for the compounds NiMnSb and Co_2MnSi.

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The Lloyd formula

Rudolf Zeller
Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich

The Lloyd formula is an important concept in the KKR Green function method since it directly gives the integrated density of states with a considerably better angular-momentum convergence than the one obtained by the use of the truncated multiple-scattering representation of the Green function. A disadvantage of the Lloyd formula is its dependence on multivalued imaginary parts of logarithms of complex arguments. It is shown how this difficulty can be avoided in numerical applications and that rather accurate results can be obtained as illustrated by calculations for the temperature dependence of the Fermi level in GaN. It is also shown how the charge density obtained from the Green function can be made consistent with the Fermi level obtained from the Lloyd formula. The consistency, which is achieved by renormalizing the Green function at each point of the complex energy charge-density integration contour, is important, e. g. in spintronic materials, where it leads to integer moments in ferromagnetic half metals and to properly aligned energetic states in metal-semiconductor systems as illustrated by density-functional calculations for zincblende CrAs, dilute $Mn_xGa_{1-x}N$ and a FeSi multilayer.

Core-Level Spectroscopy in Alloys

I.A. Abrikosov

Department of Physics, Chemistry and Biology, Linkping University, SE-581 83 Linkping, Sweden

We show that core-level binding energy shifts (CLS) can be reliably calculated within Density-functional theory [1]. The scheme includes both the initial (electron energy eigenvalue) as well as final state (relaxation due to core-hole screening) effects in the same framework. The results include CLS as a function of composition in substitutional random bulk [2,3] and surface alloys [4]. Sensitivity of the CLS to the local chemical environment in the bulk and at the surface is demonstrated [5]. A possibility to use the CLS for structural determination is discussed. Finally, an extension of the model is made for Auger kinetic energy shift calculations [6].

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Valency and SIC

W.M. Temmerman
Daresbury Laboratory, Daresbury, Warrington, WA4 4AD, Cheshire, U. K.

In this talk I will show how the application of the self-interaction correction to the local spin density (SIC-LSD) leads to an useful definition of valency. This is then explicity demonstrated on the basis of several examples which include systems such as Ce-the prototypical system showing the localization/delocalization transition-, $LiNiO_2$-transition from trivalent Jahn-Teller (JT) active Ni to divalent JT inactive Ni-, Fe in MgO-transition from high spin to low spin-, the rare earths-the discovery of two types of f-electrons-, the actinides-ls versus jj coupling- and Mn in GaAs and ZnO- applications in the field of spintronics.

Gaps and SIC

Z. Szotek
Daresbury Laboratory, Daresbury, Warrington, WA4 4AD, Cheshire, U. K.

We provide critical assessment and deeper understanding of energy gaps calculated using self-interaction corrected local spin density (LSD) approximation. We discuss the underlying formulations of the SIC-LSD method and how they impact the formal definition and interpretation of the energy gap. We present results for rare gas solids, transition metal oxides, parent compounds of high temperature superconductors, and Europium chalcogenides. Wherever possible we compare the SIC-LSD energy gaps to such approaches as LSD, GGA, LDA+U and hybrid functionals.

Single-Site Self-Interaction Correction in KKR-CPA

Martin Lueders, Daresbury Laboratory, Daresbury, Warrington, WA4 4AD, Cheshire, U. K.

Strongly correlated materials including d- and f- electrons are of common interest in many areas. For this class of systems, the standard local spin density approximation (LSD) to density functional theory often fails, which is due to a spurious self-interaction of the localized electrons. It has long been shown that this can be corrected by the so-called self-interaction corrected (SIC) LSD. The SIC-LSD is so far implemented in two ways in the KKR method.

In my talk I will overview the background of SIC in general, and then discuss in detail the approximations underlying the two current implementations, namely local SIC and Pseudo-SIC. In particular the differences and similarities in the technical implementation, as well as in the philosophy of the applications will be investigated.

Electronic Structures of Dilute Magnetic Semiconductors from LDA+SIC calculations

M. Toyoda[1], H. Akai[2], K. Sato[1] and H. Katayama-Yoshida[1]

[1] *Department of Computational Nanomaterials Designm The Institute of Scientific and Industrial Research, Osaka University, Ikaraki 567-0047, Japan.*

[2] *Department of Physics, Osaka University, Toyonaka 560-0043, Japan.*

We investigate the electronic structure and magnetic properties of dilute magnetic semiconductors from first principles calculations. Since the 3d electrons of transition-metal impurities are strongly localized[1], the local density approximation (LDA) gives a poor description of them. Thus we use the self-interaction-corrected local density approximation (LDA+SIC). Our implementation is based on the pseudopotential-like self-interaction correction scheme (pseudo-SIC) proposed by Filipetti and Spaldin[2]. We implement the pseudo-SIC with the Korringa-Kohn-Rostocker coherent potential approximation (KKR-CPA) method[3]. In order to investigate the magnetic properties, we use a classical Heisenberg-type Hamiltonian for the DMS system. The parameters of the Hamiltonian, the magnetic exchange interaction J_{ij}, are calculated from the first-principles results using Lichtenstein's formula[4]. We found that the electronic structures calculated by LDA+SIC are in better agreement with the photoemission spectrum than those calculated by LDA. We will also discuss about the difference of magnetic properties and Curie

temperature calculated by LDA+SIC from those calculated by LDA. For example, we found the enhancement of ferromagnetic interactions between the Mn ions in GaMnAs. This is because the antiferromagnetic super-exchange interaction is suppressed due to the increase of the exchange splitting. A contrary example is ZnCoO in which we found antiferromagnetic interaction between Co ions due to a large gap opening between the occupied and unoccupied d states.

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Magnetic anisotropy of an impurity in the vicinity of a surface

L. Szunyogh
Department of Theoretical Physics, Budapest University of Technology and Economics, Budapest, Hungary
and
Center for Computational Materials Science, Vienna, Austria

The most successful explanation of the size dependent Kondo effect relies on the assumption that a magnetic impurity near a surface experiences spin-orbit induced anisotropy. I present numerical investigations in terms of a simple tight–binding model that accounts correctly for the semi–infinite geometry of the host metal, while the splitting of the states of a Kondo impurity interacting with the neighboring host atoms is calculated via perturbation technique up to second order in the exchange coupling strength. Two models are investigated, namely, *(i)* an impurity with a half-filled p-shell subject to a host-induced magnetic anisotropy and *(ii)* an impurity of D^1 configuration with a strong local spin-orbit coupling. It is found that in both cases the anisotropy energy is an oscillating function of the distance d from the surface with an amplitude that, however, decays as $1/d^2$ in case *(i)*, while as $1/d^3$ in case *(ii)*. This observation is clearly supported by an asymptotic analysis of the spectral function of the host. Furthermore, numerical estimates suggests that mechanism *(ii)* gives rise to an anisotropy energy of the desired order of magnitude whereas mechanism *(i)* seems to be of negligible importance [1]. Corresponding attempts towards an ab-initio description of the phenomenon based on the Screened KKR method are also discussed.

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Ab initio description of pump-probe experiments using KKR

A. Vernes and P. Weinberger
Center for Computational Materials Science, Technical University Vienna, Gumpendorferstr. 1a, A–1060 Vienna, Austria

The Kubo response theory is extended to describe pump-probe experiments by linearizing the density of both pump- and probe-excited states and neglecting the overlap between the femtosecond pulses. Although the derived formalism includes also second order responses, as illustrated - for example - in the case of double exponential envelopes, the scheme is formally linear and all deduced formulas, e.g. in particular those of the optical conductivities, are straightforwardly implementable within any standard bandstructure method. In particular, even the time-dependent zeroth order dynamic conductivity as obtained by means of the spin-polarized relativistic screened Korringa-Kohn-Rostoker method for fcc Ni(100) predicts a relatively fast demagnetization process over 100 fs after the impact of the probe pulse, which is in a reasonably good agreement with available experimental data. Furthermore, calculations for Co/Pt(111) and Pt/Co/Pt(111) layered systems show that due to the high polarizability of Pt layers, the layer-resolved demagnetization time is not necessarily dominated by the demagnetization time of the ferromagnetic Co layer.

Magnetic properties of 3d pyrite-type mixed crystals calculated by the full potential KKR-CPA method

Masako Ogura,

Department of Physics, Graduate School of Science, Osaka University 1-1 Machikaneyama, Toyonaka, Osaka 560-0043, Japan

3d pyrite-type crystals AB2 (A=Fe, Co, Ni; B=S, Se) show various magnetism depending on the combination and concentration of elements. Due to their anisotropic structure, this system can not be treated in the conventional KKR method in which the muffin-tin potential approximation is used. In the present study, we investigate the electronic structure of the 3d pyrite compounds using the full potential KKR method combined with the CPA. The full potential treatment improves the description of this system and the results reproduce the experiments well.