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## LETTER TO THE EDITOR

# Uniaxial and planar magnetic anisotropy of thin transition-metal films

R Lorenz and J Hafner

Institut für Theoretische Physik, Technische Universität Wien, Wiedner Hauptstrasse 8-10/136, A-1040 Wien, Austria

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**Abstract.** We present a novel approach to the calculation of magnetic anisotropies in crystals and in thin films. Our technique is based on self-consistent real-space recursion calculations on a tight-binding linear-muffin-tin-orbital (TB LMTO)–Hubbard Hamiltonian including spin–orbit coupling and allowing for arbitrary orientations of the local spin-quantization axes. It allows scanning of the magnetic energy continuously as a function of the orientation of the magnetic moment, and thus avoids the computational problems that plague other techniques. Applications are presented for bulk body-centred-cubic iron and for face-centred-cubic Fe monolayers on Cu(100) and Cu(111) substrates. We predict a perpendicular direction of the magnetic moment for Fe/Cu(100) and in-plane orientation of the spins for Fe/Cu(111), with anisotropy energies of the order of 1 to 2 meV atom<sup>-1</sup>. The technique is accurate enough for studying the weak in-plane anisotropies in Fe/Cu(111) that are of the order of 1  $\mu$ eV atom<sup>-1</sup>.

The unique magnetic properties of thin transition-metal films are currently at the centre of intense research interest. The main questions concern (a) the change of the magnetic moments relative to the bulk crystals and (b) the magnetic anisotropy of the layers, in particular the conditions for the preparation of films with a perpendicular orientation of the magnetic moment [1, 2]. Recently, this effect has received much attention due to potential applications in magnetic perpendicular recording. Van Vleck has proposed that the magnetocrystalline anisotropy originates from the spin–orbit coupling, so it is a relativistic effect [3]. In cubic crystals, due to the symmetry of the energy surfaces, the first contribution to the magnetic anisotropy energy (MAE) is of fourth order. Due to the broken crystalline symmetry at the surface, lower orders in perturbation theory can contribute to the MAE as first pointed out by Néel [4].

Two different approaches to the calculation of the MAE have been developed. The first class of methods relies on *ab initio* spin-polarized total-energy calculations, including spin–orbit coupling either self-consistently within the scalar relativistic approximation [5], or as a final perturbation to a calculation neglecting spin–orbit coupling [6–9]. The disadvantage of this type of calculation is the extremely slow convergence of the Brillouin-zone sums. Wang *et al* have proposed a ‘state-tracking procedure’, using information on the change of the bandstructure with increasing spin–orbit interaction to extrapolate the Brillouin-zone integrals [9]. The second approach uses perturbation theory within a tight-binding framework, justified by the argument that the MAE is small compared with characteristic energies of the system (e.g. the bandwidth) [10–13]. The disadvantage of this technique is that the published calculations rely on parametrized non-self-consistent Hamiltonians—this allows the analysis of trends, but makes quantitative predictions for

selected systems difficult. A disadvantage common to both approaches is that they are restricted to collinear spin structures. There is experimental evidence that in certain systems (e.g. FCC Fe/Cu(100)) parallel and perpendicular components of the magnetization coexist for certain film thicknesses [14, 15]. Theoretical arguments [16] support the idea that this arises from the competition between ferro- and antiferromagnetic exchange interactions.

In this paper we present a novel approach to the problem of magnetic anisotropy based on a real-space tight-binding (TB)–Hubbard formulation of the scalar-relativistic Hamiltonian including spin–orbit coupling and magnetic dipolar interactions, allowing for arbitrary directions of the local spin-quantization axes. In a first step, we calculate self-consistently the magnitude and directions of the local magnetic moments—this also determines the easy axis of magnetization. In the second step, we can either apply a magnetic field perpendicular to the easy axis and calculate the MAE from the induced tilt of the moments as a function of the applied field, or orient the moment perpendicular to the easy axis and follow its relaxation into the preferred direction under the influence of the magnetic torque forces—this yields directly the MAE as a function of the angle relative to the easy axis. The new technique is tested by evaluating the MAE for bulk BCC Fe and the uniaxial and planar anisotropies of Fe monolayers on Cu(100) and Cu(111) substrates.

Our approach is based on the following model Hamiltonian

$$H = H_{\text{band}} + H_{\text{exch}} + H_{\text{so}} + H_{\text{dip}} \quad (1)$$

where  $H_{\text{band}}$  describes the non-magnetic part of the bandstructure,  $H_{\text{exch}}$  the magnetic exchange splitting,  $H_{\text{so}}$  the spin–orbit coupling, and  $H_{\text{dip}}$  the dipolar interaction between the magnetic moments. The two-centre tight-binding Hamiltonian  $H_{\text{band}}$  and the exchange-interaction  $H_{\text{ex}}$  are derived from a self-consistent scalar-relativistic spin-polarized calculation of the bandstructure using the linear-muffin-tin-orbital (LMTO) method [17]. A canonical transformation of the paramagnetic part yields the TB LMTO Hamiltonian  $H_{\text{band}}$  in the most localized basis [18]. The formulation of the exchange part is based on the assumption that the local exchange splitting  $\Delta_{il}$  is proportional to the local spin-polarization [19],

$$\Delta_{il} = I_l \mu_{il} \quad (2)$$

with an effective Stoner parameter  $I_l$  for the band with angular quantum number  $l$ , leading to a Hubbard-type exchange-Hamiltonian

$$H_{\text{exch}} = -\frac{1}{2} \sum_{ilm} \Delta_{il} \sum_{ss'} X_{\zeta_i, ss'} c_{ilm}^\dagger c_{ilms} \quad (3)$$

where

$$X_{\zeta_i} = D_{\zeta_i} \sigma_z D_{\zeta_i}^\dagger \quad (4)$$

is the local Pauli spin-matrix  $\sigma_z$  referring to the local quantization axis  $\zeta_i$ , rotated to a global spin axis (the  $D_{\zeta_i}$  are the rotation matrices at the site  $i$ ,  $c_{ilm}^\dagger$  and  $c_{ilms}$  the creation and annihilation operators for electrons,  $m$  and  $s$  stand for magnetic and spin quantum numbers). The *ansatz* (3) for the exchange part of the Hamiltonian is based on the observation that the proportionality (2) holds exactly, with a universal value  $I_2 = (0.95 \pm 0.015)$  eV for all 3d and 4d metals ( $l = 2$ ), if the local exchange splitting  $\Delta_{il}$  is defined in terms of the difference in the position of the centre of gravity of the spin-up and spin-down bands [20, 21] (for a general discussion of the mapping of the LSDA exchange-correlation potential on Hubbard-(or Stoner-)type models, see e.g. Anisimov *et al* [22]; here we only note that in our case the effective Stoner  $I_l$  has to be identified with Hund's rule exchange). The universality

of the effective Stoner parameter  $I_2$  is also supported by experimental investigations using spin-polarized photoemission and inverse photoemission [24]. The spin-orbit coupling term is given by

$$H_{so} = \sum_i \xi_i \sigma_i \cdot L_i \quad (5)$$

where  $\xi_i$  is the spin-orbit-coupling matrix element calculated with the self-consistent scalar relativistic wavefunctions.  $H_{dip}$  describes the standard magnetic dipole-interactions of the local moments  $\mu_{il}$ . The same Hamiltonian, but excluding spin-orbit coupling and dipolar interactions has recently been used to describe successfully non-collinear spin-structures in amorphous magnets and substitutionally disordered intermetallic compounds [25].

The first step is the self-consistent calculation of the spin-polarized scalar-relativistic bandstructure for the bulk crystal or for a slab-model of the thin layer plus substrate, using the scalar-relativistic LMTO technique in the atomic sphere approximation (ASA). For the Fe/Cu(100) and Fe/Cu(111) layers considered in the present work, we used a slab with 11 Cu layers and one Fe layer, separated by six vacuum layers. The interatomic distances correspond to the bulk lattice constants of Cu. For the lowest eight Cu layers, the LMTO potential parameters are fixed at the bulk values, those for the three Cu layers closest to the Cu/Fe interface and for the vacuum layers the potential parameters and charge densities are calculated self-consistently. For the (100) slab we used a  $(\sqrt{2} \times \sqrt{2})$  surface cell with two atoms, for the (111) slab a  $(2 \times 2)$  surface cell with four atoms. The use of a non-primitive surface cell is immaterial in the present context, but important for prospective application to systems with antiferromagnetic interactions within layers. The use of a larger surface-cell also leads to a less anisotropic cell and helps to reduce the effects of charge-sloshing during the iteration. The local spin-orbit coupling constant  $\xi_i$  is calculated in the final iteration [26, 7]. For an LMTO with energy  $E$ , an energy-dependent spin-orbit coupling parameter  $\xi_i(E)$  is defined by the expectation value of  $\xi_i(\mathbf{r})$ . In our calculations we have used the spin-orbit coupling parameters calculated at the Fermi level. The self-consistent spin-dependent LMTO  $\alpha$ ASA Hamiltonian is decomposed into a paramagnetic part and the Hubbard-type exchange part defined in (3). A two-centre tight-binding Hamiltonian in the screened, most localized TB basis and the Löwdin orthonormal representation is constructed via the canonical transformation introduced by Andersen and Jepsen [18]. For the calculation of the screened structure constants of the TB-LMTO Hamiltonian, we consider an atomic environment of up to third-nearest neighbours.

In the second step, we allow for an arbitrary orientation of the local spin-quantization axes  $\zeta_i$  and we calculate self-consistently the magnitude and orientation of the local magnetic moments. Starting with a random distribution of the local spin-quantization axes  $\zeta_i$ , for each atomic site  $i$  the local spin-polarized partial densities of state  $n_{ilms}(E)^{\parallel,\perp}$  for spins parallel and perpendicular to  $\zeta_i$  are calculated using the real-space recursion method [23]. Integrating the  $n_{ilms}(E)^{\parallel,\perp}$  up to the Fermi level defines the updated local magnetic moment  $\mu_{ilm}$ . In general,  $\mu_{ilm}$  will have transverse components with respect to  $\zeta_i$  and the new local quantization axis  $\zeta'_i$  must be rotated into the direction of the moment. The calculation is iterated until the directions are stabilized and self-consistency according to (2) has been achieved. The main advantage of this technique is that in essence we calculate the magnetic torque force that rotates the moment into the direction of easy magnetization.

The recursion calculation is performed for a larger cell. For bulk iron we use a supercell consisting of  $8 \times 8 \times 8$  BCC elementary cells and periodic boundary conditions. For the thin layers we use a supercell with periodic boundary conditions in two dimensions only. Each 2D slab consists of 25 layers: three layers of empty spheres to account for the spilling-out of charge into the vacuum, one Fe layer and 21 Cu layers (with the potential of the lowest 18

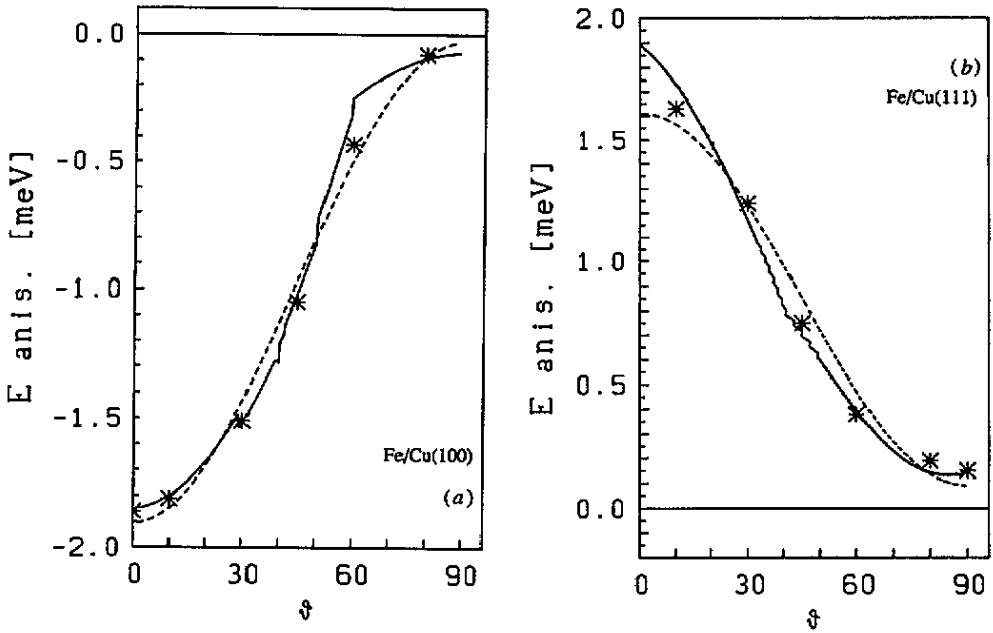
Cu layers fixed at the bulk values). Each (100) layer contains 288 atoms (a  $(12\sqrt{2} \times 12\sqrt{2})$  cell), each (111) layer 256 atoms (a  $(8 \times 8)$  cell) (set up by repeating the  $(\sqrt{2} \times \sqrt{2})$ , respectively  $(2 \times 2)$  cell in the lateral directions). Nine recursion levels were computed for the s orbitals, 12 for the p, and 27 for the d orbitals, using the Beer–Pettifor terminator [27] to get a smooth density of states.

In the third step we calculate the MAE. This may be done in the following ways. (a) adding a Zeeman term representing a magnetic field perpendicular to the easy axis to the Hamiltonian and calculating self-consistently the rotation of the magnetization as a function of the applied field. (b) orienting the magnetization nearly perpendicular to the easy axis by applying a sufficiently strong magnetic field. Then the magnetic field is switched off and the magnetic moment is allowed to relax into the preferred direction. This allows us to monitor the energy as a function of the direction of the magnetic moment. In each case the total energy was calculated exactly, including the double-counting corrections for the self-consistent charge- and spin-densities (earlier calculations used the force theorem to approximate the total energy by the sum of the one-electron eigenvalues). The disadvantage of method (a) is that it allows calculation of the MAE only at discrete orientations of the magnetization and that one has to subtract the effect of the field-induced polarization. Method (b) allows scanning of the variation of the MAE continuously. The disadvantage is that after switching off the initial magnetic field, the Hamiltonian is not completely self-consistent. Self-consistency is recovered only during the relaxation process. Both techniques lead to convergent results, except for small differences in the initial stage of the scanning process. This is shown in figure 1 for the Fe/Cu(100) and Fe/Cu(111) monolayers. With the ‘scanning’ approach (b), the variation of the energy shows small discontinuities at certain angles, arising from the changes in the bandstructure upon a re-orientation of the magnetic moments. These are precisely the effects that make the total-energy calculation in  $k$  space so cumbersome. In our technique, they are small and well controlled, since we follow these changes in the bandstructure continuously. The overall form of the MAE is well described by the  $E(\vartheta) = K_0 + K_{\text{anis}} \cos^2 \vartheta$  law for uniaxial anisotropy. In some respects, our approach is similar in spirit to the ‘state-tracking’ strategy of Freeman *et al* [8, 9]. However, Freeman *et al* study the behaviour of the bandstructure as a function of the spin–orbit coupling constant  $\xi$  at a few discrete values of the orientation of the moment, whereas we follow the change of the bandstructure as a function of the angle. Evidently this is much easier in our real-space approach than within a  $k$ -space formalism. We also note that our technique is parameter-free: all quantities determining the Hamiltonian (1) are derived from the *ab initio* LMTO calculation.

Our technique can also be used to calculate the very small anisotropies in the plane perpendicular to the easy axis. This is performed most easily in the ‘scanning’ mode (see below).

As a first test of the accuracy of our technique, we have calculated the magnetocrystalline anisotropy of crystalline BCC Fe. At the experimental lattice constant, we calculate a magnetic moment of  $\mu = 2.24 \mu_B$  oriented along the [100] direction (the easy axis). For the MAE we obtain  $\Delta E = E[100] - E[111] = -0.65 \mu\text{eV atom}^{-1}$ . Our results compare well with the calculations of Daalderop *et al* who found  $\mu = 2.25 \mu_B$  and  $\Delta E = -0.4 \mu\text{eV atom}^{-1}$  on the basis of an LMTO calculation using a set of  $\sim 500\,000$   $k$  vectors for Brillouin-zone integrations [7]. Previous calculations of the MAE of BCC Fe gave  $\Delta E = +7.4 \mu\text{eV atom}^{-1}$  (Fritsche *et al* [28]); the difference probably has to be attributed to too coarse a Brillouin-zone mesh. The experimental MAE is  $\Delta E = -1.3 \mu\text{eV atom}^{-1}$ , i.e. still slightly higher than our result [29].

For the Fe/Cu(100) monolayers we calculate spin–orbit coupling parameters  $\xi^\uparrow(E_F) =$



**Figure 1.** Variation of the total magnetic energy as a function of the angle  $\vartheta$  of the magnetization relative to the surface normal: (a) Fe monolayer on Cu(100), (b) Fe monolayer on Cu(111). Full line: calculated using method (b); relaxing the direction of the magnetic moment to the easy direction, stars: calculated for discrete orientations of the magnetic moments induced by a field perpendicular to the easy direction (method (b)), broken lines:  $\cos^2 \vartheta$ -fit. Cf. text.

59 meV and  $\xi^\downarrow(E_F) = 47$  meV for the spin-up and spin-down bands at the Fermi level. The coupling constants evaluated at the band centres are about 10% larger. These values are almost equal to the coupling constants calculated by Daalderoop *et al* [7] for bulk BCC Fe. The spin-orbit coupling constant for the Cu sites is  $\xi^{\uparrow,\downarrow}(E_F) = 108$  meV. For Fe/Cu(100) our calculations predict an easy axis oriented along the surface normal. The magnetic moment in the Fe-monolayer is enhanced to  $\mu = 2.7113 \mu_B$  for perpendicular orientation, the moments changes only very little for in-plane orientation ( $\mu = 2.7144 \mu_B$ ). The magnetic polarization of the Fe-layer induces very small moments in the first vacuum layer ( $\mu_{\text{vac}} = -0.02 \mu_B$ ) and in the first and second Cu layers from the interface ( $\mu_{\text{Cu1}} = 0.007 \mu_B$ ,  $\mu_{\text{Cu2}} = -0.012 \mu_B$ ) via the covalent coupling to the Fe spin-up and spin-down bands. The anisotropy constant (equal to the MAE for in-plane to perpendicular orientation) is  $K_{\text{anis}} = E_{\perp} - E_{\parallel} = -1.87$  meV atom $^{-1}$ , calculated via a fit of a  $\cos^2 \vartheta$ -fit to the results obtained using both techniques. These values are three orders of magnitude larger than the bulk MAE. Varying the spin-orbit coupling parameter  $\xi$  by  $\pm 10\%$  introduces changes in the MAE of the order of  $\pm 5\%$ .

For the Fe/Cu(111) monolayers, the spin-dependent difference in the spin-orbit coupling parameters is slightly larger:  $\xi^\uparrow = 60$  meV,  $\xi^\downarrow = 49$  meV. The enhancement of the magnetic moment in the Fe layer compared to the bulk value is smaller,  $\mu = 2.561 \mu_B$ , almost independent of the direction of the moment. Again small moments are induced in the vacuum and Cu layers ( $\mu_{\text{vac}} = -0.005 \mu_B$ ,  $\mu_{\text{Cu1}} = 0.010 \mu_B$ ,  $\mu_{\text{Cu2}} = -0.002 \mu_B$ ). For this orientation of the substrate, our calculation predicts an in-plane orientation of the magnetic moment, with an MAE that is of the same order of magnitude as for the Cu(100)

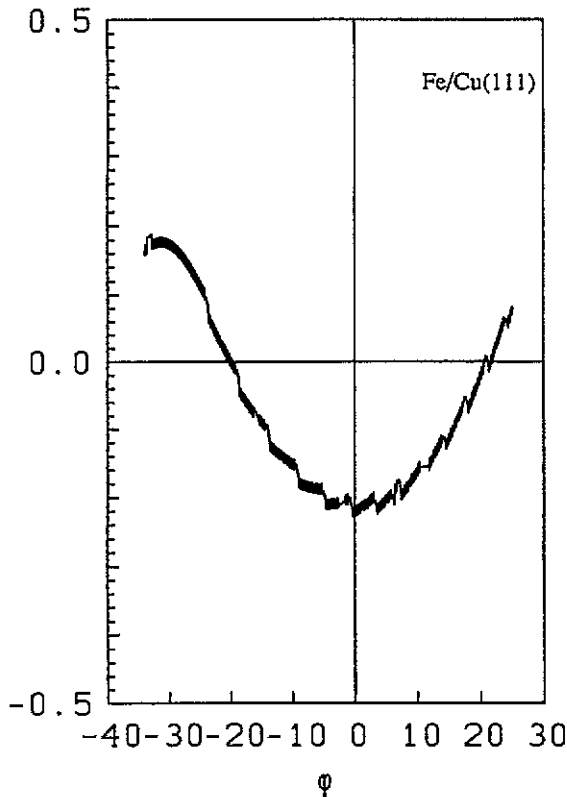


Figure 2. Variation of the total magnetic energy in Fe/Cu(111) as a function of the angle  $\varphi$  describing the orientation of the magnetization in the surface-plane ( $\varphi = 0$  corresponds to one of the basis vectors of the hexagonal surface cells).

substrate,  $K_{\text{anis}} = E_{\perp} - E_{\parallel} = 1.52 \text{ meV atom}^{-1}$ . Here again, we have checked that this result is stable against variations of the parameters, especially the spin-orbit coupling constant. We find that  $\xi(E_F)$  has to be increased to three times the value resulting from the LMTO calculations to switch the easy axis into the perpendicular direction.

The prediction of perpendicular magnetization for Fe/Cu(100) and in-plane magnetization for Fe/Cu(111) is in agreement with experiment [14, 15]. However, quantitative results for the MAE in the monolayer-limit are not available due to limitations arising from insular growth, interdiffusion or surface-segregation of Cu etc. The magnetic moments for Fe/Cu(100) are in good agreement with the result of Blügel [30],  $\mu = 2.85\mu_B$ . The MAE for free-standing transition-metal mono- and multilayers have been calculated by Bruno [11], Cinal *et al* [13], and Pick and Dreysse [12] using parametrized tight-binding Hamiltonians. The results in the monolayer case are of the same order of magnitude as our predictions, but depend crucially on adjustable parameters such as the crystal-field and spin-orbit-coupling parameters, bandfilling, etc, so that a quantitative comparison is not meaningful.

Our technique is also sufficiently accurate for studying the much smaller in-plane anisotropies for an easy axis lying in the surface plane. This may be achieved by orienting the moments in an off-symmetry direction in the plane and relaxing it into the easy direction. An example is shown in figure 2 for Fe/Cu(111). Due to the exceedingly small MAE,

the scans of the magnetic energy become more noisy, but it is still possible to follow the angular dependence and to deduce the anisotropy constants. The MAE for magnetic moments pointing into the direction of the nearest-neighbour atom and pointing into the direction of the next-nearest-neighbour atoms is  $\Delta E = 0.40 \mu\text{eV atom}^{-1}$ , i.e., even smaller than the bulk MAE.

In summary, we have presented a novel technique for calculating uniaxial and planar anisotropies in bulk and thin-film magnetic systems. Our approach exploits essentially the magnetic-torque force restoring the magnetic moment to the easy axis. By following continuously the changes introduced in the electronic structure as a function of the direction of the magnetization, it avoids the cumbersome computational problems that plague other techniques based on total-energy calculations. One of the particular advantages of our technique is that it is not restricted to collinear orientations of the magnetic moments. This will allow the investigation of systems with competing ferro- and antiferromagnetic interaction and the study of the regime where a switching of the easy axis from perpendicular to in-plane orientation occurs. These studies are now in progress.

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## References

- [1] Heinrich B and Cochran J F 1993 *Adv. Phys.* **42** 523
- [2] Allenspach R 1994 *J. Magn. Magn. Mater.* **129** 160
- [3] Van Vleck J H 1937 *Phys. Rev.* **52** 1178
- [4] Néel L 1954 *J. Phys. Radium* **15** 505
- [5] Victora R H and McLaren J M 1993 *Phys. Rev. B* **47** 11583
- [6] Gay J G and Richter R 1986 *Phys. Rev. Lett.* **56** 2728
- [7] Daalderop G H O, Kelly P J and Schuurmans M F H 1990 *Phys. Rev. B* **41** 11919; **42** 7270
- [8] Li C, Freeman A J, Jansen H J F and Fu C L 1990 *Phys. Rev. B* **42** 5433
- [9] Wang D S, Wu R and Freeman A J *Phys. Rev. Lett.* **70** 869
- [10] Takayama H, Bohnen K P and Fulde P 1976 *Phys. Rev. B* **14** 2287
- [11] Bruno P 1989 *Phys. Rev. B* **39** 865
- [12] Pick Š and Dreyssé H 1993 *Phys. Rev. B* **48** 13588
- [13] Cinal M, Edwards D M and Mathon J 1994 *Phys. Rev. B* **50** 3754
- [14] Allenspach R and Bischof A 1992 *Phys. Rev. Lett.* **69** 3385
- [15] Li D, Freitag M, Pearson J, Qiu Z Q and Bader S D 1994 *Phys. Rev. Lett.* **72** 3112
- [16] Kraft T, Marcus P M and Scheffler M 1994 *Phys. Rev. B* **49** 11511
- [17] Skriver H 1984 *The LMTO-Method* (Berlin: Springer)
- [18] Andersen O K and Jepsen O 1984 *Phys. Rev. Lett.* **53** 2571
- [19] Heine V, Samson J H and Nex C M M 1982 *J. Phys. F: Met. Phys.* **11** 177
- [20] Turek I, Becker Ch and Hafner J 1992 *J. Phys.: Condens. Matter* **4** 7257
- [21] Becker Ch and Hafner J 1994 *Phys. Rev. B* **50** 3933
- [22] Anisimov V I, Zaanen J and Andersen O K 1991 *Phys. Rev. B* **44** 943
- [23] Haydock R, Heine V and Kelly M J 1975 *J. Phys. C: Solid State Phys.* **8** 2591
- [24] Himpfel F J 1991 *Phys. Rev. Lett.* **67** 2363
- [25] Lorenz R and Hafner J *J. Magn. Magn. Mater.* **139** 209
- [26] Andersen O K 1975 *Phys. Rev. B* **12** 3060
- [27] Beer N and Pettifor D 1984 *The Electronic Structure of Complex Systems* ed P Phariseau and W M Temmermann (New York: Plenum)
- [28] Fritsche L, Noffke J and Eckhardt H 1987 *J. Phys. F: Met. Phys.* **17** 943
- [29] Escudier P 1975 *Ann. Phys.* **9** 125
- [30] Weinert M and Blügel 1993 *Magnetic Multilayers* ed L H Bennett and R E Watson (Singapore: World Scientific)