The influence of spin-independent disorder on giant magnetoresistance

1996 J. Phys.: Condens. Matter 8 L569

(http://iopscience.iop.org/0953-8984/8/40/001)

J. Phys.: Condens. Matter **8** (1996) L569–L575. Printed in the UK

LETTER TO THE EDITOR

The influence of spin-independent disorder on giant magnetoresistance

E Yu Tsymbal and D G Pettifor

Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, UK

Received 9 August 1996

Abstract. We demonstrate that the giant magnetoresistance (GMR) effect in magnetic multilayers can be explained quantitatively in terms of the scattering of electrons from a *spin-independent* random potential that arises from the grown-in defects within the multilayer. We have calculated the GMR ratio for Co/Cu and Fe/Cr multilayered systems within the Kubo–Greenwood formalism assuming that the on-site atomic energies are disordered randomly within a realistic spd tight-binding model. Our predictions are in good agreement with experiment and demonstrate how the GMR ratio depends on the features of the electronic band structure. In particular, we obtain the enhancement of GMR in Co/Cu multilayers at electron energies up to about 1 eV above the Fermi level that has recently been observed by Monsma *et al* [1995 *Phys. Rev. Lett.* **74** 5260]. We predict no such enhancement for Fe/Cr multilayers.

The mechanism of giant magnetoresistance (GMR) in magnetic multilayers [1] is usually related to the *spin dependence* of the scattering processes. In the semiclassical models of GMR this spin dependence is introduced into the theory through a number of spin-dependent phenomenological parameters such as relaxation times and transmission coefficients [2, 3]. In the quantum mechanical models the spin dependence of the scattering is assumed to arise from spin-dependent random potentials produced by magnetic impurities at the interfaces or in the bulk of the ferromagnetic layers [4–6]. These models usually take the electronic structure of the host material to be spin independent, assuming that transport is carried out by the s electrons which are described by the free-electron model [2–5] or by the single-band tight-binding model [6]. Recently, however, it has been shown that in the ballistic regime of conductance the spin dependence of the electronic structure already gives a sizeable contribution to GMR [7]. Several attempts have been made at combining this realistic description of the electronic structure with spin-dependent scattering potentials for a predictive model of the diffusive regime [8–10]. Unfortunately, these calculations imply unrealistically large values of the GMR compared to experiment.

In this letter we demonstrate that a realistic prediction of GMR can be obtained within a model that introduces *spin-independent disorder* in the on-site atomic energy levels in conjunction with accurate *spin-dependent electronic structure*. In particular, we will show that this model predicts the recently observed enhancement of GMR for hot electrons in Co/Cu spin valves [11]. Our model is motivated by experimental observations that magnetic multilayers contain a lot of defects such as vacancies, impurities and grain boundaries that are produced during the process of deposition (e.g. [12]). Each of these defects makes its own contribution to the scattering potential and, consequently, to the resistivity. However, on average there is no reason for this random scattering potential to be spin dependent. One would expect the appearance of a strong spin dependence only for the case when chemically

0953-8984/96/400569+07$19.50

abrupt interfaces of thin magnetic layers have a high density of steps and the bulk resistivity is small [13, 14]. Otherwise, there will be no significant contribution to resistivity and GMR from spin-dependent scattering potentials. We believe that the dominant mechanism of GMR is driven by the spin-independent scattering potential within the magnetic multilayers.

We, therefore, model the influence of the defects on the electron scattering by assuming spin-independent disorder in the on-site atomic energy levels within a realistic tight-binding (TB) description of the electronic structure. The Hamiltonian of the magnetic multilayer system may, thus, be written in the form $\hat{H} = \hat{H}_0 + \hat{V}$, where the first term is the TB Hamiltonian of the perfect multilayer and the second term is the random potential. The Schrödinger equation $\hat{H}_0 |n\boldsymbol{k}\rangle = E_n(\boldsymbol{k}) |n\boldsymbol{k}\rangle$ for the unperturbed system is solved within the two-centre, orthogonal TB approximation by expanding the eigenstate with band index n and wave vector $\boldsymbol{k}$ in terms of atomic orbitals:

$$|n\boldsymbol{k}\rangle = \frac{1}{\sqrt{N}} \sum_{mj\alpha} \mathrm{e}^{\mathrm{i}\boldsymbol{k}(\boldsymbol{R}_m + \boldsymbol{r}_j)} c_{n,j\alpha}(\boldsymbol{k}) |mj\alpha\rangle \tag{1}$$

where $|mj\alpha\rangle$ are the atomic basis functions with respect to the unit cell m, basis site j, and atomic orbital α. Here N is the total number of cells in the crystal, $\boldsymbol{R}_m$ is the mth lattice vector and $\boldsymbol{r}_j$ is the basis vector. The expansion coefficients $c_{n,j\alpha}(\boldsymbol{k})$ are obtained by solving the TB secular equation in the usual way.

The scattering potential $\hat{V}$ is assumed to be diagonal with respect to cell, site and orbital indices so that

$$\langle mj\alpha| \hat{V} |m_1 j_1 \alpha_1\rangle = V_{mj\alpha} \delta_{mm_1} \delta_{jj_1} \delta_{\alpha\alpha_1}. \tag{2}$$

The diagonal elements of the potential are then taken to be randomly distributed such that

$$\left\langle V_{mj\alpha} \right\rangle = 0 \qquad \left\langle V_{mj\alpha} V_{m_1 j_1 \alpha_1} \right\rangle = \gamma^2 \delta_{mm_1} \delta_{jj_1} \delta_{\alpha\alpha_1} \tag{3}$$

where $\langle ... \rangle$ denotes the configuration average and γ^2 is the mean square displacement of the on-site energies reflecting the defect scattering. The configurational averaging is performed using the standard technique which requires that the original configuration-dependent Green function of the system $\hat{G}^{(\pm)} = (E_F - \hat{H} \pm \mathrm{i}\varepsilon)^{-1}$, when averaged over all random configurations, should equal the Green function of the effective medium [15]

$$\left\langle \hat{G}^{(\pm)} \right\rangle = \left(E_F - \hat{H}_0 - \hat{\Gamma}^{(\pm)} \pm \mathrm{i}\varepsilon \right)^{-1}. \tag{4}$$

The self-energy $\hat{\Gamma}$ characterizes the relaxation properties of the system and yields the energy shift resulting from the scattering as well as the damping of the electronic states. It is found by applying the weak scattering limit [15]:

$$\hat{\Gamma}^{(\pm)} = \left\langle \hat{V} \hat{G}_0^{(\pm)} \hat{V} \right\rangle \tag{5}$$

where $\hat{G}_0^{(\pm)}(E_F) = (E_F - \hat{H}_0 \pm \mathrm{i}\varepsilon)^{-1}$ is the Green function of the perfect crystal. For the weak scattering limit to be valid the random potential must be such that $\gamma n(E_F) \ll 1$, where $n(E_F)$ is the average density of states (DOS) per orbital at the Fermi energy E_F. In order to obtain typical values of the resistivity of 3d metal multilayer structures, i.e. 1–100 $\mu\Omega$ cm, one needs γ to be in the range 0.1 to 1 eV, whereas $n(E_F)$ is of the order of 0.1 eV^{-1}. Therefore, we see that the weak scattering approximation is justified. It follows from equations (1)–(3) that the self-energy can be written as

$$\hat{\Gamma}^{(\pm)}_{j\alpha,\, j_1\alpha_1}(E_F) = \gamma^2 \sum_n \int \frac{\Omega \, \mathrm{d}\boldsymbol{k}}{(2\pi)^3} \frac{\left| c_{n,j\alpha}(\boldsymbol{k}) \right|^2}{E_F - E_n(\boldsymbol{k}) \pm \mathrm{i}\varepsilon} \delta_{jj_1} \delta_{\alpha\alpha_1} \tag{6}$$

where Ω is the volume of the unit cell.

The conductivity is calculated using the Kubo–Greenwood formula [16, 17]

$$\sigma_{\mu\nu} = \frac{\pi\hbar e^2}{N\Omega}\mathrm{Tr}\left\langle \hat{v}^{\mu}\delta\left(E_F - \hat{H}\right)\hat{v}^{\nu}\delta\left(E_F - \hat{H}\right)\right\rangle \tag{7}$$

where $\sigma_{\mu\nu}$ is the conductivity tensor for a definite spin direction and $\hat{v}^{\mu}$ is the velocity operator. The δ function is defined by $\delta(E_F - \hat{H}) = (\hat{G}^{(-)} - \hat{G}^{(+)})/2\pi\mathrm{i}$. We decouple the configurational average of the product of the two Green functions in equation (7) as $\left\langle \hat{G}^{(\pm)}\hat{v}^{\mu}\hat{G}^{(\pm)}\right\rangle = \left\langle \hat{G}^{(\pm)}\right\rangle \hat{v}^{\mu}\left\langle \hat{G}^{(\pm)}\right\rangle$. This is exact within the weak scattering limit of our model for the current-in-plane (CIP) geometry, but is an approximation for the current-perpendicular-to-plane (CPP) geometry as the vertex corrections do not vanish by symmetry in this latter case [18]. Using equation (4) we finally obtain

$$\sigma_{\mu\nu} = -\frac{e^2}{\pi\hbar}\int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}\left[\hat{\Lambda}^{\mu}(\boldsymbol{k})\hat{\Lambda}^{\nu}(\boldsymbol{k})\right] \tag{8}$$

where the scattering-path operator $\hat{\Lambda}^{\mu}(\boldsymbol{k})$ is defined by

$$\hat{\Lambda}^{\mu}(\boldsymbol{k}) = \frac{\hbar\hat{v}^{\mu}(\boldsymbol{k})}{2}\left[\frac{1}{E_F - \hat{H}_0(\boldsymbol{k}) - \hat{\Gamma}^{(-)}} - \frac{1}{E_F - \hat{H}_0(\boldsymbol{k}) - \hat{\Gamma}^{(+)}}\right]. \tag{9}$$

The spin dependence of the conductivity enters equation (8) not only through the spin-dependence of the perfect multilayer Hamiltonian and velocity operators, $\hat{H}_0$ and $\hat{v}^{\mu}$ respectively, but also through the spin dependence of the scattering rate $\tau_n^{-1}(\boldsymbol{k}) = 2\mathrm{Im}\Gamma_{nn}^{(-)}(\boldsymbol{k})/\hbar$. Unlike Zahn *et al* [10], who neglected the state dependence of the scattering rate within the semiclassical treatment of electronic transport, our approach includes explicitly the state dependence of the scattering which is central to obtaining realistic values of GMR. Moreover, we have found that the semiclassical approximation itself significantly over estimates GMR if it is applied in the interval of resistivities typical for multilayers due to the neglect of interband transitions [18].

Using equation (8) we have calculated the conductivity of (100) oriented fcc $\mathrm{Co}_n/\mathrm{Cu}_n$ and bcc $\mathrm{Fe}_n/\mathrm{Cr}_n$ multilayers with $n = 1$, 2 and 3 for the parallel (P) and antiparallel (AP) alignment of their magnetizations. Calculations were performed using s, p, and d TB bands fitted to *ab initio* band structures of the corresponding metals [19]. The resulting DOS for the multilayers are in good agreement with LMTO calculations. The parameter γ was chosen to be 0.6 eV to provide realistic resistivities of corresponding multilayers at the saturation field, namely about 18 $\mu\Omega\,$cm for Co/Cu and 38 $\mu\Omega\,$cm for Fe/Cr [1, 12]. Calculations were performed for both CIP and CPP geometries. We note that the results of our computations show that for the CPP geometry the difference in the local conductivities is negligible and, consequently, the electric field can be considered to be uniform within the unit cell and, hence, there is no spin mixing of currents [20].

In this letter we focus on $\mathrm{Co}_2/\mathrm{Cu}_2$ and $\mathrm{Fe}_2/\mathrm{Cr}_2$ multilayers. We see in figure 1(a) that as expected the DOS of the P-aligned $\mathrm{Co}_2/\mathrm{Cu}_2$ multilayer is asymmetric between the majority and minority spins. The DOS for the AP configuration is similar to the DOS for the P configuration averaged between the majority and minority spins. In figure 1(b) the conductivity σ is plotted as a function of the energy, or equivalently as a function of the Fermi energy E_F. The features displayed in this figure can be qualitatively understood as follows. The conductivity is expected to be proportional to the mean free path of the electrons, which, in its turn, is proportional to the electron velocity at the Fermi level and inversely proportional to the DOS. Therefore, σ will be low if E_F lies within the d band due to the low velocity and the high density of states. As is seen from figure 1(b) the

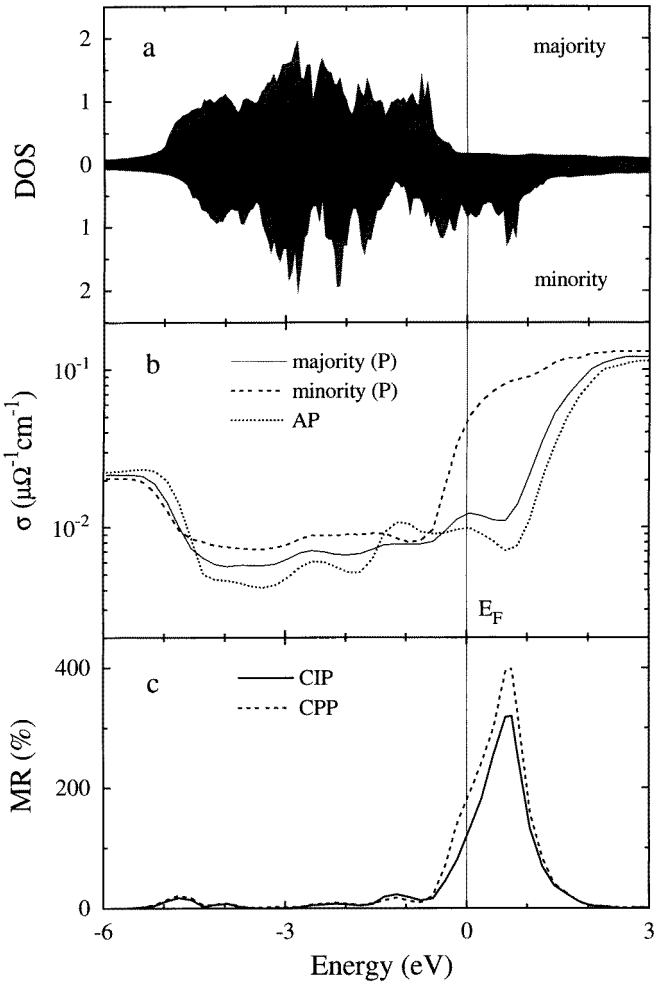

Figure 1. The calculated results for the Co_2/Cu_2 multilayer. (*a*) Density of states (in eV^{-1} per atom) for the parallel alignment of the magnetizations for majority spins (top) and minority spins (bottom). (*b*) Conductivity in the CIP geometry as a function of the electron energy for majority spins (dashed line) and minority spins (dotted line) for parallel (P) configuration and for antiparallel (AP) configuration (solid line). (*c*) Magnetoresistance as a function of the electron energy for CIP (solid line) and CPP (dotted line). The vertical line denotes the position of the Fermi level.

variation in σ as a function of the energy qualitatively reproduces the washed-out features of the DOS for the d bands of the majority and minority spins. Above the top of the d band σ increases rapidly. This increase is due to the contribution of the sp band which is characterized by a high velocity of electrons. For the Co_2/Cu_2 multilayer the Fermi level lies approximately 0.6 eV above the top of the majority-spin d band (figure 1(a)) which results in the crucial difference between the conductivities of the P and AP configurations. The GMR ratio we have found for the Co_2/Cu_2 system is approximately 110% for CIP and 180% for CPP. The result for the CIP geometry is in good agreement with experiment where a GMR ratio of 115% has been measured at 4.2 K [21].

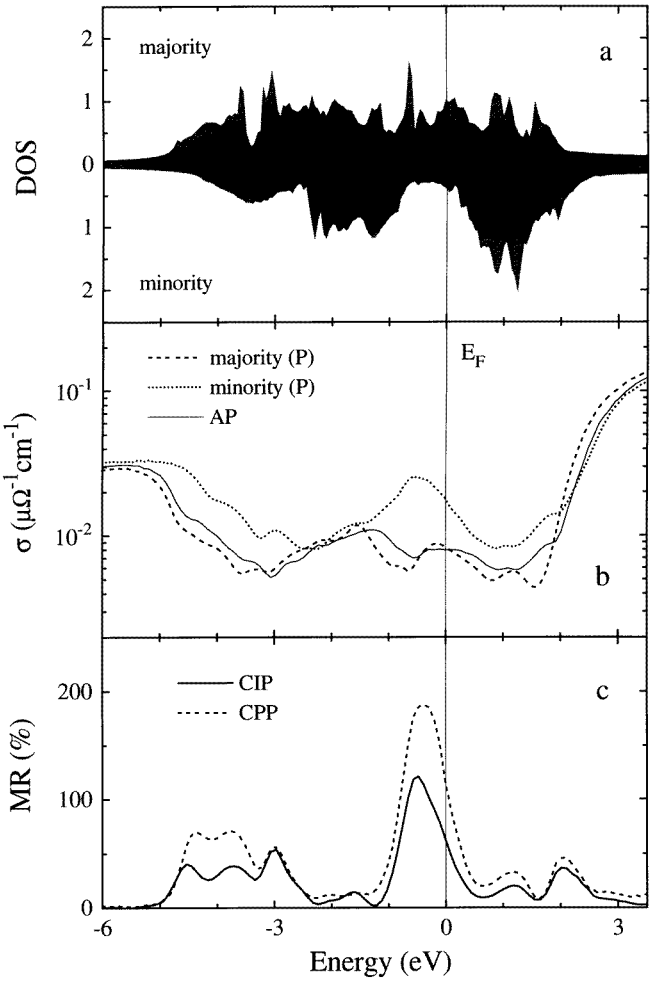

Figure 2. The same as in figure 1 for the Fe_2/Cr_2 multilayer.

The GMR ratio is plotted as a function of the electron energy in figure 1(c). We see that the magnetoresistance in the Co_2/Cu_2 system increases with increasing energy *above* the Fermi level. A pronounced peak in the GMR ratio appears at about 0.7 eV above the Fermi level, taking the large value of nearly 400% for the CPP geometry. The position of this peak is shifted to about 0.8 eV for the Co_3/Cu_3 multilayer. Using the calculated DOS for the Co_n/Cu_n multilayers for $n > 3$, we have concluded that for large n this peak will be shifted up to about 1 eV above the Fermi energy, i.e. to the top of the d band for the minority spin electrons. The enhancement of GMR in Co/Cu multilayers has very recently been observed where more than the 390% CPP magnetocurrent change has been measured in a 'spin-valve transistor' [11]. The energy of the hot electrons in this device was defined by the Schottky barrier heights of about 0.7 eV for the collector and about 0.6 eV for the emitter. Our calculations give a very clear interpretation of this enhanced GMR effect.

The Fe/Cr system behaves differently from the Co/Cu system. As is evident from figure 2(a), the Fermi level in the Fe_2/Cr_2 multilayer lies within the d band for *both*

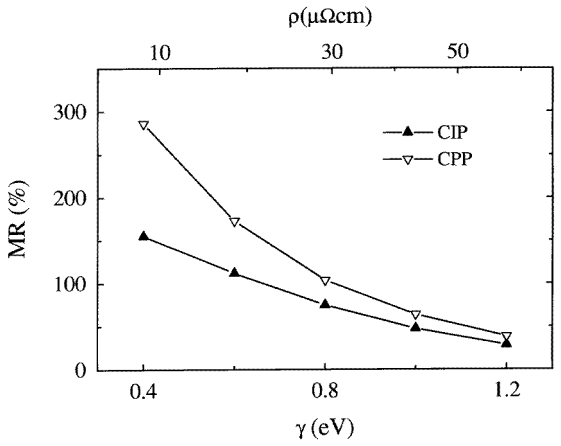

Figure 3. Magnetoresistance as a function of the mean square displacement of the on-site energies for the Co_2/Cu_2 multilayer. Corresponding CIP resistivity at the saturation field is schematically shown for comparison.

spin orientations. Consequently, there is no large difference in the Fermi velocities for the majority and minority spin electrons. However, due to its bcc structure, the Fe_2/Cr_2 multilayer exhibits a pronounced valley in the DOS of the minority spins, with the Fermi level lying almost at the bottom of this valley. Therefore, the spin asymmetry of the conductivity in figure 2(b) is connected mainly with the spin asymmetry of the DOS. The calculated CIP GMR ratio is approximately 65% which is in good agreement with experiment [1]. The CPP GMR value is about 120% . The variation of the magnetoresistance as a function of the energy in figure 2(c) shows that there are several peaks related mainly to the differences in the DOS. Increasing GMR is expected for the energies *below* the Fermi level. Hence, our calculation does not predict the possibility of enhanced GMR in hot electron devices for the Fe/Cr system.

Figure 3 shows the predicted dependence of the GMR on the mean square displacement of the on-site energies for Co_2/Cu_2 multilayers. We have varied γ in the interval 0.4 to 1.2 eV so that the resistivity of the multilayer changes by one order of magnitude, spanning the range of experimental values. The curve shows that increasing γ always causes a drop in the magnetoresistance. For example, the CIP GMR ratio decreases from about 160% to about 30% (as γ changes from 0.4 to 1.2 eV). The corresponding change of resistivity for the P orientation is from about 8 to 56 $\mu\Omega$ cm. As γ characterizes the degree of disorder in the system due to defects, an improvement in the perfection of the multilayer should enhance the magnetoresistance. We note, however, that there should be a limitation to this enhancement at small γ due to spin-flip processes arising from electron–magnon interactions [12].

The spin independence of the scattering potential within our model does *not* imply that the interfaces between the magnetic and non-magnetic layers are peripheral to the origin of GMR. In this letter we have performed calculations for small unit cells, so that in a sense all the atoms are at the interface. However, preliminary calculations based on our model show that GMR in the $Co_1Ni_2Co_1Cu_2$ multilayer is about twice as large as that for the $Ni_1Co_2Ni_1Cu_2$ multilayer, demonstrating the importance of the different magnetic layers at the interfaces. This result is in qualitative agreement with experiments

on interfacial 'dusting' [22].

In conclusion, we have shown that *spin-independent disorder* in the on-site atomic energy levels accounts for the observed enhancement of GMR in Co/Cu spin valves provided the electronic structure is realistically modelled. We predict no such enhancement for Fe/Cr systems. The model is currently being used to explore other materials that might lead to enhanced values of GMR for spin-valve applications.

This research was supported by Hewlett–Packard Laboratories in Palo Alto through a collaborative research program. We happily acknowledge the stimulation provided by discussions with Drs Tom Anthony, Alex Bratkovsky, Jim Brug, Girvin Harkins, Chuck Morehouse and Stan Williams at Hewlett-Packard and Drs Pascale Bayle-Guillemaud, John Jakubovics, Amanda Petford-Long, and Tchavdar Todorov at Oxford. The calculations were performed in the Materials Modelling Laboratory at the Department of Materials, University of Oxford.

References

[1] Baibich M N *et al* 1988 *Phys. Rev. Lett.* **61** 2472
[2] Camley R E and Barnas J 1989 *Phys. Rev. Lett.* **63** 664
[3] Hood R Q and Falicov L M 1992 *Phys. Rev.* B **46** 8287
[4] Levy P M, Zhang S and Fert A 1990 *Phys. Rev. Lett.* **65** 1643
[5] Vedyayev A, Dieny B and Ryzhanova N 1992 *Europhys. Lett.* **19** 329
[6] Itoh H, Inoue J and Maekawa S 1995 *Phys. Rev.* B **51** 342
[7] Schep K M, Kelly P J and Bauer G E W 1995 *Phys. Rev. Lett.* **74** 568
[8] Butler W H, MacLaren J M and Zhang X G 1993 *MRS Symp. Proc.* **313** 59
[9] Nesbet R K 1994 *J. Phys.: Condens. Matter* **6** L449
[10] Zahn P, Mertig I, Richer M and Eschrig H 1995 *Phys. Rev. Lett.* **75** 2996
[11] Monsma D J, Lodder J C, Popma T J A and Dieny B 1995 *Phys. Rev. Lett.* **74** 5260
[12] Bland J A C and Heinrich B (ed) 1994 *Ultrathin Magnetic Structures I, II: An Introduction to the Electronic, Magnetic and Structural Properties* (Berlin: Springer)
[13] Schad R *et al* 1995 *J. Magn. Magn. Mater.* **148** 331
[14] Todorov T N, Tsymbal E Yu and Pettifor D G 1996 *Phys. Rev.* B at press
[15] Velicky B 1969 *Phys. Rev.* **184** 614
[16] Kubo R 1957 *J. Phys. Soc. Japan* **12** 570
[17] Greenwood D A 1958 *Proc. Phys. Soc. London* **71** 585
[18] Tsymbal E Yu and Pettifor D G 1996 *Phys. Rev.* B at press
[19] Papaconstantopoulos D A 1986 *Handbook of the Band Structure of Elemental Solids* (New York: Plenum)
[20] Camblong H E, Zhang S and Levy P M 1993 *Phys. Rev.* B **47** 4735
[21] Parkin S S P, Li Z G and Smith D J 1991 *Appl. Phys. Lett.* **58** 2710
[22] Parkin S S P 1993 *Phys. Rev. Lett.* **71** 1641