Ab Initio Calculations of the Giant Magnetoresistance

P. Zahn and I. Mertig
Technische Universität Dresden, Institut für Theoretische Physik, D-01062 Dresden, Germany

M. Richter and H. Eschrig
Technische Universität Dresden, MPG-Arbeitsgruppe "Elektronensysteme" D-01062 Dresden, Germany
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Ab initio calculations are presented for the giant magnetoresistance (GMR) of Fe/Cr multilayers. The electronic structure of Fe/Cr superlattices is calculated within the spin density functional theory. The spin-dependent bulk impurity scattering is treated in a fully quantum mechanical way using a Green function formalism. The transport is described quasiclassically. The calculations suggest that the GMR in thin multilayers is mainly determined by the electronic structure of the layered system. Spin-dependent impurity scattering can reduce, increase, or invert the effect.

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The discovery of giant magnetoresistance (GMR) in magnetic multilayer systems [1,2] initiated experimental and theoretical work to elucidate the microscopic origin of the phenomenon. The GMR effect occurs when subsequent ferromagnetic layers in the stack have their magnetization antiparallel. Application of an external magnetic field brings the magnetization of the ferromagnetic layers into alignment and causes a decrease of resistivity for both the current-in-plane (CIP) and the current-perpendicular-to-plane (CPP) geometries. Most theories [3–8] try to explain the GMR by spin-dependent scattering at interfaces or bulk defects but neglect the electronic structure of the multilayer system. Attempts to include the electronic structure have been made by several authors [9–11]. Oguchi [9] examined the dependence of the GMR on Fermi velocities but neglected the specific influence of scattering. Schep, Kelly, and Bauer [11] investigated the influence of the electronic structure on the GMR effect but only in the ballistic regime of transport, which is not the regime where the experiments are carried out. The first attempt to include the electronic structure of the layered system and the scattering by interface roughness was done by Butler *et al.* [10] using a coherent potential approximation to simulate interface roughness. All calculations predict a strong influence of the electronic structure of the multilayer on the GMR.

In this paper we present *ab initio* calculations of the GMR for layers thin compared to the electron mean free path, by including the full electronic structure of the superlattice, as well as a first principles description of spin-dependent electron scattering by impurities. The transport is treated quasiclassically by solving the linearized Boltzmann equation, an approach recently successfully applied to the calculation of residual resistivities for dilute ferromagnetic alloys [12–14].

The GMR in magnetic multilayers is defined to be

$$\mathrm{GMR} = \frac{\sigma^P}{\sigma^{AP}} - 1\,, \tag{1}$$

where σ^P and σ^{AP} are the conductivities of the multilayer for parallel and antiparallel alignment of subsequent ferromagnetic layer magnetization, respectively. Experimentally, one can be sure of the realization of parallel alignment due to the marked transition from strong to weak magnetic field dependence of the conductivity at the saturation field. Because of real structure at the interfaces, on the other hand, the ideal antiparallel alignment is probably never realized at zero field, and the GMR given by (1) will always be an upper bound of the experimentally found value.

To be specific, the main part of our considerations focuses on (100) oriented $\mathrm{Fe}_m\mathrm{Cr}_n$ layer sequences (m monolayers Fe followed by n monolayers Cr), where the Fe layers are intrinsically ferromagnetic and the Cr layers are intrinsically antiferromagnetic. We consider only cases with n even, because otherwise no antiparallel alignment of subsequent Fe layer magnetization (up to $n = 24$) appears. *Ab initio* electronic structure calculations have been performed using spin-density functional theory within the frame of an optimized LCAO (linear combination of atomic orbitals) scheme [15]. For initially parallel and antiparallel spin configurations of subsequent Fe layers, the iterations converge to local energy minima corresponding to those configurations, respectively. The unit cell is prolonged in stacking direction and is to be doubled in the antiparallel case compared to the parallel one. The lattice parameter was chosen to be that of bcc bulk Fe. For the self-consistency of the potentials 1440 **k** points in the Brillouin zone have been used. The final electronic structure was calculated at a mesh of 74 400 **k** points in the Brillouin zone. The Fermi surface and the necessary Fermi surface integrations have been treated using a modified tetrahedron method [16]. Given the band structure ε_k^σ of the just described situation, potential scattering causes transitions from a state k into a state k', where k is a shorthand notation for the wave vector **k** and the band index ν, with

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probability

$$P^{\sigma}_{kk'} = 2\pi |T^{\sigma}_{kk'}|^2 \delta(\varepsilon^{\sigma}_k - \varepsilon^{\sigma}_{k'}). \quad (2)$$

σ indicates the spin direction and T is the scattering T matrix. We restrict our considerations to spin-conserving scattering only, assuming it to be dominant. For a concentration c of scatterers the electron lifetime is

$$(\tau^{\sigma}_k)^{-1} = c \sum_{k'} P^{\sigma}_{kk'}. \quad (3)$$

Averaging over the Fermi surface leads to a state-independent but spin-dependent relaxation time

$$\tau^{\sigma} = \frac{\sum_k \delta(\varepsilon^{\sigma}_k - \varepsilon_F)\tau^{\sigma}_k}{\sum_k \delta(\varepsilon^{\sigma}_k - \varepsilon_F)}. \quad (4)$$

ε_F is the Fermi energy. The spin dependence of the scattering is expressed by the anisotropy ratio $\beta = \tau^{\uparrow}/\tau^{\downarrow}$. Following Mott's idea [17], the transport properties of ferromagnetic alloys can be explained by assuming conduction in parallel by electrons in the majority bands (spin up, $\uparrow$) and the minority bands (spin down, $\downarrow$). For each spin direction the quasiclassical Boltzmann equation was solved in relaxation time approximation, which leads to a conductivity tensor per spin direction

$$\sigma^{\sigma} = e^2 \tau^{\sigma} \sum_k \delta(\varepsilon^{\sigma}_k - \varepsilon_F) \mathbf{v}^{\sigma}_k \mathbf{v}^{\sigma}_k; \quad (5)$$

$\mathbf{v}^{\sigma}_k$ is the Fermi velocity. The integration is performed over the Fermi surface of the parallel configuration of the layered system. Addition of the two currents yields the total conductivity

$$\sigma^{P} = \sigma^{\uparrow} + \sigma^{\downarrow}. \quad (6)$$

In the antiparallel configuration the electronic states are spin degenerate, and the conductivity becomes

$$\sigma^{AP} = 2e^2 \tau^{AP} \sum_k \delta(\varepsilon^{AP}_k - \varepsilon_F) \mathbf{v}^{AP}_k \mathbf{v}^{AP}_k. \quad (7)$$

If we assume z to be the growth direction of the layered system, CIP corresponds to the xx or yy component of the conductivity tensor and CPP to the zz component in Eqs. (5) and (7). τ^{AP} is the relaxation time in the antiparallel configuration and can be obtained by addition of the scattering operators. For equal concentration of defects in adjacent ferromagnetic layers the relaxation time is then given by

$$\frac{1}{\tau^{AP}} = \frac{1}{2}\left(\frac{1}{\tau^{\uparrow}} + \frac{1}{\tau^{\downarrow}}\right). \quad (8)$$

The simplest situation appears if all scattering centers are localized in the antiferromagnetic Cr layers (for example, Fe impurities). In this case, the lifetimes τ^{σ} for both spin directions must be equal, since the spin up wave-function amplitudes of one-half of the Cr atoms equal the spin down wave-function amplitudes of the other half. This corresponds to $\beta = 1$. Equation (8) now implies $\tau^{AP} = \tau^{\uparrow} = \tau^{\downarrow}$. Hence the lifetime completely drops out of the GMR expression,

$$\mathrm{GMR} = \frac{\sum_{\sigma}\sum_k \delta(\varepsilon^{\sigma}_k - \varepsilon_F) v^{\sigma}_{k_i} v^{\sigma}_{k_i}}{2\sum_k \delta(\varepsilon^{AP}_k - \varepsilon_F) v^{AP}_{k_i} v^{AP}_{k_i}} - 1, \quad (9)$$

where v_{k_i} are the Cartesian components of the Fermi velocity. This GMR is completely determined by the Fermi surface and Fermi velocities as functions of the magnetization configuration, and is hence a pure band-structure effect. Note that this result is different from that obtained for the ballistic regime [11], and, in a certain sense, is more general, since the conductivity itself remains dependent on the scattering. The obtained GMR values are about 200% in CIP and 700% in CPP [dots in Figs. 1(a) and 1(b)], which is in agreement with the experimentally obtained maximum CIP-GMR (220%) for ultrathin Fe layers [18].

The spin dependence of the relaxation time ($\beta \neq 1$) due to scattering centers in the ferromagnetic layers (Cr impurities in Fe layers, for example) leads to modifications. GMR is now to be calculated via (1), (6), and (7).

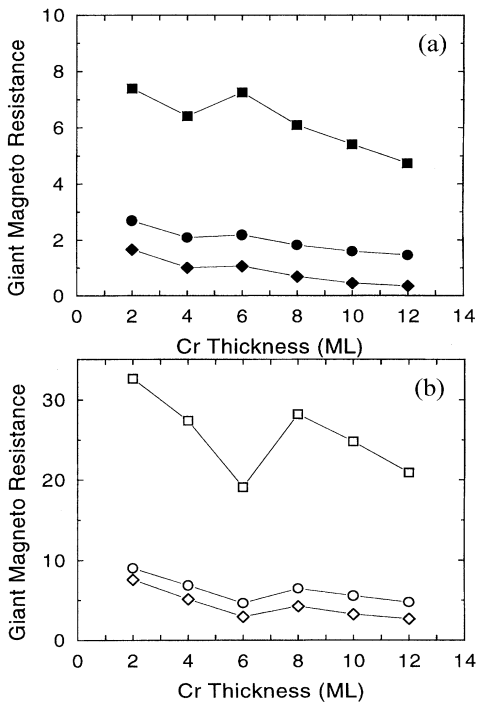

FIG. 1. Calculated GMR for Fe_3Cr_n in dependence on the Cr layer thickness $n = 2, 12$ monolayers (ML). The results assuming Cr defects in Fe layers are indicated by squares ($\beta = 0.11$). Dots mark the results for spin-independent scattering ($\beta = 1$) and diamonds for $\beta = \beta_{\min}$. (a) CIP geometry (closed symbols). (b) CPP geometry (open symbols).

The transition matrix elements $T^{\sigma}_{kk'}$ for the scattering of Bloch electrons by an impurity cluster embedded in an ideal host crystal are described within the KKR Green function formalism [16,19–21]. Under the assumption that the extension of the impurity is small compared to the layer thickness, the scattering properties are described by the spin-dependent relaxation times calculated for the Cr impurities in Fe [12,14]. For this case, $\beta = 0.11$, that is, the majority electrons are scattered strongly at a Cr defect and the minority electrons pass the defect weakly scattered.

The results, assuming Cr defects in the Fe layers, only lead to a GMR of about 600% in CIP [solid squares in Fig. 1(a)] and 2500% in CPP [open squares in Fig. 1(b)], which is larger than the experimental results [22–25]. The anisotropy ratios β obtained in [12,14] for a variety of defects scatter from 0.1 to 10. Therefore, GMR is discussed as a function of β in relaxation time approximation. If only one type of scatterer is included, GMR takes on a minimum for

$$\beta_{\min} = \frac{\tau^{\uparrow}}{\tau^{\downarrow}} = \sqrt{\frac{\sum_k \delta(\varepsilon_k^{\downarrow} - \varepsilon_F) v_{k_i}^{\downarrow 2}}{\sum_k \delta(\varepsilon_k^{\uparrow} - \varepsilon_F) v_{k_i}^{\uparrow 2}}}. \quad (10)$$

That is, GMR can be enhanced or reduced by spin-dependent impurity scattering. The results for $\beta = \beta_{\min}$ are shown in Figs. 1(a) and 1(b) (diamonds).

Finally, the combination of alternating Fe layers with Cr defects ($\beta = 0.11$) and with Cu defects ($\beta = 3.68$) is discussed. For simplicity, equal concentration is assumed. The relaxation times in parallel configuration are

$$\frac{1}{\tau^{\sigma}} = \frac{1}{2}\left(\frac{1}{\tau^{\sigma}_{\mathrm{Cr}}} + \frac{1}{\tau^{\sigma}_{\mathrm{Cr}}}\right), \quad (11)$$

and in antiparallel configuration

$$\frac{D_{\uparrow}^{+} + D_{\uparrow}^{-}}{\tau^{\uparrow}} = \left(\frac{D_{\uparrow}^{+}}{\tau^{+}_{\mathrm{Cr}}} + \frac{D_{\uparrow}^{-}}{\tau^{-}_{\mathrm{Cu}}}\right) \quad (12)$$

and

$$\frac{D_{\downarrow}^{+} + D_{\downarrow}^{-}}{\tau^{\downarrow}} = \left(\frac{D_{\downarrow}^{-}}{\tau^{-}_{\mathrm{Cr}}} + \frac{D_{\downarrow}^{+}}{\tau^{+}_{\mathrm{Cu}}}\right), \quad (13)$$

respectively, where the superscripts + and − correspond to majority and minority electrons. D_{σ}^{+} and D_{σ}^{-} are the local spin-dependent density of states in a ferromagnetic layer where spin-σ electrons are majority or minority electrons, respectively. In the considered configuration the factors have been chosen to be 1. The conductivity in the antiparallel state including both defects is given by

$$\sigma^{AP} = e^2(\tau^{\uparrow} + \tau^{\downarrow}) \sum_k \delta(\varepsilon_k^{AP} - \varepsilon_F) \mathbf{v}_k^{AP} \mathbf{v}_k^{AP}. \quad (14)$$

The results are shown in Fig. 2. The main message from this calculation is that the GMR can be reduced drastically or even change sign if scatterers with different spin anisotropy are combined. (If they would be located in the same layer they would merely act as an effective scattering potential.) One should note that in Fig. 2 the inverse effect occurs in CIP but not in CPP. Experimentally, the inverse effect was obtained for Fe/Cr/Fe/Cu/Fe/Cu multilayers [26], as well as in $Fe_{1-x}V_x$/Au/Co multilayers [27], both in CIP geometry. Preliminary calculations for Fe_3Cu_9 multilayers with Cr and Cu defects in subsequent Fe layers show a CIP-GMR of −35% and a CPP-GMR of 200%. Concerning V instead of Cr defects, the scattering potential of V impurities in Fe is very similar to Cr defects in Fe. The spin anisotropy of Fe(V) is 0.15 in comparison to 0.11 for Fe(Cr).

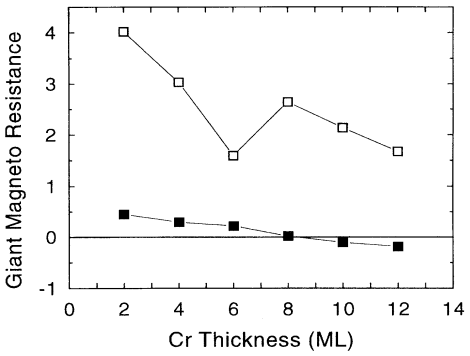

FIG. 2. Calculated GMR for Fe_3Cr_n with Cr and Cu defects in adjacent Fe layers in CIP (closed squares) and CPP (open squares). For $n \geq 10$ the inverse GMR is obtained.

Generally, it should be noted that CPP-GMR for comparable scattering is always larger than CIP by a factor of about 4 in agreement with experimental results [25]. This factor stems from the difference of the Fermi velocity components in plane and perpendicular to the plane, and it finds its natural explanation in the fact that the carriers with large momentum components in direction of the current are more influenced by the superstructure in CPP.

Moreover, GMR (Figs. 1–3) shows characteristic variations with layer thicknesses. Both the magnetic moments

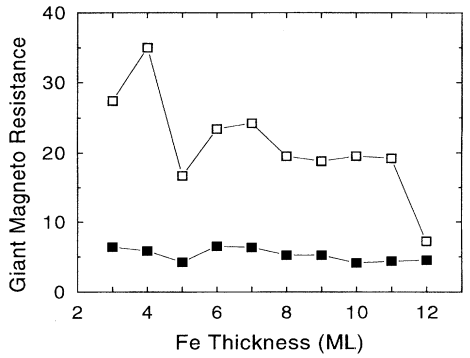

FIG. 3. Calculated GMR for Fe_mCr_4 in dependence on the Fe layer thickness $m = 3, 12$ monolayers (ML) ($\beta = 0.11$) in CIP (closed squares) and CPP (open squares).

and the *averaged* Fermi velocities show little dependence on the layer thickness. The variations stem from a subtle game of the superstructure gaps on the Fermi surface. In Fig. 3, these variations are reminiscent of experimentally found oscillations [28].

In conclusion, it has been shown that GMR under the assumption of a coherent multilayer system and a spin-independent impurity scattering is determined by the changes of the electronic structure as a function of the magnetization direction. Spin-dependent impurity scattering can enhance or reduce the effect. The combination of impurities with different spin anisotropy can cause in particular the inverse GMR.

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