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LETTER TO THE EDITOR

Giant magnetoresistance due to spin-dependent interface scattering

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Abstract. Spin-dependent electrical resistivity due to scattering by displaced interface atoms has been computed for a layered CuCo superlattice, using full-potential multiple-scattering theory with no free parameters. The magnetoresistance ratio $\Delta R/R(\uparrow\uparrow)$ obtained for this scattering mechanism is 25.03. When interface resistivity, weighted by interpretation concentration $c \simeq 0.10$, is combined with bulk resistivity, $\Delta R/R$ is in the expected experimental range for adjacent CuCoCu layers. This resistance mechanism produces spin-dependent steady-state chemical potentials, relevant to the bipolar spin switch.

Giant magnetoresistance (GMR) is observed in layered Fe/Cr [1, 2], and in other materials in which magnetic layers are separated by a conducting non-magnetic spacer material [3–9]. If the magnetization of successive magnetic layers is originally not parallel, electrical resistivity is greatly reduced when parallel alignment is forced by an applied magnetic field. Application of GMR to magnetic recording devices may greatly increase data densities compared with existing technology.

The present work is intended to clarify the underlying mechanism of GMR. The relaxation time compared here is strongly spin dependent and varies over the Fermi surface. Hence the Boltzmann equation implies that the displacement of the Fermi distribution function in momentum space due to an applied electric field depends on spin orientation. This produces a spin-polarized steady-state current flow that depends on spin orientation. It also implies the existence of spin- and space-dependent chemical potentials. This chemical potential response (CPR) will be discussed here as a contribution to the mechanism for the recently discovered bipolar spin switch [10].

GMR occurs when the mean free path in a magnetized material is strongly spin dependent, primarily due to spin-dependent interface scattering, but also from spin-dependent bulk scattering. GMR has been studied [11–15] by a spin-dependent generalization of the Fuchs–Sondheimer theory of thin-film conductivity [16, 17]. The Fuchs–Sondheimer theory treats interface scattering differently from bulk scattering. Hood and Falicov [15] have recently extended the theory to include potential wells that depend on spin alignment, using a more complete description of spin-dependent interface scattering. This parametrized theory is in reasonable accord with experiment. Several authors [18–21] have used quantum theory based on the Kubo formalism. All of these prior theoretical studies depend on parametrization and on simplified models of the electronic energy band structure.

The present work reports first-principles calculations of the electronic structure and electrical conductivity of a superlattice model of spin-polarized CuCo. The translational

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unit cell is (CuCoCu)/(CuCoCu) on a 001 tetragonal lattice whose atoms have the geometry of FCC Cu. Transport theory is simplified here in several ways in order to make calculations tractable, but consequences of the spin-dependent energy band structure are worked out in detail. The essential approximations are that the interface penetration scattering mechanism is treated in the weak-scattering limit of isolated impurities, and scattering-in terms (vertex corrections in Kubo theory) are neglected in the Boltzmann equation. This latter approximation was used by Butler and Stocks [22] in calculations on substitutional alloys, later extended using Kubo theory by Butler [23]. The approximation appears to be justified for transition elements when s, d–s, d transitions are important, and has recently been used for GMR calculations [24]. The formalism developed by these authors can be applied to extend the present calculations to fully quantitative results, but this was not considered essential for the present exploration of basic scattering mechanisms. Calculation of interface scattering beyond the present weak-scattering limit requires treatment of interpenetrating atomic layers as a binary alloy.

If there is no thermal gradient, and if mean free paths are larger than the translational unit cell, the Boltzmann equation for a regular periodic solid requires only Fermi surface data and state to state transition probabilities for dissipative scattering mechanisms. All required quantities relevant to the Fermi surface are obtained here from first-principles calculations, as are transition probabilities for scattering due to random interpenetration of adjacent Co and Cu layers. Bulk scattering is treated much less specifically by using empirical values of the mean free path for residual resistivity in pure Co and Cu.

In multiple-scattering theory, as used here in the LACO (linearized atomic-cell orbital) method [25], energy bands $\varepsilon(k, s)$ are determined by the secular equation $\det(\mathbf{I} - \mathbf{tg}) = 0$. Here $\mathbf{t}(\varepsilon)$ is a site-diagonal atomic-cell scattering matrix and $\mathbf{g}(k)$ is an energy-independent matrix of structure constants. The energy-band structures of magnetic and spacer species in typical GMR materials match closely at the Fermi energy for one spin direction, while there is substantial mismatch for the opposite spin [26]. The **t**-matrices of the two species must have a similar relationship, since energy bands in a fixed space lattice are determined entirely by the **t**-matrices. If atoms are interchanged due to interpenetration at a spacer interface, the difference of **t**-matrices must be small for one spin direction and large for the other, implying strong dependence of scattering on spin. The transition amplitude for this scattering mechanism is determined in the weak-scattering limit by the impurity scattering matrix $\Delta\mathbf{t}$, the difference between the **t**-matrices of the interchanged species, evaluated at the Fermi energy.

Comparison of Co and Cu densities of states indicates that matrix elements $\Delta\mathbf{t}$ should be small for majority-spin electrons and large for minority-spin electrons in layered CuCo with parallel Co spins. This expectation is confirmed by the present computations. Matrix elements $\Delta\mathbf{t}$ are of intermediate size for antiparallel Co spins. Spin dependence of the computed transition probabilities implies spin dependence of the resistivity. The present computations indicate that for parallel alignment of magnetization in the Co layers the minority-spin resistivity is much greater than the majority-spin resistivity. Since the resistivity for anti-parallel alignment is of intermediate magnitude, the magnetoresistance is very large.

This spin-dependent scattering mechanism differs from the well known Mott effect, which provides an explanation of the increased resistivity of transition metals compared with noble metals. Since transitions into final d states reduce the relaxation time of a wave packet at the Fermi energy, resistivity due to any dissipative scattering mechanism depends on spin if the density of states does so. When the transition probability itself depends on spin, stronger spin dependence is possible than that due to the Mott effect alone. Scattering

due to the atomic-cell **t**-matrices alone is phase-coherent, described by transmission and reflection coefficients that depend on spin and direction, but does not contribute to electrical resistivity. In contrast, scattering due to $\Delta\mathbf{t}$ is dissipative, in the sense that each Bloch wave acquires an energy width or inverse lifetime, characteristic of impurity scattering by a displaced atom at a random site.

A relaxation time τ is defined if collisions cause a perturbed distribution function to relax locally in its parameter space and exponentially in time to the equilibrium distribution f_0. If τ is defined, the classical steady-state Boltzmann equation for a homogeneous material with no thermal gradient implies the electrical conductivity tensor

$$\sigma_{ij} = -e^2 \int \tau v_i \left(\partial f_0 / m\, \partial v_j\right) \mathrm{d}^2 v. \tag{1}$$

The semiclassical Boltzmann equation for electrons replaces $f(x; v)$ via the Fermi–Dirac distribution function $f(k, b, s)$, indexed by distinct eigenstates, specified by k in the reduced Brillouin zone, by a band index b, and by a spin index s. Classical electron velocity is replaced by wave-packet group velocity, $v = (1/2\hbar)\nabla_k \varepsilon$, in atomic units if the energy ε is in Rydberg units. In the reduced Brillouin zone for a translational cell of volume Ω, the number-of-states element is $([\![2]\!]\Omega/8\pi^3)(1/2\hbar v)\,\mathrm{d}\varepsilon\,\mathrm{d}S$, where $\mathrm{d}S$ is an element of the constant-ε surface. In a spin-polarized system, the factor indicated here by $[\![2]\!]$ is replaced by a sum over the spin index. On the Fermi surface, $v_j = v_r l_j$, where l is a unit normal vector, and at $T = 0$ K, $\partial f_0/\hbar\,\partial k_j = -2v_F l_j \delta(\varepsilon - \varepsilon_F)$.

If scattering-in terms in the Boltzmann equation are neglected, the relaxation time τ is the reciprocal of the transition probability at the Fermi surface,

$$\tau^{-1}(k, b, s) = \frac{\Omega}{8\pi^3} \sum_{b'} \int \mathrm{d}S' \left(\hbar v'_F\right)^{-1} \frac{2\pi}{\hbar} \left|\left(k', b', s\left|\Delta\mathbf{t}_s\right|k, b, s\right)\right|^2. \tag{2}$$

In equation (2), the final density of states is proportional to the reciprocal Fermi velocity. An increased final density of states for either spin implies a relative decrease of the relaxation time. This is the Mott effect giving spin-dependent conductivities even for spin-independent transition probabilities.

The implied semiclassical electric current density is $j_i = \sum_i \sigma_{ij} E_j$, where the conductivity tensor is

$$\sigma_{ij} = \frac{e^2}{8\pi^3\hbar} \sum_s \int_{\varepsilon=\varepsilon_F} \tau v_F l_i l_j \,\mathrm{d}S. \tag{3}$$

A sum over distinct energy bands, assumed in equation (3), is not indicated explicitly. Equation (3) implies that electrical conductivity can depend on spin only through the spin dependence of the Fermi surface or of the mean free path $\lambda(k, b, s) = \tau v_F$. In general, σ is dominated by the largest values of λ, and spin dependence of λ implies magnetoresistance.

The electronic distribution function is modified by an electric field, implying a change ΔV in the chemical potential $\mathrm{d}E/\mathrm{d}N$, whose steady-state value is $\varepsilon_F + V(E) + \Delta V$. By analogy with the electrical conductivity tensor, $\Delta V = \sum_i \nu_i E_i$ defines a chemical potential response (CPR) vector ν. The Schrödinger–Dirac operator for $\mathrm{d}E/\mathrm{d}N$ is the one-electron Hamiltonian $\mathcal{H}$, whose eigenvalues at the Fermi level are one-electron removal energies. Hence the Kubo formula for σ is converted to that for ν by replacing an electronic current operator by $\mathcal{H}$. In the semiclassical Boltzmann equation $-ev$ for each Bloch wave $\phi_{k,b,s}(x)$ is replaced by the one-electron energy. The resulting CPR vector is

$$\nu_i = -\frac{e}{8\pi^3\hbar} \varepsilon_F \sum_s \int_{\varepsilon=\varepsilon_F} \tau l_i \,\mathrm{d}S. \tag{4}$$

Electrical potential at a point x is obtained from equation (4) by multiplying $|\phi_{k,b,s}(x)|^2$ into the integrand. Full translational symmetry implies that the Fermi surface integral of the vector integrand in equation (4) must vanish, but this does not hold for the component of v normal to a free surface. When translational symmetry is broken, an experiment that measures a voltage difference in the presence of spin-dependent current flow would measure the net result of CPR and of the IR drop due directly to resistivity along the path of current flow. Both effects must show the same dependence on spin orientation that is observed in GMR. Such a voltage difference has recently been observed by Johnson [10]. To model this experiment in principle requires solving the coupled Maxwell and Schrödinger equations for the three-dimensional local electric field and the local current density, in order to distinguish between the CPR as such and the IR drop due to the detailed local current flow.

The LACO system of programs for electronic structure calculations, implementing full-potential multiple-scattering theory in the local density approximation, has recently been extended to ferromagnetic metals. New methodology, used recently for empty-lattice tests [25], has been applied to self-consistent calculations. Preliminary calculations on FCC Cu and on both paramagnetic and ferromagnetic FCC Co provided initial radial density functions for 001-tetragonal (CuCoCu)(CuCoCu), on the Cu lattice, for which self-consistent calculations were carried to convergence for both parallel $\uparrow\uparrow$ and antiparallel $\uparrow\downarrow$ alignments of the Co spin axes. A local basis designated by spd{fg} was used. The orbital basis sets on each atom included (spd) orbital basis functions at each electronic band energy, with intermediate boundary sums carried through (fg) solid harmonics.

The matrix $\Delta\mathbf{t}$ for the interface scattering mechanism considered here is the difference of spin-dependent self-consistent atomic-cell $\mathbf{t}$-matrices computed, as described recently [27], from rectangular matrices characteristic of full-potential theory. Matrix elements of $\Delta\mathbf{t}$ were computed using Bloch wave eigenvectors of the secular matrix. Incoherent scattering by each displaced atom was assumed. Equations (2) and (3) were used to compute the state-dependent relaxation time and spin-dependent electrical conductivity tensor, respectively. Integrals over the Fermi surface required in these expressions were carried out by a tetrahedral interpolation scheme. These calculations have not been carried to convergence with respect to the number of points in the Brillouin zone, but test calculations indicate that the interpolation method used is relatively stable, and that the preliminary results given here are not subject to large error. A subsequent publication will give details of the computations.

Relaxation times due to the interpenetration effect were computed in the form $c\tau$, where c is the (unknown) concentration of Co atoms exchanged with Cu in adjacent planes. The spin dependence of $c\tau$ and dependence on spin polarization is very large, characterized in atomic units by the orders of magnitude

$$\uparrow\uparrow\ (F) \qquad c\tau(\uparrow) \simeq 10^4 \qquad c\tau(\downarrow) \simeq 10^1$$

$$\uparrow\downarrow\ (a) \qquad c\tau(\uparrow) \simeq 10^2 \qquad c\tau(\downarrow) \simeq 10^2.$$

The resulting conductivity tensor is strongly spin dependent. Computed in-plane conductivities, again multiplied by the concentration c, are (in atomic units)

$$\uparrow\uparrow\ (F) \qquad c\sigma(\uparrow) = 2.0378 \qquad c\sigma(\downarrow) = 0.0032 \qquad c\sigma(F) = 2.0410$$

$$\uparrow\downarrow\ (a) \qquad c\sigma(\uparrow) = 0.0392 \qquad c\sigma(\downarrow) = 0.0392 \qquad c\sigma(a) = 0.0784.$$

The implied magnetoresistance ratio, defined here by $(\rho(F) - \rho(a))/\rho(F) = (\sigma(F) - \sigma(a))/\sigma(a)$, is 25.03 from this mechanism alone. Results of a similarly large magnitude have recently been reported from layered KKR/CPA calculations [24].

Recent first-principles calculations by Sticht [28] are similar to the present work, but do not evaluate a specific scattering mechanism, and take the relaxation time to be constant and

independent of spin. This is inconsistent with equation (2) here, since the spin-dependent Fermi surface and Fermi velocity imply spin-dependent τ. The present results include this Mott effect, together with the direct effect of spin-dependent interface scattering. In order to separate these two aspects of the physical mechanism, the calculations were repeated with the state-dependent transition probability replaced in equation (2) by its average over initial state and spin. The modified values of $c\sigma$ computed in this way are

$$\uparrow\uparrow\ (F) \qquad c\sigma(\uparrow) = 0.0419 \qquad c\sigma(\downarrow) = 0.0027 \qquad c\sigma(F) = 0.0446$$

$$\uparrow\downarrow\ (a) \qquad c\sigma(\uparrow) = 0.0038 \qquad c\sigma(\downarrow) = 0.0038 \qquad c\sigma(a) = 0.0076.$$

The implied magnetoresistance ratio is 4.87. Hence, in the present example, the Mott effect by itself leads to significantly smaller GMR than does the combination of Mott effect and spin-dependent interface scattering.

A model of bulk scattering is provided by using spin- and state-independent values of the mean free path λ in equation (3). Empirical low-temperature values for typical experimental materials are [29] λ(Cu) = 200 Å, λ(Co) = 70 Å. $1/\lambda$ for each species is weighted by the relative number of atoms in the unit cell. The implied values of σ for pure bulk scattering are

$$\uparrow\uparrow\ (F) \qquad \sigma(\uparrow) = 1.2288 \qquad \sigma(\downarrow) = 1.1610 \qquad \sigma(F) = 2.3898$$

$$\uparrow\downarrow\ (a) \qquad \sigma(\uparrow) = 1.4350 \qquad \sigma(\downarrow) = 1.4350 \qquad \sigma(a) = 2.8700.$$

The implied magnetoresistance ratio for spin-independent bulk scattering is -0.17, opposite in sign to the observed GMR. This result indicates that despite the spin dependence of the Fermi surface, purely spin-independent scattering cannot account for GMR.

For comparison with observed GMR, resistivities due to bulk scattering and to interface scattering must be combined. In the weak-scattering limit, scattering probabilities due to bulk and interface mechanisms are additive, and the implied spin-dependent resistivities must be added separately for the spin-up and spin-down electrons in the two-fluid model postulated here. The resulting spin-indexed current densities are independent, and are of greatly different magnitude in the spin-aligned material. Adding the spin-indexed conductivities gives a rational formula that interpolates $\Delta R/R$ between pure bulk and interface scattering limits as a function of c. If the interpenetration concentration is $c \simeq 0.10$, which is not unreasonable for atomic species closely spaced in the periodic table, and other quantities are taken from the data given above, this formula implies $\Delta R/R \simeq 1.0$, comparable to an estimate of $\simeq$2.0 [29] for the empirical limit in Cu/Co at low temperature, with adjacent magnetic and spacer atomic layers, but no true bulk spacer material.

The relationship between conductivity and chemical potential response given by equations (3) and (4) here indicates that GMR and CPR are different manifestations of the same underlying phenomenon—spin-selective dissipative scattering in a magnetic material. CPR must occur in all GMR materials. Johnson [10] has recently reported voltage measurements on an electronic switching device consisting of a paramagnetic metal film P sandwiched between two ferromagnetic films, F1 and F2. A potential difference detected between P and F2 is proportional to the electric current in F1 and changes depending on the relative orientation of the magnetization vectors of F1 and F2. The expected CPR in such a system will contribute to the measured voltage difference, but is combined with the IR drop due to the full three-dimensional current flow, which is not separately measured. The impedance mechanism considered by Johnson [10] is a spin-dependent change of the electronic distribution function, and as such must be describable by the Boltzmann or Kubo theory. If so, some dissipative scattering effect must always be taken into account in order

to characterize the dynamical balance in a steady state. The results given here indicate that interpenetration scattering, which certainly is present in layered magnetic materials, may be the predominant spin-dependent mechanism in these materials.

In conclusion, the present first-principles calculations show that interface scattering due to interpenetration of adjacent magnetic and spacer atomic layers can produce large magnetoresistance and, by implication, a corresponding chemical potential response, if the difference between atomic scattering t-matrices at the Fermi energy depends strongly on spin orientation. Hence spin-dependent energy band structure can be correlated with the occurrence of GMR and of CPR. Comparing published densities of states [30], Co/Cu bands agree for majority-spin electrons and disagree for minority-spin electrons at the Fermi energy, while the opposite is true for Fe/Cr and Fe/Rh. The present study predicts that electric current is spontaneously polarized in spin-aligned magnetized GMR materials. Opposite polarization is expected in Co/Cu and Fe/Cr (or Fe/Rh), respectively.

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