Interplay of the specular and diffuse scattering at interfaces of magnetic multilayers

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Both specular and diffuse scattering at interfaces of magnetic multilayers play a significant role in their resistivity and magnetoresistance. Within the framework of the semiclassical Boltzmann approach, we show that the resistance across an interface for currents perpendicular to the plane of the layers is sensitive to the nature of the scattering and that perturbative treatments of diffuse interface scattering break down. Depending on reflection coefficients diffuse scattering can *assist* or *suppress* conduction across interfaces.
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Giant magnetoresistance (GMR) has been observed in transition-metal magnetic multilayers in two principal geometries. For the current parallel to the plane of the layers (CIP), a number of authors[1–4] have studied the interface roughness from different angles. Noticeably, Hood, Falicov, and Penn[3] considered the relation between the structures of roughness and its effect on resistivity in detail. Recently, much attention has centered on currents perpendicular to the plane of the layers (CPP) since the magnitude of the GMR is larger in this geometry. One of the most important issues in this geometry is the origin of interface resistance. Valet and Fert[5] introduced a phenomenological potential drop at interface due to diffuse scattering but without potential steps, and Stiles[6] considered realistic specular reflection but without diffuse scattering. Vedyaev *et al.*[7] evaluated effects of interference between incident and reflected electrons for an ideal interface. Dugaev *et al.*,[8] and Barnas and Fert[9] considered the effects of diffuse scattering at interfaces in the presence of potential steps, but they limited their calculations to second order of the scattering potential. Finally, Ustinov and Kravtsov[10] made a study similar to the one presented here; as we discuss later on they made an assumption about the variation of the electric field that is in general unwarranted.

In the transition-metal magnetic multilayers, diffuse scattering is strong and specular reflection at interfaces is significant as shown in Refs. 11,12. What is needed is a general analysis which can assess the interplay of the putatively *strong* diffuse and specular scattering at interfaces. By extending the semiclassical Boltzmann treatment of Hood and Falicov[3] to the CPP geometry we find (1) a term exists in the boundary conditions for the CPP geometry which is absent in CIP, (2) while diffuse scattering increases resistance for small reflection coefficients at interfaces, it can *assist* conduction for large reflection coefficients, i.e., it can induce impurity assisted conduction, and (3) while general solutions are difficult to obtain, an exact solution of the interface resistance is found for a special form of the specular and diffuse coefficients; with this special solution, we show the interplay of the specular and diffuse scattering on the interface resistance.

The semiclassical linearized Boltzmann equation for metallic layered structures is

$$ev_z E_z \frac{\partial f^0}{\partial \epsilon} + v_z \frac{\partial f}{\partial z} = -\frac{1}{(2\pi)^3}\int d^3\mathbf{k}'\,\delta(\epsilon_k - \epsilon_{k'})W_{\mathbf{k}\mathbf{k}'}(z) \times[f(\mathbf{k},z) - f(\mathbf{k}',z)], \tag{1}$$

where E_z is the external electric field applied in the direction perpendicular to the plane of the layers, f is the distribution function (f^0 is the equilibrium distribution function), $v_z = \partial\epsilon_{\mathbf{k}}/\partial k_z$ is the velocity in z direction, and it is assumed that the collision is *local and elastic*. One can greatly simplify the above equation when the scattering probability $W_{\mathbf{k}\mathbf{k}'}(z) = W_0(z)$ is independent of momenta $\mathbf{k}$ and $\mathbf{k}'$. Following,[5] we can separate the distribution function as follows:

$$f(\mathbf{k},z) = f^0(\mathbf{k}) + \left(-\frac{\partial f^0}{\partial \epsilon}\right)[g(\mathbf{k},z) + e\mu(z)], \tag{2}$$

where the condition $\int g(\mathbf{k},z)d^3\mathbf{k} = 0$ is imposed. By placing Eq. (2) into Eq. (1) with the above constraint on $g(\mathbf{k},z)$, one arrives at

$$v_z \frac{\partial g}{\partial z} + \frac{g}{\tau(z)} = ev_z E(z); \tag{3}$$

the effective electric field $E(z) = E_z - (\partial/\partial z)\mu$ and the relaxation time $\tau(z)$ is defined as $\tau^{-1}(z) = \pi N(\epsilon)W_0(z)$, where N is the local density of states at the Fermi level. A commonly used approximation for the relaxation time $\tau(z)$ is to assume that it is layerwise constant, and here we adopt this convention. In the absence of spin-flip scattering Eq. (3) can be applied independently to each spin channel; as we are not stressing the implication of our analysis solely on GMR we omit the spin index for simplicity of notation.

In the presence of specular reflection *and* diffuse scattering at interfaces, solutions for the electric field and distribution functions are highly nontrivial. In the absence of the current, a dipole layer forms at an interface, this comes from the charge redistribution at the interface to insure a common chemical potential across layers. This dipole layer does not give rise to a voltage drop at the interface. What we are interested in is the current-driven (nonequilibrium) dipole layer at an interface which comes from charge transport across the interface. Such a dipole layer, which does not exist

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in CIP geometry, leads to finite voltage drops at interfaces in the CPP geometry. Our goal is to find this voltage drop in the presence of arbitrary strong diffuse scattering and specular reflections.

In current experimental realizations of metallic multilayered structures, specular (potential step) and diffuse scattering coexist. There are two limiting cases. When the layer thickness is much smaller than the mean free path, one needs to calculate the electronic structure of the multilayer in the present of potential steps. The coherent scattering from different interfaces leads to superlattice bands which are weakly perturbed by diffuse scattering either in the layers or at the interfaces. Schep *et al.*,[15] and Mertig[16] have performed such calculations.

The case most relevant to current experimental realization is that of layer thickness comparable to the mean free paths in the layers. The interference of scattering from different interfaces is unimportant,[3,13,14] and one can single out each interface separately as suggested by Barnas and Fert.[9] In the following, we consider two semi-infinite layers (layer 1 for $z<0$ and layer 2 for $z>0$). The dipole layer, of atomic dimensions, is assumed to be small compared to the mean free paths in the layers; in a first approximation we assume it infinitely thin. To solve the Boltzmann equation, one must specify boundary conditions at the interface $z=0$, which have the following generalized forms in the presence of diffuse and specular scattering:

$$f(\mathbf{v}_2^>)=S(\mathbf{v}_1,\mathbf{v}_2)R(\mathbf{v}_1,\mathbf{v}_2)f(\mathbf{v}_2^<)+S(\mathbf{v}_1,\mathbf{v}_2)T(\mathbf{v}_1,\mathbf{v}_2)f(\mathbf{v}_1^>) +[1-S(\mathbf{v}_1,\mathbf{v}_2)]F_1(f) \tag{4}$$

and

$$f(\mathbf{v}_1^<)=S(\mathbf{v}_1,\mathbf{v}_2)R(\mathbf{v}_1,\mathbf{v}_2)f(\mathbf{v}_1^>)+S(\mathbf{v}_1,\mathbf{v}_2)T(\mathbf{v}_1,\mathbf{v}_2)f(\mathbf{v}_2^<) +[1-S(\mathbf{v}_1,\mathbf{v}_2)]F_2(f), \tag{5}$$

where the subscript of $\mathbf{v}$ represents the electron in layer 1 or 2, while the superscripts $>$ and $<$ refer to $v_z>0$ and $v_z<0$; $R(\mathbf{v}_1,\mathbf{v}_2)$ and $T(\mathbf{v}_1,\mathbf{v}_2)$ are reflection and transmission coefficients, $1-S(\mathbf{v}_1,\mathbf{v}_2)$ describes the diffuse scattering. The function F_1 and F_2 are defined below in Eqs. (6) and (7). All the coefficients S, T, and R are unchanged when one exchanges their argument $\mathbf{v}_1$ and $\mathbf{v}_2$ or when one reverses the sign of velocity, i.e., v_1 is replaced by $-v_1$, and v_2 is replaced by $-v_2$, due to microscopic reversibility. The velocity in layers 1 and 2 is related via the electron refraction law. For example, for a free electron with different potentials in layer 1 (U_1) and layer 2 (U_2), one has $mv_{1z}^2/2+U_1=mv_{2z}^2/2+U_2$, $T(\mathbf{v}_1,\mathbf{v}_2)=4|v_{1z}v_{2z}|/(v_{1z}+v_{2z})^2$, and $R=1-T$. The physical processes underlying Eq. (4) [and similarly Eq. (5)] is rather transparent: the outgoing particle flux $f(\mathbf{v}_2)$ comes from the reflected particle flux (the first term), the transmitted particle flux (the second term), and the particle flux from all directions diffusely scattered into $\mathbf{v}_2$ (last term).

There are two broad categories of interface roughness in lattice matched metallic multilayers, e.g., Co/Cu and Fe/Cr; they are depicted in Fig. 1. Figure 1(a) is a model for short range impurity scattering at the interface which neglects possible short range order inherent to the interface, e.g., pairwise interdiffusion.[17] In this model, an incoming electron ($\mathbf{v}_1^>$) is

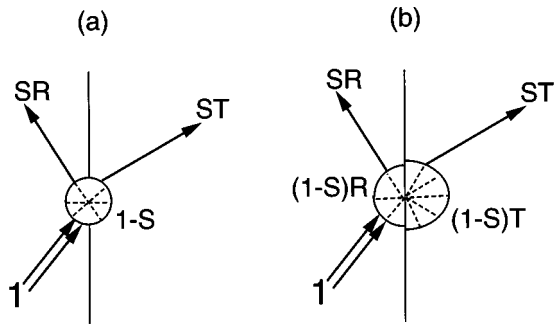

FIG. 1. Models for diffuse scattering. (a) For short range roughness isotropic impurity scattering. (b) For large-scale roughness.

scattered by an interface via reflection (SR, in the direction of $\mathbf{v}_1^<$), transmission (ST, in the direction of $\mathbf{v}_2^>$), and isotropic diffuse scattering (1-S, in all directions). Figure 1(b) is a model for large scale geometric roughness, in which the diffuse scattering is not isotropic in all directions; we discuss it later on. These two scattering processes are described by the following equations. For Fig. 1(a)

$$F_1^{(a)}=\frac{1}{\Omega}\int d^3\mathbf{k}'[|v'_{2z}|f(\mathbf{v}_2'^<)+|v'_{1z}|f(\mathbf{v}_1'^>)] \tag{6}$$

and $F_2^{(a)}=F_1^{(a)}$, where $\Omega\equiv\int d^3\mathbf{k}'|v'_z|$ is the normalization factor of the incoming particle flux to the interface. For Fig. 1(b), we have

$$F_1^{(b)}=\frac{1}{\Omega}\int d^3\mathbf{k}'[|v'_{2z}|R(\mathbf{v}_1',\mathbf{v}_2')f(\mathbf{v}_2'^<) +|v'_{1z}|T(\mathbf{v}_1',\mathbf{v}_2')f(\mathbf{v}_1'^>)] \tag{7}$$

and a similar expression $F_2^{(b)}$.

The key difference in the boundary conditions for the CIP and CPP geometries is that F_1 and F_2 in Eqs. (4) and (5) are absent in CIP because the distribution function is proportional to v_x (x is the direction of the electric field in the plane of the layers) and thus F_1 and F_2 are identically zero. For CPP, current conservation requires the presence of the F_1 and F_2. It is this boundary condition and the presence of the nonequilibrium dipole layer (as well as charge and spin accumulations) which make the CIP and CPP transport quite different.

The Boltzmann equation (3) along with the boundary conditions, Eqs. (4) and (5), determine interface resistance. Up to this point, our formulation is quite general and valid for arbitrary strengths of diffuse scattering; however, solutions of these equations are difficult to obtain for arbitrary $T(\mathbf{v})$ since electric fields (chemical potentials) and distribution function $g(\mathbf{v},z)$ must be determined self-consistently. Even without diffuse scattering, various approximations have to be made in order to solve the Boltzmann equation. In general, the self-consistent chemical potential $\mu(z)$ has two length scales: a rapid change occurs within a screening length from the interface; then there is a gradual change within a mean free path of the interface.[18–20] This gradual change was overlooked in an earlier study.[10] In Fig. 2, we illustrate this profile of the chemical potential; both $\Delta\mu$ and $\Delta\mu'$ contribute

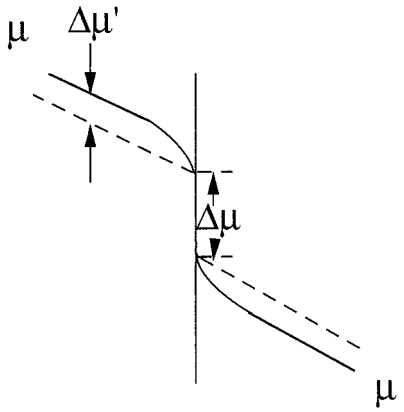

FIG. 2. The general chemical potential profile near an interface. $\Delta\mu$ is the voltage drop across the dipole layer (screen length) and $\Delta\mu'$ is the voltage drop across the diffusion layer due to detailed balancing with a length scale of the mean free path.

to the interface resistance $R_s=(\Delta\mu+2\Delta\mu')/j$ for identical layers. Without diffuse scattering ($S=1$), several groups[21,18,19] find approximate expressions for the interface resistance in terms of arbitrary transmission coefficient $T(\mathbf{v})$. In the presence of diffuse scattering one can solve the Boltzmann equation with the additional terms in the boundary conditions given by Eqs. (4) and (5) by a rather tedious self-consistent calculation. Instead of doing so in this paper, we examine a special case where an exact solution can be found. Our purpose is to demonstrate that diffuse and specular scattering are inseparable, i.e., one cannot treat them independently. For example, we show below, diffuse scattering can enhance or diminish conductance depending on the specular scattering. However, since we have not solved the boundary equations (4)–(7) for arbitrary reflection and transmission coefficients, which in general are functions of the velocities, but rather have resorted to the free electron approximation, the solutions obtained below should not be regarded as quantitative answers to these general cases. Rather these transparent solutions provide insight to what happens in more realistic situations. More importantly, we alert the reader that interface scattering problems are more complicated than Matthiessen's rule which envisages resistance as the sum of individual contributions from diffuse and specular scattering.

We choose a simple solution such that the long range variation of the chemical potential, $\Delta\mu'$ is *zero*, see Fig. 2, i.e., the electric field is constant outside a screening length from the interface. Then the distribution function is simply $g(\mathbf{v})=ev_z\tau E+e\mu(0^{\pm})$, where $\pm$ is for $z\gtrless 0$, where we assume an equal relaxation time and identical electronic structures for both layers[22] so that one can simply label $T(\mathbf{v}_1,\mathbf{v}_2)\equiv T(v_z)$, and similarly for S and R. By placing this trial function into Eq. (6), we find the diffuse scattering term is $F_1^{(a)}=[\mu(0+)+\mu(0-)]/2$. Then, from the boundary conditions, Eqs. (4) and (5), we arrive at

$$\mu(0^-)-\mu(0^+)=2v_z\tau E\frac{1+S(v_z)-2S(v_z)T(v_z)}{1-S(v_z)+2S(v_z)T(v_z)}. \quad (8)$$

As long as $\mu(0^-)-\mu(0^+)$ is independent of v_z, a constant electric field is a solution of this equation. For example, without diffuse scattering, i.e., $S(v_z)=1$, $T=|v_z|/(|v_z|+\alpha)$ satisfies this condition for any positive constant α.[6] If we continue to use $T=|v_z|/(|v_z|+\alpha)$ in the presence of the diffuse scattering, S must take the form

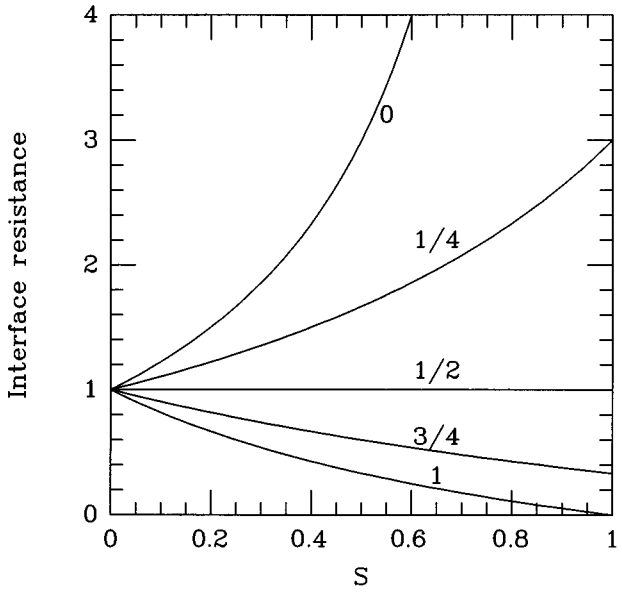

FIG. 3. Interface resistance R_s as a function of the diffuse scattering parameter for a set of fixed transmission coefficients $T_{k_F}=0$, 1/4, 1/2, 3/4, and 1; see Eq. (11). Note that $S_{k_F}=0$ represents total diffuse scattering, while for $S_{k_F}=1$ there is no diffuse scattering.

$$S(v_z)=\frac{\beta-|v_z|}{\beta+|v_z|}\frac{\alpha+|v_z|}{\alpha-|v_z|}, \quad (9)$$

where β is restricted to values such that $0\leqslant S\leqslant 1$. For these forms of $S(v_z)$ and $T(v_z)$ the interface resistance is

$$R_s\equiv\frac{\mu(0^-)-\mu(0^+)}{j}=2\beta\tau/\sigma_0, \quad (10)$$

where σ_0 is the conductivity of the bulk layers. One might immediately notice that Eq. (9) is different from the diffuse scattering found for the model of correlated roughness considered in Ref. 3. Our choice of the form Eq. (9) stems from our requirement of obtaining an exact solution of the Boltzmann equation for CPP.

While this solution gives us some insight into the interplay of specular and diffuse scattering in producing interface resistance, $S(v_z)$ and $T(v_z)$ vary between zero and one, as the component of the velocity perpendicular to the interface varies, and we cannot speak of cases where there is little or no diffuse or specular scattering. To confine $S(v_z)$ and $T(v_z)$ to a narrow range we consider a case where conduction is quasi one dimensional so that we are limited to electrons with k_z near k_F. Then interface resistance has a simple relation with S_{k_F} and T_{k_F},

$$R_s=2\frac{\tau k_F}{\sigma_0}\frac{1+S_{k_F}-2S_{k_F}T_{k_F}}{1-S_{k_F}+2S_{k_F}T_{k_F}}. \quad (11)$$

For $T_{k_F}=1$ (complete transmission), resistance increases as diffuse scattering increases (S_{k_F} decreases), while for $T_{k_F}=0$, resistance decreases as diffuse scattering increases. In Fig. 3, we show the resistance as a function of diffuse scattering parameter S_{k_F} for several fixed transmission coefficients. As seen from Fig. 3, diffuse scattering can *assist* conduction through interfaces. Although this conclusion was derived from this simple special case, Eq. (11), it holds for realistic specular and diffuse scattering coefficients $T(v_z)$ and $S(v_z)$ by the following argument. For small specular reflection coefficients (nearly perfect transmission) diffuse scattering is the sole source of interface resistance; for large specular reflection coefficients (nearly no transmission) diffuse scattering is the sole source of conduction because without diffuse scattering electrons cannot cross the interface.

For large scale geometric roughness interfaces are flat on the scale of atomic dimensions, and one has *locally* perfect reflections and transmissions. However, on a larger scale the locally flat regions are at an angle to one another, so that the specular reflections and transmissions from the locally flat regions are partially randomized by large scale roughness; this is depicted in Fig. 1(b). The diffuse scattering that comes from this process only redistributes the original specular reflection and transmission in such a way that the integrated backscattering is given by $\int|v_z|R(\mathbf{v})d^3\mathbf{k}$, and the forward transmission is given by $\int|v_z|T(\mathbf{v})d^3\mathbf{k}$. When $T(v_z)=0$ the interface resistance is infinite *regardless* of the diffuse scattering parameter $S(v_z)$ because all electrons are totally reflected by the interface, i.e., there is no diffuse scattering assisted conduction. While such diffusive scattering is certainly unacceptable for scattering from impurities it is applicable to large scale geometrical roughness.

In summary, we have considered interface resistance in the presence of both specular and diffuse scattering at interfaces. In the presence of diffuse scattering, new boundary conditions are required for the current perpendicular to the plane of the layers. These two sources of resistance, diffuse and specular scattering, are not additive, rather they are strongly coupled. Depending on specular reflection coefficients, diffuse scattering can either assist or suppress conduction. Our nonperturbative results alter the conclusions one derives from perturbative treatments which considered the effects of diffuse scattering in the presence of specular reflections, e.g., that diffuse scattering increases the resistance from an interface. For large reflection R (small T) this no longer holds; diffuse scattering *decreases* the resistance.

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[1] R. E. Camley and J. Barnas, Phys. Rev. Lett. **63**, 664 (1989).
[2] S. Zhang, P. M. Levy, and A. Fert, Phys. Rev. B **45**, 8689 (1992).
[3] R. Q. Hood and L. M. Falicov, Phys. Rev. B **46**, 8287 (1992); R. Q. Hood, L. M. Falicov, and D. R. Penn, *ibid.* **49**, 368 (1994).
[4] A. Vedyayev, B. Dieny, and N. Ryzhanova, Europhys. Lett. **19**, 329 (1992).
[5] T. Valet and A. Fert, J. Magn. Magn. Mater. **121**, 378 (1993); Phys. Rev. B **48**, 7099 (1993).
[6] M. Stiles (private communication).
[7] A. Vedyaev, C. Cowache, N. Ryzhanova, and B. Dieny, Phys. Lett. A **198**, 267 (1995).
[8] V. K. Dugaev, V. I. Litvinov, and P. P. Petrov, Phys. Rev. B **52**, 5306 (1995).
[9] J. Barnas and A. Fert, Phys. Rev. B **49**, 12 835 (1994); J. Magn. Magn. Mater. **136**, 260 (1994).
[10] V. V. Ustinov and E. A. Kravtsov, J. Phys.: Condens. Matter **7**, 3471 (1995).
[11] M. D. Stiles, J. Appl. Phys. **79**, 5805 (1996).
[12] W. H. Butler *et al.*, Phys. Rev. Lett. **76**, 3216 (1996).
[13] H. E. Camblong, S. Zhang, and P. M. Levy, Phys. Rev. B **47**, 4735 (1993).
[14] P. M. Levy and S. Zhang, J. Magn. Magn. Mater. **164**, 284 (1996).
[15] K. M. Schep, P. J. Kelly, and G. E. W. Bauer, Phys. Rev. Lett. **74**, 586 (1995).
[16] I. Mertig *et al.*, J. Magn. Magn. Mater. **151**, 363 (1995).
[17] S. Zhang and P. M. Levy, Phys. Rev. Lett. **77**, 916 (1996).
[18] B. Laikhtman and S. Luryi, Phys. Rev. B **49**, 17 177 (1994).
[19] C. Kunze, Phys. Rev. B **51**, 14 085 (1995).
[20] R. Landauer, Phys. Rev. B **52**, 11 225 (1995).
[21] R. S. Sorbello, Phys. Rev. B **39**, 4984 (1989).
[22] One can think of a situation where two identical metals are separated by a thin barrier, e.g., a small vacuum gap, so that the transmission coefficient can be varied from 0 to 1. Another example is the case where two identical metals are not perfectly stacked at the interface such that T differs from 1.