Enhanced Spin-Dependent Scattering at Interfaces

Shufeng Zhang and Peter M. Levy
Physics Department, New York University, 4 Washington Place, New York, New York 10003
(Received 21 February 1996)

Correlations in the scattering from the ordering of impurities due to interdiffusion between metals at interfaces are taken into account. The constructive interference between scattering amplitudes produces enhanced spin-dependent scattering from the interface between magnetic and nonmagnetic metals. [S0031-9007(96)00764-8]

PACS numbers: 73.40.–c, 73.50.Bk, 75.70.Pa

The giant magnetoresistance (GMR) observed in the transition-metal magnetic multilayers results from the change in the scattering of the conduction electrons as the magnetic configuration of the multilayer goes from one where the layers are on the average antiparallel in zero or coercive field to one where they are aligned in parallel at the saturation field. There is some debate as to the origin of the dependence of this scattering on the magnetic configuration [1]; however, it is clear that spin-dependent scattering is needed for GMR to occur, and that the scattering at interfaces plays a critical role in producing this effect [2].

For interfacial scattering to produce resistance it is necessary for it to be diffusive; this requires defects or roughness at the interface. For metals which are well lattice matched, most of the important roughness will be of two types: interdiffusional and geometrical; see Fig. 1. In this figure we show a pair of atoms that have exchanged their host positions [3]; rearrangements involving more atoms are often indistinguishable from short-range geometrical roughness. Geometrical roughness is more complicated, because this type of roughness has several length scales associated with it; its effect on conduction electron scattering has received some attention [4]. Notwithstanding other sources of scattering [5] we focus our attention on one source that has heretofore not been considered in either model or *ab initio* calculations [6] of the GMR: the *correlated* scattering from pairs of interdiffused atoms at interfaces.

In the bulk of the layers the impurities are not spatially correlated; e.g., they can exist as isolated impurities or as uncorrelated clusters. The scattering between randomly located impurities is phase incoherent, and one adds scattering probabilities to arrive at the total. However, at interfaces the scattering from a pair of interdiffused impurities is phase coherent; therefore one adds the scattering amplitudes before calculating the probability. In this latter case the resultant scattering can be greater or less than the sum of the individual ones due to quantum interference.

To illustrate the difference between scattering from uncorrelated and correlated impurities we develop multiple scattering theory for correlated impurities; for clarity we restrict ourselves to an ordered pair that is created at an interface. We show that for the same single-site scattering parameters correlated scattering from a pair of impurities produces scattering which is much more strongly spin dependent than the scattering from the individual impurities.

We choose as our Hamiltonian for a magnetic multilayer

$$H = H_c + V = H_c + \sum_i v_i , \tag{1}$$

where H_c is the Hamiltonian for any chosen coherent potential which may be complex, and v_i is the difference between the actual scattering potential v_α (if the site is occupied by the atom α) and the coherent potential Σ_i at i site. As we are discussing a multilayered periodic

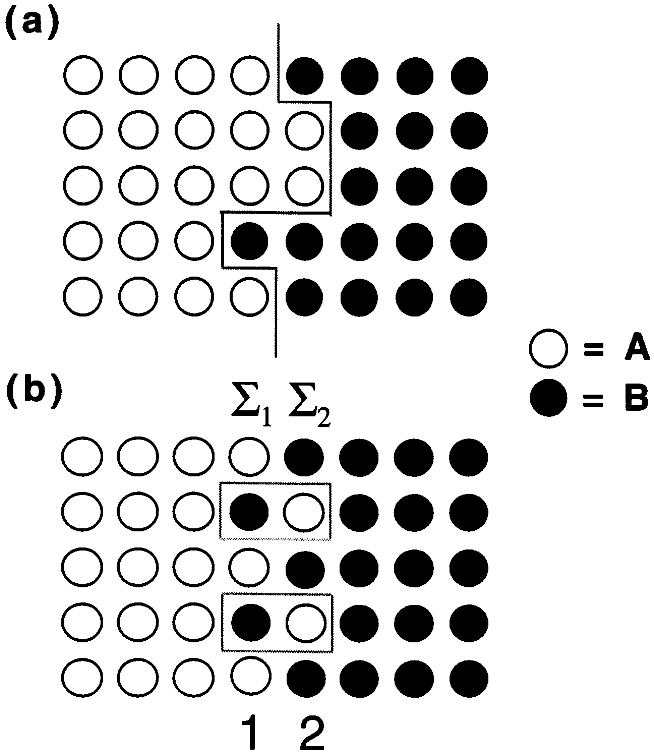

FIG. 1. Profiles of the interface due to geometrical roughness (a), and due to interdiffusion (b) (exchange of a pair of atoms). While there exists a clear interface for (a), it is not true for interdiffusion. The present calculation is focused on interdiffusional interfaces (b).

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structure, our coherent potential varies from one atomic layer to another within a unit cell [7]. The Green's function is written as

$$G = (\epsilon - H_c - V)^{-1} = G_0 + G_0 T G_0 , \qquad (2)$$

where $G_0 = (\epsilon - H_c)^{-1}$,

$$T = \sum_i v_i (1 + G_0 T) \equiv \sum_i T_i , \qquad (3)$$

$$T_i = t_i + t_i G_0 \sum_{j \neq i} T_j , \qquad (4)$$

and

$$t_i = (1 - v_i G_0)^{-1} v_i \qquad (5)$$

represent repeated scattering by the impurity at site i. Equation (4) is exact; it separates the total scattering at site i as a part coming from single-site scattering t_i and that from the surrounding $T_j (j \neq i)$. In the single-site approximation (SSA) one neglects correlations between scattering at different sites, so that the condition for the coherent potential in the coherent potential approximation (CPA) reduces to the requirement that the impurity averaged scattering t matrix at a site vanishes, $\langle t_i \rangle = 0$ [8].

To account for correlations in the scattering from a pair of impurity sites we regroup the T matrix into pairwise t matrices. From Eq. (4) this is written as

$$T_i = t_i + t_i G_0 T_j + t_i G_0 \sum_{m \neq i,j} T_m \qquad (6)$$

and

$$T_j = t_j + t_j G_0 T_i + t_j G_0 \sum_{m \neq i,j} T_m . \qquad (7)$$

By replacing Eq. (7) into Eq. (6), we arrive at

$$T_i = p_i^j + p_i^j G_0 \sum_{m \neq i,j} T_m , \qquad (8)$$

where

$$p_i^j = (1 - t_i G_0 t_j G_0)^{-1} t_i (1 + G_0 t_j) \qquad (9)$$

represents repeated scattering at sites i and j. The structure of Eq. (8) is similar to Eq. (4); one replaces the single-site t matrices by the pair matrices p_i^j.

With our classification of the T_i, Eq. (8), it is possible to keep correlations between scattering impurities at a pair of sites and neglect those between this pair and other sites, by uncoupling the impurity averages of the T matrix, Eq. (8), as follows,

$$\langle T \rangle = \sum_i \langle T_i \rangle = \sum_i \langle p_i^j \rangle + \sum_i \langle p_i^j \rangle G_0 \sum_{m \neq i,j} \langle T_m \rangle . \qquad (10)$$

Since all pairs at interfaces are the same after impurity average, the CPA condition ($\langle T \rangle = 0$) for determining the coherent potential within the approximation of pairwise ordering is

$$\langle p_i^j \rangle = 0 . \qquad (11)$$

For comparison, in the SSA it is $\langle t_i \rangle = 0$.

In magnetic multilayers, impurity scattering occurs in the bulk and at the interfaces. The CPA treatment for the scattering in the bulk is the same as the alloy problem, i.e., by using the uncorrelated single-site approximation, $\langle t_i \rangle = 0$ when the site i is not at an interface. For correlated pairs at interfaces, we use our pairwise CPA $\langle p_i^j \rangle = 0$. When solved self-consistently these equations determine the coherent potentials at each site within the unit cell of the superlattice; compared to the homogeneous alloy CPA, they are considerably more complicated to solve because we have N coupled equations (N is number of monolayers in the unit cell). Since the main purpose of this Letter is to show the critical role of short-range ordering at interfaces in producing spin-dependent scattering, we focus our attention purely on interface scattering. For simplicity, we first consider a simple cubic structure; then we outline our results for the bcc structure.

Let us designate two metals A and B in contact, sharing a common [100] interface with monolayers 1 and 2 nearest to the interface, see Fig. 1(b). When atom A with potential v_a occupies monolayer 2, atom B is in layer 1 directly to the left of A; i.e., we limit ourselves to nearest neighbor diffusion. We introduce the site-diagonal coherent potential $\hat{\Sigma}$ for a layered structure [9]

$$\hat{\Sigma} = |1\rangle \Sigma_1 \langle 1| + |2\rangle \Sigma_2 \langle 2| + \sum_{m \neq 1,2}^{N} |m\rangle v_m \langle m| , \qquad (12)$$

where we have only included the disorder of the two layers adjacent to the interface, and V_m is a potential at site m which is not at the interface. With this coherent potential, the potential at site i ($i = 1, 2$) is either $v_a - \Sigma_i$ or $v_b - \Sigma_i$ depending whether the site i is occupied by the A or B atom. The Green's function for our chosen coherent potential is $G_0 = (\epsilon - H_0 - \hat{\Sigma})^{-1}$. Explicitly, we can write down the CPA condition, Eq. (11), as

$$(1 - c) \frac{(v_a - \Sigma_1) - (v_a - \Sigma_1)(v_b - \Sigma_2)(G_{22} - G_{12})}{1 - (v_a - \Sigma_1) G_{11} - (v_b - \Sigma_2) G_{22} + (v_a - \Sigma_1)(v_b - \Sigma_2)(G_{11} G_{22} - G_{12} G_{21})}$$
$$+ c \frac{(v_b - \Sigma_1) - (v_b - \Sigma_1)(v_a - \Sigma_2)(G_{22} - G_{12})}{1 - (v_b - \Sigma_1) G_{11} - (v_a - \Sigma_2) G_{22} + (v_b - \Sigma_1)(v_a - \Sigma_2)(G_{11} G_{22} - G_{12} G_{21})} = 0 \qquad (13)$$

and a similar equation by exchanging the labels 1 and 2 of the above equation, where $G_{ij} = \langle i|(\epsilon - H_0 - \Sigma)^{-1}|j\rangle$, and c is the concentration (probability) that atom B is found in layer 1. These are the equations that must be solved self-consistently to determine the self-energy (coherent potentials) Σ_1 and Σ_2.

The key difference entering the pairwise CPA conditions for the coherent potentials, Eq. (13), is the site off-diagonal Green's functions, which are absent in the single-site CPA condition. These propagators keep track of the correlations in the scattering from the ordered impurities that may exist at interfaces; the SSA makes the unrealistic assumption that impurities on the two sides of an interface are uncorrelated. At the present time there is no direct evidence for ordering of interdiffused impurities; however, its existence explains the enhanced spin dependence of the interface scattering that has been observed [2].

To demonstrate the importance of short-range order on the resistivity and magnetoresistance, we examine the limit where both the concentration c and the relative scattering potential $v_a - v_b$ are small, i.e., dilute and weak scattering limit. We linearize Eq. (13) with respect to c, and only keep terms up to second order in the potentials. We find the simple expression for the self-energies (coherent potentials),

$$\Sigma_1 = v_a + c\{v_b - v_a + (v_a - v_b)^2(G_{11}^0 - G_{12}^0)\} \tag{14}$$

and

$$\Sigma_2 = v_b + c\{v_a - v_b + (v_b - v_a)^2(G_{22}^0 - G_{21}^0)\}, \tag{15}$$

where $G_{ij}^0 = \langle i|(\epsilon - H_0)^{-1}|j\rangle$ is the matrix element of the Green's function for the perfect superlattice. The local relaxation time, which is given by the inverse of the imaginary part of the self-energy, is

$$1/\tau_1 = -\mathrm{Im}\Sigma_1 = c(v_a - v_b)^2[-\mathrm{Im}(G_{11}^0 - G_{12}^0)] \tag{16}$$

and a similar expression holds for $1/\tau_2$. For comparison the self-energy without taking account of the short-range order of the impurities at the interfaces is

$$1/\tau_i = c(v_a - v_b)^2(-\mathrm{Im}G_{ii}^0) = c(v_a - v_b)^2\pi\rho(\epsilon) \tag{17}$$

for $i = 1, 2$, where $\rho(\epsilon)$ is the density of states at the interface.

As Eqs. (16) and (17) hold for each direction of spin the ratio of scattering rates for up spins to down spins at an interface, due to correlated scattering, is from Eq. (16)

$$\alpha_{\mathrm{int}} = \frac{(v_a^\uparrow - v_b^\uparrow)^2[-\mathrm{Im}(G_{11}^{0\uparrow} - G_{12}^{0\uparrow})]}{(v_a^\downarrow - v_b^\downarrow)^2[-\mathrm{Im}(G_{11}^{0\downarrow} - G_{12}^{0\downarrow})]}, \tag{18}$$

while that from *uncorrelated* scattering at the interface or in the bulk is from Eq. (17)

$$\alpha_{\mathrm{bulk}} = \frac{(v_a^\uparrow - v_b^\uparrow)^2\rho^\uparrow(\epsilon_F)}{(v_a^\downarrow - v_b^\downarrow)^2\rho^\downarrow(\epsilon_F)}. \tag{19}$$

The enhancement from correlated scattering at the interface is the ratio of Eqs. (18) and (19), in which the factors $(v_a^\sigma - v_b^\sigma)^2$ cancel.

To quantitatively determine the enhancement of the spin-dependent scattering at interfaces, we model the interfacial region by spin-polarized tight binding bands. For a simple cubic lattice we use

$$\epsilon_{\mathbf{k}}^\sigma = \epsilon^\sigma - t[\cos(k_x a_0) + \cos(k_y a_0) + \cos(k_z a_0)], \tag{20}$$

where a_0 is the lattice constant, and σ is the spin index. The spin dependence of the band is described by a constant exchange potential $\epsilon^\uparrow - \epsilon^\downarrow$. Taking the bandwidth t to be one, we show in Fig. 2 the spin-dependent scattering ratios at interfaces with, Eq. (18), and without, Eq. (19), short-range correlations. The spin dependence of scattering from correlated pairs at interfaces is indeed enhanced compared to scattering in the bulk. This provides one explanation for the observation [2] that interface scattering is more spin dependent than the scattering in the bulk. The enhancement of the spin dependence of interface scattering will be in proportion to the fraction of scattering at interfaces coming from interdiffused pairs of atoms.

We have also carried out similar calculations for bcc and fcc structures. For a [100] bcc interface, an impurity can diffuse to four possible nearest neighbors; therefore, we need to extend our formulation to include four pairs for each site at an interface. Following our decoupling procedure of impurity averaging, we regroup the T matrix

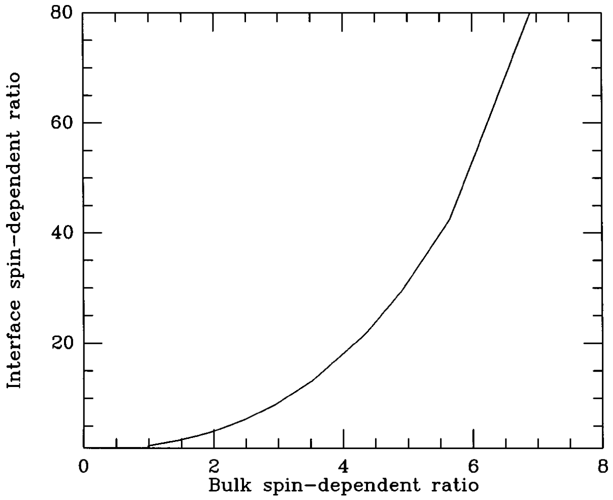

FIG. 2. The ratio of the spin-dependent scattering rates at the interface compared to that in the bulk from Eqs. (16) and (17) for the [001] interface of a simple cubic lattice. For simplicity we have assumed that the spin dependence of the scattering from the bulk and from interfaces *without* correlations is the same. Note that the ratio at the interface is one when the bulk is one.

as follows:

$$T_0 = t_0 + t_0 G_0 \sum_{\alpha=1}^{4} T_\alpha + t_0 G_0 \sum_{\beta \neq 0,\ldots,4} T_\beta \,, \tag{21}$$

where the label "0" in the t matrix represents a site at the monolayer 1 with nearest neighbors "1–4" at the monolayer 2. Similar equations for T_α can also be rearranged. In the dilute and weak scattering limit we can discard the last term in Eq. (21), and we find

$$1/\tau_0 = c(\upsilon_a - \upsilon_b)^2 \left[-\mathrm{Im}\left(G_{00}^0 - \sum_{\alpha=1}^{4} G_{0\alpha}^0/4 \right) \right], \tag{22}$$

where again we have limited ourselves to scattering from nearest neighbors. By using the tight binding band for a bcc lattice $\epsilon_{\mathbf{k}}^{\sigma} = \epsilon^{\sigma} - t\cos(k_x b_0)\cos(k_y b_0)\cos(k_z b_0)$ where $b_0 = (\sqrt{2}/2)a_0$, we are able to reach the same conclusion, i.e., the spin dependence of scattering is enhanced by the correlated scattering at interfaces from pairs of interdiffused impurities. For example, if one uses the same band parameters, the enhancement will be *four* for bcc structures if that for the simple cubic is *ten.* This is indeed quite significant. For the [100] fcc interface the four nearest neighbors are rotated by 45° with respect to the bcc lattice; from the consideration below we can anticipate the enhancement to be less than that for simple cubic lattices and the same order as the bcc quoted above.

While quantitative calculations for realistic multilayers are much more complicated than our tight binding band approximation, our simple model in the limit of dilute and weak scattering provides physical insights of the effect of the short-range order at interfaces. The essential ingredient in Eqs. (16) and (22) is the appearance of interference in the scattering through the Green's functions. If we limit ourselves to M nearest neighbors at an interface, the interference term is

$$\frac{1}{M} \sum_{\alpha}^{M} G_{0\alpha} = \frac{1}{M} \sum_{\alpha}^{M} \sum_{\mathbf{k}} \frac{\exp(i\mathbf{k} \cdot \mathbf{r}_{0\alpha})}{\epsilon - \epsilon_{\mathbf{k}} + i\delta} \,, \tag{23}$$

where $\mathbf{r}_{0\alpha}$ is the vector connecting two correlated sites. When the diffusion process happens beyond nearest neighbors (bad multilayers), the effect of interference is much smaller (for example, if the diffusion happens for the third nearest neighbors, the interference effect is practically zero) since the average of $\exp(i\mathbf{k} \cdot \mathbf{r}_{12})$ approaches zero, and one recovers the single-site CPA approximation. The calculations for various crystallographic orientations of the multilayered growth will be presented elsewhere.

In summary, we have accounted for correlations in the scattering from a pair of impurity sites at an interface by replacing the single-site CPA condition $\langle t_i \rangle = 0$ by the condition $\langle p_i^j \rangle = 0$, and have developed a multiple scattering theory to explicitly take into account this short-range order. The new CPA algorithm goes beyond single-site CPA, and we have demonstrated that it accounts for an enhanced spin dependence of the interface scattering. As structural evidence for short-range ordering of impurities at interfaces becomes available, *ab initio* calculations of the transport properties of metallic multilayers should take into account these important correlations.

This research was supported by the Office of Naval Research under Grant No. N00014-96-1-0203.

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[1] That is, whether the spin dependence of the scattering is primarily due to the band structure of the superlattice and thus to the different density of states at the Fermi level for the majority and minority conduction electrons, or whether the spin dependence is intrinsic to the scattering potential itself.

[2] S. S. P. Parkin, Phys. Rev. Lett. **71**, 1641 (1993); B. A. Gurney *et al.,* IEEE Tran. Magn. **26**, 2747 (1990); P. Baumgart *et al.,* J. Appl. Phys. **69**, 4792 (1991). From fits to GMR data on magnetic multilayers one arrives at two conclusions: that the strength and that the spin dependence of the scattering at the interfaces between layers are stronger than in the bulk of the layers. That the scattering is larger at interfaces is not surprising; after all, there is a rapid change in the electronic structure (potential step) in the interfacial region; the layers are not flat so that there is geometrical roughness and elements from one layer diffuse into the neighboring layer; see Fig. 1. Each of these aspects has received some attention; the "band mismatch" between the dissimilar metals constituting the interface is a crude indicator of the strength of the scattering induced by roughness (geometrical and interdiffusional). Geometrical roughness produces an angular dependence to the scattering that is nominally isotropic in the layers; i.e., the interfacial scattering is stronger for electron trajectories perpendicular to the plane of the layers; see, for example, Ref. [4]. Also, interfaces have inordinately high concentrations of impurities relative to the bulk of the layers, so that it is natural that the scattering is stronger than in the bulk of the layers.

[3] One possibility for interdiffusion is for atoms in adjoining layers to fill the vacancies left by the diffusing atom from the host layer.

[4] R. Q. Hood, L. M. Falicov, and D. R. Penn, Phys. Rev. B **49**, 368 (1994).

[5] There are sources of scattering in the multilayers that are difficult to model and to take into account, e.g., extended defects such as stacking faults in the layers, and the magnetization at the interfaces is hardly collinear, i.e., while magnetic moments in the bulk of a layer are collinear, the competing forces at the interface do not guarantee collinearity there. W. H. Butler (private communication).

[6] W. H. Butler, J. M. MacLaren, and X. G. Zhang, Mater. Res. Soc. Symp. Proc. **313**, 59 (1993); W. H. Butler, X. G. Zhang, and D. M. C. Nicholson, J. Appl. Phys. **76**, 6808 (1994).

[7] J. M. MacLaren *et al.,* Phys. Rev. B **40**, 12 164 (1989); **40**, 12 176 (1989).

[8] H. Ehrenreich and L. M. Schwartz, Solid State Phys. **31**, 149 (1976).

[9] H. Itoh, J. Inoue, and S. Maekawa, Phys. Rev. B **51**, 342 (1995).