Theory of tunneling magnetoresistance in a junction with a nonmagnetic metallic interlayer

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(Received 23 February 1999)

It is demonstrated by numerical evaluation of the real-space Kubo formula that the tunneling magnetoresistance (TMR) due to tunneling between two cobalt electrodes separated by a vacuum gap remains nonzero when one of the electrodes is covered with a copper layer. This contradicts the classical theory of tunneling that predicts zero TMR. It is shown that a nonzero TMR is due to quantum well states in the Cu layer that do not participate in transport. Since these only occur in the down-spin channel, their loss from transport creates a spin asymmetry of electrons tunneling from a Cu overlayer, i.e., nonzero TMR. This mechanism could provide an explanation of the observed nonzero TMR for junctions with Cu or Ag interlayers. A simple method for modifying the classical theory of tunneling so that it can describe correctly tunneling in the presence of quantum well states is proposed and implemented for the Co junction with a Cu interlayer.
[S0163-1829(99)10525-3]

The conductance $\Gamma(H_s)$ of a tunnel junction with two ferromagnetic electrodes whose magnetic moments are aligned parallel in an applied saturating field H_s is much higher than its conductance $\Gamma(0)$ in zero field when the moments are antiparallel.[1–3] The effect is called tunneling magnetoresistance (TMR) and the relative change in the resistance of the junction, i.e., the so-called optimistic magnetoresistance ratio

$$R_{TMR}=\frac{\Gamma(0)^{-1}-\Gamma(H_s)^{-1}}{\Gamma(H_s)^{-1}} \tag{1}$$

can be as high as 40%. The traditional explanation of the TMR effect is based on the assumption that electrons tunneling from a ferromagnet are spin polarized and their polarization P is given in terms of the spin-dependent density of states D^{σ} of the ferromagnet by $P=[D^{\uparrow}(E_F)-D^{\downarrow}(E_F)]/[D^{\uparrow}(E_F)+D^{\downarrow}(E_F)]$. Since the classical theory of tunneling[4] states that the junction conductance is proportional to the product of the densities of states of the left and right electrodes, it is easy to show that the TMR ratio (1) can be written in terms of the spin polarizations P_L, P_R of the left and right electrodes,

$$R_{TMR}=\frac{2P_LP_R}{1-P_LP_R}. \tag{2}$$

This is the well-known Julliere formula[5] that is remarkably successful in predicting the TMR ratio from the observed values[4] of the spin polarization of electrons tunneling from Fe, Ni, and Co into a superconductor.

However, when the Julliere formula is applied to a tunneling junction with a thin nonmagnetic metallic interlayer, such as Cu or Ag, inserted between one of the ferromagnetic electrodes and the insulating barrier, it fails to explain the observed[6] nonzero TMR ratio. In fact, since the density of states of the Cu layer adjacent to the barrier is spin independent, $P_{\mathrm{Cu}}=0$, it follows from Eq. (2) that $R_{TMR}=0$, which contradicts the experiment.[6] The failure of the Julliere formula calls into question the validity of the whole classical theory of tunneling. To resolve this problem, we shall first demonstrate in the case of coherent tunneling that rigorous quantum theory of transport based on the real-space Kubo formula,[7,8] and a realistic band structure gives a nonzero TMR for the junction with a nonmagnetic metallic interlayer. We then examine the derivation of the Julliere formula from the Kubo formula to identify the physical reasons for its failure in the case of a junction with a nonmagnetic interlayer. Finally, using a concept of the transport density of states, we propose how the Julliere formula should be generalized to describe correctly tunneling in the presence of a nonmagnetic interlayer.

The first theoretical demonstration of a nonzero TMR in a junction with a nonmagnetic interlayer was given by Vedyaev *et al.*[9] for a single band model. Zhang and Levy[10] reviewed their argument using Slonczewski's model of tunneling[11] (simple parabolic band). They concluded that loss of coherence between the scattering from the ferromagnet/nonmagnet and nonmagnet/barrier interfaces destroys TMR. Zhang and Levy[10] also argued that quantum well states arising from the insertion of a nonmagnetic layer are detrimental to TMR. In contrast to their results, obtained for the Slonczewski model, our more general approach based on the Kubo formula and a realistic band structure shows that quantum well states in the nonmagnetic interlayer are, in fact, a necessary ingredient for nonzero TMR. We shall also demonstrate that in the presence of quantum well states loss of coherence does not necessarily destroy TMR.

To obtain a clear-cut answer to the question whether there is a nonzero TMR for a tunneling junction with a nonmagnetic interlayer, one needs to treat realistically the band structure of the junction. We, therefore, investigate tunneling from a cobalt electrode covered with an overlayer of N atomic planes of Cu across a vacuum gap into another cobalt electrode (Fig. 1). It is assumed that the electrodes are perfect so that the electron wave vector parallel to the layers $\vec{k}_{\parallel}$ is conserved in tunneling. This restriction will be relaxed later. In contrast to the junction with an amorphous Al_2O_3 barrier used in the experiment, tunneling across a vacuum gap considered here has the great advantage that the real-

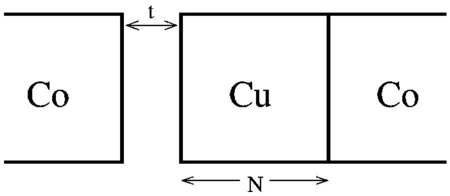

FIG. 1. Schematic representation of a Co/vacuum/Cu/Co tunneling junction, with hopping matrix **t** across the vacuum gap.

space Kubo formula[7,8] can be evaluated without any approximations for a fully realistic band structure of all the components of the junction.

We use a tight-binding parametrization of an *ab initio* band structure of fcc Co and Cu (for details, see Ref. 8). The total conductance of the junction Γ^σ in a spin channel σ is expressed[12] in terms of the one-electron Green's functions $\mathbf{G}_L^\sigma(E_F,\vec{k}_\parallel)$, $\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel)$ at the surface of the left and right electrodes,

$$\Gamma^\sigma=\frac{4e^2}{h}\sum_{\vec{k}_\parallel}\ \mathrm{Tr}\{[\mathbf{T}_\sigma\ \mathrm{Im}\,\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel)]\times[\mathbf{T}_\sigma^\dagger\,\mathrm{Im}\,\mathbf{G}_L^\sigma(E_F,\vec{k}_\parallel)]\}. \quad (3)$$

We recall that the left electrode is a semi-infinite Co slab and the right electrode is a semi-infinite Co slab covered with N atomic planes of Cu. The summation in Eq. (3) is over the two-dimensional Brillouin zone and the trace is over the orbital indices corresponding to s,p,d orbitals that are required in a tight-binding parametrization of Co and Cu. Since we use a multiorbital band structure, then **G** and **T** are matrices whose size depends on the number of orbitals. The matrix $\mathbf{T}_\sigma$ is given by

$$\mathbf{T}_\sigma=\mathbf{t}(\vec{k}_\parallel)[\mathbf{I}-\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel)\mathbf{t}^\dagger(\vec{k}_\parallel)\mathbf{G}_L^\sigma(E_F,\vec{k}_\parallel)\mathbf{t}(\vec{k}_\parallel)]^{-1}, \quad (4)$$

where **I** is a unit matrix in the orbital space and $\mathbf{t}(\vec{k}_\parallel)$ is the matrix of tight-binding hopping integrals connecting across vacuum gap atomic orbitals in the surface of the Cu overlayer on the right Co electrode to atomic orbitals in the surface of the left Co electrode. This is schematically depicted in Fig. 1. Following Refs. 12 and 13 we model tunneling across vacuum gap by turning off gradually the hopping matrix $\mathbf{t}(\vec{k}_\parallel)$ across the gap. As discussed in Ref. 12, tunneling between d orbitals is suppressed owing to their weak overlap across the gap. In the case of an Al_2O_3 barrier, suppression of d-type tunneling is due to the fact that there are no d orbitals present in the barrier. It follows that hopping between s-p orbitals should dominate the tunneling current. We, therefore, include in the tunneling matrix $\mathbf{t}(\vec{k}_\parallel)$ only s-p hopping matrix elements and their values are obtained by scaling down uniformly the bulk tight-binding hopping matrix between Co and Cu. It was shown in Ref. 12 that TMR becomes very quickly independent of the actual magnitude of the hopping matrix elements in the tunneling regime. We, therefore, scaled in our calculations the s-p hopping matrix elements to 10% of their bulk values, which is more than sufficient to reach the tunneling regime.

The dependence of the TMR ratio obtained by numerical evaluation of the Kubo formula (3) on the thickness of the Cu overlayer is shown in Fig. 2 (continuous line). The calculation is for (111) orientation of the layers.

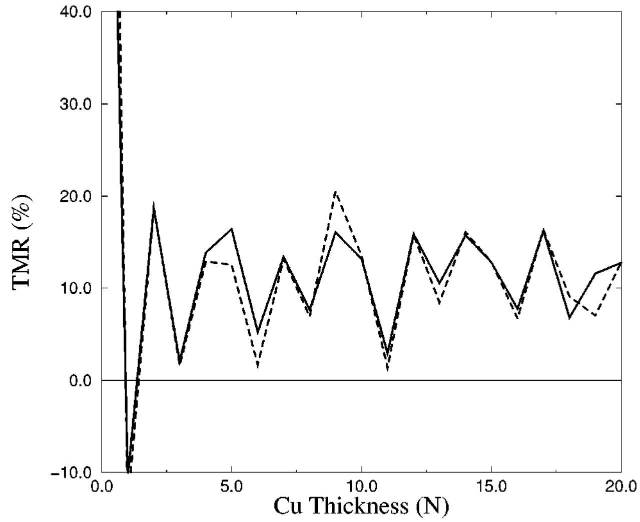

FIG. 2. TMR (%) as a function of Cu overlayer thickness. The continuous line represents the TMR evaluated by the full Kubo formula. The broken line represents the TMR evaluated by the linearized Kubo formula.

In contrast to the Julliere formula (2), the TMR determined from the Kubo formula (3) is nonzero and oscillates as a function of Cu thickness due to quantum interference of electrons on the Cu interlayer. It is interesting that for a small Cu thickness (1 ML) the TMR ratio becomes negative. A negative TMR with a very thin gold interlayer has been observed by Moodera.[14]

The physical explanation of a nonzero TMR is that the Cu layer acts as a spin filter. Since the Fermi surfaces of Cu and of the majority-spin electrons in Co are very similar (the Co majority d band lies below E_F), majority-spin electrons easily cross the Co/Cu interface and participate in tunneling as if there were no intervening Cu layer. On the other hand, there is a poor match between the Cu bands and the minority-spin bands in Co, which results in formation of down-spin quantum well states in the Cu overlayer.[15,16] Since the quantum well states are localized in the Cu layer they do not contribute to transport of charge in the down-spin channel, which gives rise to a spin asymmetry (nonzero polarization P) of the tunneling current and, hence, nonzero TMR. It should be noted that resonances, i.e., virtual bound states, have the same spin-filtering effect in the case of coherent tunneling. However, it will be seen that true bound states are required for the spin-filtering effect to persist in the case of noncoherent tunneling.

The apparent paradox that the Julliere formula predicts zero TMR but the Kubo formula gives a nonzero TMR can now be easily resolved. Since the down-spin quantum well states in the Cu layer contribute to the ordinary density of states (DOS) they are, incorrectly, counted in the Julliere formula (2) as contributing to the tunneling current. The total DOS of down-spin electrons, which is made up of propagating and quantum well states, is equal to the DOS of up-spin states that are all propagating. There is, therefore, no spin asymmetry in the DOS of the Cu overlayer and, hence, the Julliere formula gives zero TMR. On the other hand, the Kubo formula excludes automatically all the quantum well states. Since these only occur in the down-spin channel, their loss from transport creates a spin asymmetry of electrons tunneling from a Cu overlayer, i.e., nonzero TMR.

To pinpoint the reason why the classical theory of tunneling fails in the presence of quantum well states, we need to examine the approximations that are made in its derivation from the Kubo formula. There are three approximations involved.

(i) Given that $\mathbf{t}\approx 0$ (electron hopping between the electrodes is weak), it is assumed that the Kubo formula can be linearized, i.e., $\mathbf{T}_\sigma = \mathbf{t}(\mathbf{I}-\mathbf{G}_R^\sigma \mathbf{t}^\dagger \mathbf{G}_L^\sigma \mathbf{t})^{-1} \rightarrow \mathbf{t}$.

(ii) Only tunneling between the same orbitals is considered and assumed to be equally probable, i.e., the hopping matrix $\mathbf{t}$ is replaced by $t_0\mathbf{I}$, where $\mathbf{I}$ is a unit matrix in the orbital space and t_0 is a single tunneling matrix element independent of $\vec{k}_\parallel$.

(iii) Complete loss of coherence across the barrier (vacuum gap) is imposed, i.e., it is assumed that a state $\vec{k}_\parallel$ tunnels with an equal probability to any other state $\vec{k}_\parallel'$.

With these approximations, the Kubo formula (3) takes the form

$$\Gamma^\sigma = \frac{4e^2}{hN_\parallel}|t_0|^2 \left[\sum_{\vec{k}_\parallel} \mathrm{Tr\,Im}\,\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel) \right] \times \left[\sum_{\vec{k}_\parallel'} \mathrm{Tr\,Im}\,\mathbf{G}_L^\sigma(E_F,\vec{k}_\parallel') \right], \tag{5}$$

where $N_\parallel$ is the number of atoms in the plane of the junction. Since the expressions in the brackets are (up to a factor $1/\pi$) the total densities of states of the right and left electrodes, Eq. (5) reduces to the usual expression for the conductance obtained in the classical theory of tunneling.[4]

It is easy to see that the linearization [approximation (i)] of the Kubo formula is the reason for the failure of the classical theory of tunneling (Julliere formula) in the presence of quantum well states. It is crucial to preserve the full $\mathbf{T}$ matrix defined by Eq. (4) in the Kubo formula since it contains complete information about the interaction of the right and left electrodes (to all orders in $\mathbf{t}$). This is needed to remove the contribution of quantum well states to the conductance.

Approximation (ii) is responsible for the incorrect sign of the spin polarization P predicted by Eq. (5) for Fe, Co, and Ni. This is because formula (5) based on the total DOS allocates equal weights to tunneling via d states and s-p states. In reality, tunneling via s-p states dominates.[17]

Approximation (iii) is useful since it provides the simplest way of dealing with loss of coherence in tunneling (nonconservation of $\vec{k}_\parallel$), which almost certainly occurs in tunneling through an amorphous Al_2O_3 barrier.

Based on this analysis, it is clear how the Julliere formula should be generalized so that it describes correctly tunneling in the presence of quantum well states and, at the same time, gives the correct sign of the tunneling current. We first address the linearization problem. One expects that the linearization should be a very good approximation in the absence of bound (quantum well) states. On the other hand, we know from the Kubo formula that bound states do not contribute to the conductance. We, therefore, propose to correct linearization (i) by explicitly removing all the bound states. This can be done by the following simple device. For each tunneling state $\vec{k}_\parallel$, we test whether it is propagating in the electrodes at $\pm\infty$. If it is, its contribution is included in the conductance, otherwise it is excluded. The accuracy of the linearized Kubo formula with all the quantum well states manually removed can be tested by comparing it with the full Kubo formula. The TMR ratio computed from the linearized Kubo formula (broken line) is compared in Fig. 2 with the TMR ratio determined from the full Kubo formula (solid line). It can be seen that, for a scaling of the vacuum matrix elements to 10% of their metalic values, the linearization gives a very good approximation to the calculation based on the full Kubo formula. Since the TMR calculated from the linearized formula is independent of the scaling of the vacuum hopping matrix [see Eq. (1)], the linearized formula for TMR is exact in the limit $\mathbf{t}\rightarrow 0$.

We now consider the second and third approximations made in the Julliere formula. As already discussed, the second approximation cannot be made since it leads to an incorrect sign of the tunneling current. It is, therefore, necessary to keep the full matrix $\mathbf{t}(\vec{k}_\parallel)$ of hopping integrals across the vacuum gap. The elements of the hopping matrix are again scaled down as described earlier for the case of coherent tunneling.

Finally, we need to model the loss of coherence in tunneling due to disorder. The effect of disorder on tunneling has previously been studied within a single orbital model using the coherant potential approximation and direct numerical evaluation of the Kubo formula for small clusters.[18,19] Such a study would be difficult for a realistic band structure. However, the work on single orbital models shows that the main effect of disorder at the electrode/vacuum interface is the loss of conservation of in-plane momentum $\vec{k}_\parallel$. To model the loss of conservation of $\vec{k}_\parallel$ phenomenologically, and to make contact with Julliere's formula we wish to reduce the Kubo formula into a product of two factors characterizing the left and right electrodes, i.e., bring it to the form of Eq. (5). We first need to generalize the linearized Kubo formula [deduced from Eq. (3)] to allow for loss of coherence across the vacuum gap. We assume that the electrodes remain translationally invariant in the direction parallel to the layers and, therefore, the surface Green's functions of the disconnected electrodes are diagonal. However, tunneling from any state $\vec{k}_\parallel$ in the left electrode to any other state $\vec{k}_\parallel'$ in the right electrode is now allowed to model disorder in the gap between the electrodes. That means that the hopping matrix across the vacuum gap becomes nondiagonal, i.e., $\mathbf{t}(\vec{k}_\parallel,\vec{k}_\parallel')$. The Kubo formula for such incoherent tunneling takes the form

$$\Gamma^\sigma = \frac{4e^2}{hN_\parallel} {\sum_{\vec{k}_\parallel}}' {\sum_{\vec{k}_\parallel'}}' \mathrm{Tr}[\mathbf{t}(\vec{k}_\parallel,\vec{k}_\parallel')\,\mathrm{Im}\,\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel')] \times [\mathbf{t}^\dagger(\vec{k}_\parallel',\vec{k}_\parallel)\,\mathrm{Im}\,\mathbf{G}_L^\sigma(E_F,\vec{k}_\parallel)], \tag{6}$$

where the prime indicates that all the quantum well states have been omitted. The assumed complete loss of coherence implies that tunneling from any state $\vec{k}_\parallel$ to any other state $\vec{k}_\parallel'$ is equally probable. It follows that $\mathbf{t}(\vec{k}_\parallel,\vec{k}_\parallel')$ is a constant

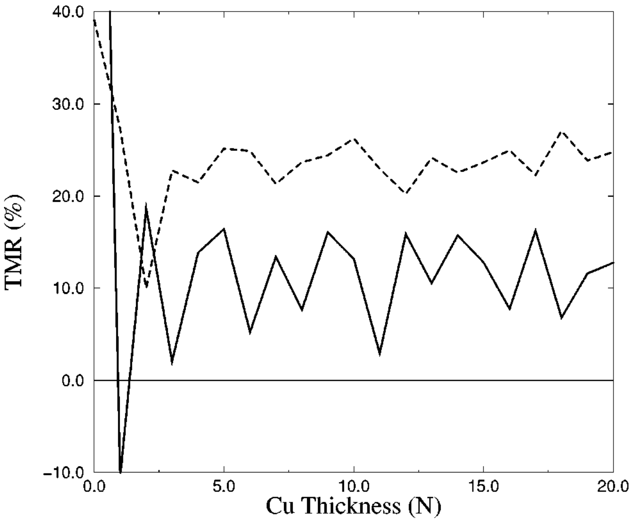

FIG. 3. TMR (%) as a function of Cu overlayer thickness. The continuous line represents the TMR evaluated by the full Kubo formula for coherent tunneling. The broken line represents the TMR evaluated by the linearized Kubo formula for incoherent tunneling.

matrix, independent of $\vec{k}_\parallel,\vec{k}'_\parallel$. We may approximate this constant matrix by $\mathbf{t}(\vec{k}_\parallel,\vec{k}'_\parallel)\approx\mathbf{t}(0)$, where $\mathbf{t}(0)$ is the value of the diagonal hopping matrix element for $\vec{k}_\parallel=0$. It is reasonable to make such an approximation since the perpendicular tunneling with $\vec{k}_\parallel=0$ is expected to dominate. It is now possible to define a transport density of states $D_R^\sigma(E_F)$ for the right electrode by

$$D_R^\sigma(E_F)=\mathbf{t}(0)\sum_{\vec{k}_\parallel}{}' \; \mathrm{Im}\,\mathbf{G}_R^\sigma(E_F,\vec{k}_\parallel), \tag{7}$$

where once again the prime indicates that all the quantum well states have been omitted. The transport DOS for the left electrode $D_L^\sigma(E_F)$ is also defined by Eq. (7) but $\mathbf{t}$ is replaced by $\mathbf{t}^\dagger$ and $\mathbf{G}_R$ by $\mathbf{G}_L$. It should be noted that the transport DOS is a matrix whose size is dependent on the number of orbitals.

The transport DOS incorporates all the corrections to the classical theory of tunneling discussed above. The generalization of expression (5) of the classical theory of tunneling is, therefore, straightforward. One simply multiplies the transport densities of states for the right and left electrodes taking into account that they are matrices and takes the trace over the orbital indices. This leads to

$$\Gamma^\sigma=\frac{4e^2}{hN_\parallel}\mathrm{Tr}[D_R^\sigma(E_F)][D_L^\sigma(E_F)]. \tag{8}$$

Equation (8) describes incoherent ($\vec{k}_\parallel$ nonconserving) tunneling between two electrodes. In particular, it can be applied to the Co junction with a Cu interlayer. Assuming the same uniform scaling of the *s-p* hopping matrix elements as for the coherent tunneling, we have evaluated from Eq. (8) the dependence of the TMR ratio on the thickness of the Cu overlayer. In Fig. 3, we compare the results for incoherent tunneling deduced from Eq. (8) (broken line), with those for coherent tunneling (continuous line). It can be seen from Fig. 3 that the TMR ratio for incoherent tunneling is not only nonzero but even somewhat larger than that for the coherent tunneling. Oscillations due to quantum interference are weaker than for coherent tunneling and the TMR is again positive. The latter implies that the sign of the polarization of electrons tunneling from the Cu overlayer is the same as that of electrons tunneling from Co, i.e., positive, which is as observed.[4]

Direct evaluation of the Kubo formula for coherent tunneling and of the generalized Julliere formula (8) for incoherent tunneling give nonzero TMR for a Co tunneling junction with a nonmagnetic Cu interlayer. Both these calculations explain a nonzero TMR as being due to quantum well states formed in the down-spin band in the Cu interlayer. This indicates that, for a nonzero TMR effect to occur, one needs a strong scattering at the ferromagnet/nonmagnet interface in one of the spin channels and weak scattering in the other spin channel (strong magnetic contrast). These are the same conditions as those required for a large giant magneto resistance (GMR) in the corresponding ferromagnet/nonmagnet multilayer. It is, therefore, clear that Co/Cu is a particularly good combination but, for example, an Al interlayer should not lead to any sizable TMR since GMR for an Al spacer is very small. This is in agreement with the well-known result (see, e.g., Ref. 17) that an Al interlayer ''kills'' the TMR very effectively.

For a nonzero TMR to be observable, it is necessary that quantum well states in one of the spin channels are well defined (long lived). This is certainly the case when the effect of impurities in the nonmagnetic interlayer is negligible (ballistic transport across the interlayer) and the scattering at the ferromagnet/nonmagnet interface is specular. Scattering from impurities or/and diffuse scattering at the ferromagnet/nonmagnet interface may allow quantum well states to evolve into propagating states, in which case the spin asymmetry of electrons tunneling from the nonmagnetic interlayer is lost (and with it the TMR effect). The fact that the calculated TMR shown in Figs. 2 and 3 is nondecaying as a function of Cu thickness is due to our neglect of impurity/interfacial scattering. For thicker layers impurity scattering/corrugation of the interface becomes important and the TMR is, therefore, expected to decay as a function of the interlayer thickness. However, the precise mechanism (characteristic length) that governs redistribution of quantum well states into propagating states needs further investigation. There is also the possibility that the decay of TMR with the interlayer thickness is caused by spin-flip scattering that mixes the up- and down-spin channels. Spin-flip scattering may be due to magnetic impurities or spin-orbit interaction in the interlayer.

Finally, we need to reconcile the conclusions of Zhang and Levy[10] with our results. Zhang and Levy[10] argued that total conductance of a tunneling junction in the case when coherence is lost is dominated by the reflection coefficient r_2 for the nonmagnet/barrier interface. Since electron scattering at the nonmagnet/barrier interface is spin independent, they go on to conclude that TMR vanishes. However, this argument depends entirely on two assumptions: the reflection coefficient r_1 for the ferromagnet/nonmagnet interface satisfies $r_1\ll r_2$ and $r_2\approx 1$. This is correct only in the absence of quantum well states. Electrons in quantum well states incident on the ferromagnet/nonmagnet interface are of course totally reflected. It follows that $r_1=1$, and we have the opposite limit $r_1\gg r_2$. In that case, the ferromagnet/spacer interface with spin-dependent scattering dominates tunneling and nonzero TMR occurs. (This is an alternative formulation of our spin filtering argument.)

To illustrate the point, we present an idealized example of a junction with a nonmagnetic interlayer for which TMR is demonstrably nonzero regardless of the fact whether scattering is coherent or incoherent. Consider a ferromagnet whose up-spin band is full and the Fermi surface intersects only the down-spin band. We further assume that the bands of the nonmagnetic interlayer match perfectly those of the down-spin carriers in the ferromagnet. All the up-spin states in the interlayer are quantum well states and they remain nonpropagating even if scattering at the interfaces is diffuse since there are no propagating states in the ferromagnet into which they can evolve. Furthermore, the conductance of down-spin electrons is totally unaffected by the interlayer since down-spin electrons see the same potentials in the interlayer and ferromagnet. This conclusion again holds even in the presence of interfacial roughness due to intermixing of atoms. It follows that TMR is completely unaffected by the insertion of the nonmagnetic interlayer. While the model just described applies literally to a junction based on half-metallic ferromagnets, its assumptions are quite well satisfied by the band structures of Co and Cu.

One of us (J.M.) wishes to thank Stuart Parkin for helpful discussions. The support of the Engineering and Physical Sciences Research Council (EPSRC UK) under Grant No. GR/L92945 is gratefully acknowledged.

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