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Theory of resonant tunneling in an epitaxial Fe/Au/MgO/Au/Fe(001) junction

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Calculation of the magnetoresistance of an epitaxial Fe/Au/MgO/Au/Fe(001) tunneling junction is reported. The tunneling magnetoresistance (TMR) is determined without any approximations from the real-space Kubo formula using tight-binding bands fitted to an *ab initio* band structure of Fe, Au, and MgO. A very high TMR ratio of ≈1000% is predicted for a junction with seven to nine atomic planes of MgO sandwiched between two Au layers, each consisting of eight atomic planes. The insertion of two Au interlayers should prevent oxidation of the Fe electrodes, which is detrimental to TMR in the junction without Au interlayers. It is also found that the TMR oscillates as a function of Au thickness with a period determined by the spanning vector of the Au Fermi surface belly. The underlying physics is explained in terms of a simple parabolic band model of resonant tunneling for a barrier sandwiched between two quantum wells.

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Until very recently virtually all experiments on spin-dependent tunneling were performed with magnetic tunnel junctions (MTJ) based on an amorphous aluminium oxide barrier.[1] Disorder in the barrier has the unwelcome consequence that the electron wave vector parallel to the layers $\mathbf{k}_{\parallel}$ is not conserved and the tunneling magnetoresistance ratio (TMR) for such noncoherent tunneling is thus determined only by the spin polarization of the magnetic electrodes. Since this cannot be manipulated for conventional transition metal ferromagnets, the scope for significantly improving the TMR ratio and, in general, for engineering other properties of MTJ with amorphous barriers is very limited. All this has changed with the theoretical prediction[2,3] of a very large TMR for an epitaxial Fe/MgO/Fe(001) junction. In an epitaxial junction $\mathbf{k}_{\parallel}$ is conserved and tunneling is thus coherent. It follows that tunneling in an epitaxial junction is determined by the matching of electron wave functions across the whole junction and, therefore, the TMR ratio depends critically on the particular combination and crystalline orientation of the materials from which the junction is composed. Since the theoretical prediction of a very large TMR for an epitaxial Fe/MgO/Fe(001) junction has been confirmed experimentally,[4,5] it is now appropriate to theoretically explore other epitaxial tunneling junctions based on MgO whose properties can be engineered to suit applications. We propose that epitaxial Fe/Au/MgO/Au/Fe(001) is a system with particularly interesting properties. Our choice of this system was guided by two considerations. One of them has to do with growth of Fe/MgO/Fe. We suggest the introduction of two gold interlayers separating Fe from MgO to prevent oxidation of Fe, which is the problem that plagued the first attempts to grow Fe/MgO/Fe junctions with a very high TMR.[6]

However, our principal motive for choosing Fe/Au/MgO/Au/Fe(001) is that it is a fully epitaxial system in which one can study under controlled conditions the effect of a nonmagnetic interlayer on TMR. One might assume naively that nonmagnetic interlayers between the ferromagnetic electrodes and the barrier destroy the TMR effect since the surface density of states at the Au/MgO interface is spin independent. This would be true for noncoherent tunneling. However, we showed earlier[7] that the TMR remains nonzero for coherent tunneling in a cobalt junction with vacuum gap when a copper interlayer is inserted between one of the Co electrodes and the gap. Our explanation of a nonzero TMR was that a large mismatch between the minority-spin bands in Co and Cu leads to the formation of quantum well (QW) states in the Cu interlayer. Since the QW states are nonconducting and are formed only in the down-spin channel, a large spin asymmetry in transport and, hence a nonzero TMR, remains. This prediction was confirmed experimentally for a Co/Cu/Al_2O_3/Py junction[8] in which the alumina barrier was amorphous. Although only coherence of transport in the Co/Cu bilayer is strictly required for a nonzero TMR,[9] the fully epitaxial junction we propose here is clearly a much cleaner system for studying this effect. Examination of the majority- and minority-spin bands for Fe and Au shows[10] that, as for the Co/Cu combination, there is again a good match for the majority-spin electrons but a poor match for the minority-spin electrons. One might, therefore, expect that the Fe/Au/MgO/Au/Fe(001) junction should have properties similar to those of a junction based on the Co/Cu combination. However, the calculated results for Fe/Au/MgO/Au/Fe(001) junction we present here are completely different, and the explanation of a large TMR that we obtain for this system, requires a qualitatively different physical mechanism. It will be seen that the crucial factor here is the presence of nonmagnetic interlayers on either side of the MgO barrier.

We first address the question of Fe/Au/MgO/Au/Fe(001) growth. There is a very good lattice match between bcc Fe and fcc Au with the Au lattice rotated by 45° relative to that of Fe and it is well known that Fe/Au(001) superlattices can be grown on MgO with monatomic control.[11] Similarly, there is a good lattice match between fcc Au and rocksalt MgO with Au atoms sitting above oxygen sites.[12] We, therefore, assume that the growth is pseudomorphic in the whole structure and neglect in our cal-

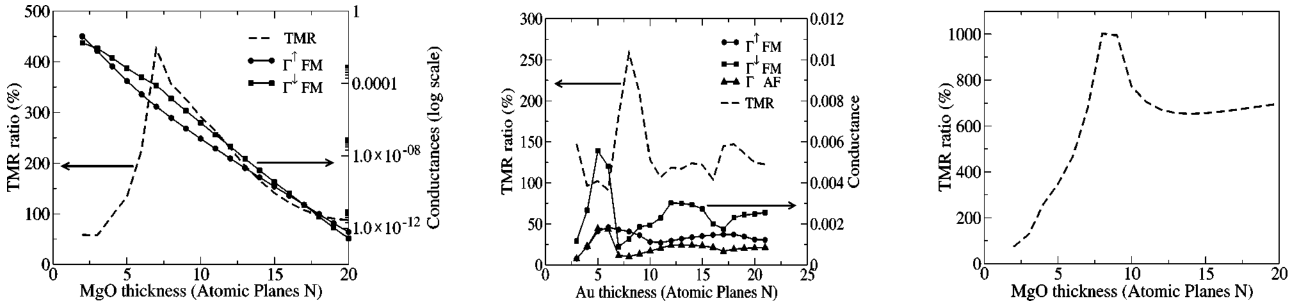

FIG. 1. (a) TMR and conductances as a function of MgO thickness for an Au thickness of 4 atomic planes. The left-hand axis refers to the TMR, and the right-hand axis to the conductances $\Gamma^{\uparrow}_{\mathrm{FM}}$ and $\Gamma^{\downarrow}_{\mathrm{FM}}$; (b) TMR and conductances as a function of Au thickness for a MgO thickness of 4 atomic planes. The left-hand axis refers to the TMR, and the right-hand axis to the conductances $\Gamma^{\uparrow}_{\mathrm{FM}}$, $\Gamma^{\downarrow}_{\mathrm{FM}}$, and Γ_{AF} ; (c) TMR as a function of MgO thickness for an Au thickness of 8 atomic planes.

culations any small lattice mismatch. We describe the band structure of the Fe electrodes and that of the Au interlayers by tight-binding bands fitted to the *ab initio* band structure of bcc Fe and fcc Au.[10] Similarly, the barrier is described by tight-binding bands fitted to the band structure of bulk MgO.[13] The on-site potentials in the Fe/Au and Au/MgO interface planes were adjusted self-consistently to preserve charge neutrality, and to reproduce the correct surface moment of Fe. The band gap for the band structure of bulk MgO we use is 7.6 eV. Further details of our tight-binding parametrization may be found in Ref. 2.

The tunneling conductance Γ^{σ} in the spin channel σ of a Fe/Au/MgO/Au/Fe(001) junction was determined by the same method as in our previous calculation for Fe/MgO/Fe.[2] The tunneling current between two neighboring atomic planes in the MgO barrier, labeled 0 and 1, was evaluated from the real-space Kubo formula. Using a mixed representation that is Bloch-like in the plane of the layers and atomiclike in the perpendicular direction, it is easy to express the Kubo formula in terms of one-electron Green's functions at the Fermi surface ($E=E_F$). The conductance Γ^{σ} is given by

$$\Gamma^{\sigma} = \frac{4e^2}{h}\sum_{\mathbf{k}_{\parallel}} \mathrm{Tr}([\mathbf{T}_{\sigma}\,\mathrm{Im}\,\mathbf{G}_0^{\sigma}(\mathbf{k}_{\parallel})]\cdot[\mathbf{T}_{\sigma}^{\dagger}\,\mathrm{Im}\,\mathbf{G}_1^{\sigma}(\mathbf{k}_{\parallel})]). \quad (1)$$

The summation in Eq. (1) is over the two-dimensional Brillouin zone (BZ) and the trace is over the orbital indices corresponding to s,p,d orbitals that are required in a tight-binding parametrization of the junction. Finally, $\mathbf{G}_0^{\sigma}(\mathbf{k}_{\parallel})$ and $\mathbf{G}_1^{\sigma}(\mathbf{k}_{\parallel})$ are the one-electron Green's functions at the left (right) surfaces of a junction that is separated into two independent parts by an imaginary cleavage plane drawn between the atomic planes 0,1. The separation of the junction into two independent parts is made simply for calculational purposes. The junction remains physically connected and the interaction between the left and right parts is fully restored in Eq. (1) by the matrices $\mathbf{T}_{\sigma}$ and $\mathbf{T}_{\sigma}^{\dagger}$ defined by

$$\mathbf{T}_{\sigma} = \mathbf{t}_{01}(\mathbf{k}_{\parallel})[\mathbf{I} - \mathbf{G}_1^{\sigma}(\mathbf{k}_{\parallel})\mathbf{t}_{01}^{\dagger}(\mathbf{k}_{\parallel})\mathbf{G}_0^{\sigma}(\mathbf{k}_{\parallel})\mathbf{t}_{01}(\mathbf{k}_{\parallel})]^{-1}, \quad (2)$$

where $\mathbf{I}$ is a unit matrix in the orbital space and $\mathbf{t}_{01}(\mathbf{k}_{\parallel})$ is the tight-binding hopping matrix connecting the surfaces 0 and 1. The calculation of the surface Green's functions and the problems of numerical accuracy are discussed in Ref. 2.

We are now ready to present our results for the system Fe/Au/MgO/Au/Fe(001). We use the "optimistic" tunneling magnetoresistance ratio $R_{\mathrm{TMR}}=(\Gamma_{\mathrm{FM}}-\Gamma_{\mathrm{AF}})/\Gamma_{\mathrm{AF}}$, where Γ_{FM} and Γ_{AF} are the total conductances in the ferromagnetic (FM) and antiferromagnetic (AFM) configurations of the junction. In Figs. 1(a) and 1(c), we show R_{TMR} as a function of MgO thickness for fixed thicknesses of four and eight atomic planes of Au, respectively. In Fig. 1(a) we also show the majority-spin $\Gamma^{\uparrow}_{\mathrm{FM}}$ and minority-spin $\Gamma^{\downarrow}_{\mathrm{FM}}$ conductances in the ferromagnetic configuration of the junction on a logarithmic scale. In Fig. 1(b), R_{TMR}, $\Gamma^{\uparrow}_{\mathrm{FM}}$, $\Gamma^{\downarrow}_{\mathrm{FM}}$, and the conductance Γ_{AF} in the antiferromagnetic configuration are plotted as a function of the Au layer thickness for a fixed thickness of four atomic planes of MgO. All the conductances are measured in units of the quantum conductance (e^2/h).

The first rather remarkable result seen in Figs. 1(a) and 1(c) is that, in contrast to Fe/MgO/Fe(001), the TMR ratio of the Fe/Au/MgO/Au/Fe(001) junction peaks as a function of MgO thickness in the region of seven to nine atomic planes of MgO. For an Au thickness of 8 atomic planes [Fig. 1(c)], R_{TMR} reaches $\approx$1000%, which is almost as high as the highest calculated TMR for the Fe/MgO/Fe(001) junction without Au interlayers.[2] The other even more surprising result seen in Figs. 1(a) and 1(b) is that the conductance $\Gamma^{\downarrow}_{\mathrm{FM}}$ in the minority-spin channel is generally much higher than that in the majority-spin channel. We recall that for the Fe/MgO/Fe(001) junction just the opposite is true.[2]

In fact, the very high TMR for the Fe/MgO/Fe(001) junction is obtained because perpendicular tunneling at the $\overline{\Gamma}$ point $\mathbf{k}_{\parallel}=0$, which dominates in the majority-spin channel, is forbidden in the minority-spin channel by the symmetry of wave functions.[3] On the basis of the quantum well argument we advanced for a Co junction with a Cu interlayer,[7] one might expect that tunneling in the minority-spin channel in the Fe/Au/MgO/Au/Fe(001) junction should be even further reduced because of the formation QW states in this channel. Clearly this simple QW argument is not applicable here. On the other hand, large-amplitude oscillations of $\Gamma^{\downarrow}_{\mathrm{FM}}$ as a function of Au thickness, seen in Fig. 1(b), are a clear indication that QW states are involved in tunneling of minority-spin electrons. This is further supported by the fact that the period of such oscillations $\approx$10 atomic planes is very

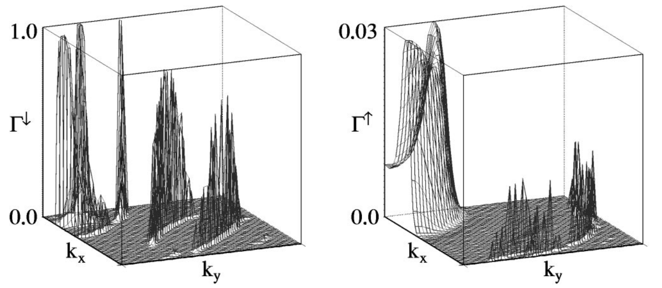

FIG. 2. (a) The $\mathbf{k}_\parallel$ dependences of the minority-spin $\Gamma_{FM}^{\downarrow}(\mathbf{k}_\parallel)$ and (b) majority-spin $\Gamma_{FM}^{\uparrow}(\mathbf{k}_\parallel)$ conductances for Fe/Au(4)/MgO(4)/Au(4)/Fe junction.

close to the period obtained from the spanning vector at the belly of the Au Fermi surface.[16]

A clue to the explanation of this unexpected behavior can be found in Fig. 2(a), which shows the $\mathbf{k}_\parallel$ dependence of the minority-spin conductance $\Gamma_{FM}^{\downarrow}(\mathbf{k}_\parallel)$ in the irreducible segment of the two-dimensional BZ for Fe/Au(4)/MgO(4)/Au(4)/Fe. The corresponding dependence for the majority-spin conductance $\Gamma_{FM}^{\uparrow}(\mathbf{k}_\parallel)$ is shown in Fig. 2(b). While $\Gamma_{FM}^{\uparrow}(\mathbf{k}_\parallel)$ has the expected peak close to the $\bar{\Gamma}$ point $\mathbf{k}_\parallel=0$ with the highest tunneling conductance at the peak of only $\approx 3\times 10^{-2}$ (in units of quantum conductance), we can clearly see in Fig. 2(a) that there are resonance peaks of $\Gamma_{FM}^{\downarrow}(\mathbf{k}_\parallel)$ where $\Gamma_{FM}^{\downarrow}(\mathbf{k}_\parallel)\approx 1$. It follows that there are points in the two-dimensional BZ located close to, but not exactly at $\mathbf{k}_\parallel=0$, where full transmission across the MgO barrier occurs in the minority-spin channel. We propose that the correct explanation of such a very high tunneling conductance in the minority-spin channel, and the corresponding very high TMR, is resonant tunneling of minority-spin electrons mediated by QW states in the gold interlayers. To prove that the effect is due to QW states in gold, we have computed the dependence of the partial conductance $\Gamma_{FM}^{\downarrow}(\mathbf{k}_\parallel^0)$ on Au thickness, where $\mathbf{k}_\parallel=\mathbf{k}_\parallel^0$ is a point of full conductance close to $\mathbf{k}_\parallel=0$ (see Fig. 2). This calculation shows a perfectly periodic sequence of peaks of full transmission that persist up to the largest computed thicknesses of 1000 atomic planes of Au. Such a periodic dependence on Au thickness unambiguously rules out any involvement of interfacial states, and validates our claim that the cause of resonant tunneling is QW states in the gold interlayers.

We now need to address the question of the physical origin of such resonant tunneling. Since all the evidence points to the existence of quantum wells, we can make a simple parabolic band model of the potential profile seen by minority-spin electrons. This is shown in Fig. 3(a), where k_0, k_1, and k_2 are the magnitudes of the electron wave vectors in the corresponding regions (the wave vector in the barrier is, of course, imaginary). A straightforward but mildly tedious calculation, using the standard transfer-matrix method to match the wave functions, shows that, indeed, a full transmission through a barrier sandwiched between two symmetric wells may occur provided an integer number of electron half-wavelengths can be fitted into each well. This implies that the transmission coefficient (conductance) oscillates as a function of the nonmagnetic layer thickness with a period determined by the electron wavelength $2\pi/k_1$ in the direction perpendicular to the layers.

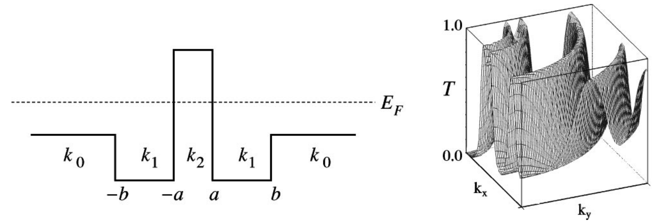

FIG. 3. (a) Potential profile for minority-spin electrons and (b) the $\mathbf{k}_\parallel$ dependence of their transmission coefficient T, in a parabolic band model.

However, fitting an integer number of electron half-wavelengths into the spacer layer QW is only a necessary but not sufficient condition for full transmission. The necessary and sufficient condition takes the form

$$\sin[2k_1(b-a)+k_1a-\psi]=\Delta, \qquad (3)$$

where

$$\Delta=\frac{(1+x)(1+y)\sinh(2ak_2)}{(1-y)[4x\cosh^2(ak_2)+(1-x)^2\sinh^2(ak_2)]^{1/2}}, \qquad (4)$$

with $x=k_1^2/k_2^2$, $y=k_0^2/k_1^2$, and $\tan(\psi)=[(1-x)/2x]\tanh(ak_2)$. It follows from Eq. (3) that full transmission occurs only if $|\Delta|\leqslant 1$. This is a more stringent condition than the requirement that one should be able to fit an integer number of half-wavelengths into the well, which is sufficient for the well-known resonant tunneling through a double barrier.[14] (For an application of double barrier resonance to the TMR in semiconductor systems see Ref. 15.) However, it is clear that the condition for full transmission may be satisfied for a narrow or low barrier, when the factor $\sinh(2ak_2)$ is small. However such a resonance will eventually disappear as the barrier height or width are increased.

To obtain the total transmission coefficient (conductance) we need to sum over all $\mathbf{k}_\parallel$. Since $\mathbf{k}_\parallel^2+k_\perp^2=k_F^2$, where k_F is the Fermi wave vector, the partial conductances with different $k_\perp$ oscillate as a function of the well width with different periods. However, applying the usual stationary-phase argument,[16] we find that all such oscillations cancel out and only the oscillation with a period coming from an extremum of the spacer layer Fermi surface (FS) survives. This argument explains why it is the spanning vector of the Au FS belly that determines oscillations of the TMR in Fe/Au/MgO/Au/Fe as a function of Au thickness [Fig. 1(b)]. Other possible periods coming from the Au FS necks and from cutoff points at the edge of the Au QW (Ref. 16) are suppressed since perpendicular tunneling with $\mathbf{k}_\parallel\approx 0$ dominates.[9]

In Fig. 3(b) we show the $\mathbf{k}_\parallel$ dependence of the transmission coefficient T (conductance) for a parabolic band model. The resonance rings of full transmission are clearly analogous to the conductance peaks seen in Fig. 2(a) for $\Gamma_{FM}^{\downarrow}$ in the Fe/Au(4)/MgO(4)/Au(4)/Fe(001) junction. We have verified that the dominant contribution to $\Gamma_{FM}^{\downarrow}$ arises from these resonances. This fact enables us to explain the main qualitative features of the TMR as a function of MgO thickness. Let us consider the case of four atomic planes of Au as depicted in Fig. 1(a) [the case of eight atomic planes of Au shown in Fig. 1(c) is qualitatively similar]. We find for the parabolic band model that the contribution of resonances to the total tunneling conductance decreases slowly with barrier thickness as long as the system remains in resonance (i.e., for thin barriers). Hence, initially, $\Gamma_{FM}^{\downarrow}$ in the Fe/Au/MgO/Au/Fe junction, decreases relatively slowly while $\Gamma_{FM}^{\uparrow}$ decreases exponentially as in normal tunneling, and this produces the initial rise in the TMR seen in Fig. 1(a). The resonances in $\Gamma_{FM}^{\downarrow}$ remain until MgO reaches a thickness of about seven atomic planes, which coincides with a sudden drop in $\Gamma_{FM}^{\downarrow}$, leading to a subsequent decrease in the TMR. For very thick MgO, we expect that all resonance contributions disappear and, therefore, the system should behave eventually like Fe/MgO/Fe. A thick insulator increasingly favors perpendicular tunneling $\mathbf{k}_{\parallel}=0$ and the symmetry of wave functions dictates that $\Gamma_{FM}^{\downarrow}(\mathbf{k}_{\parallel}=0)=0$ but $\Gamma_{FM}^{\uparrow}(\mathbf{k}_{\parallel}=0)$ remains nonzero.[3] It follows that the TMR should start to increase as a function of MgO thickness, as in Fe/MgO/Fe. The change of regime from resonant to nonresonant Fe/MgO/Fe-like tunneling is completed when the majority-spin conductance $\Gamma_{FM}^{\uparrow}$ becomes greater than $\Gamma_{FM}^{\downarrow}$. It can be seen in Fig. 1(a) that this occurs for a MgO thickness of ≈15 atomic planes. The increase of the TMR for thick MgO is clearly visible in Fig. 1(c), but not verifiable in Fig. 1(a) because it occurs for MgO thicknesses so large that the conductances are too small to be computed reliably.

In conclusion, our calculations based on a fully realistic band structure and rigorous Kubo formula predict a very high TMR ratio for an epitaxial Fe/Au/MgO/Au/Fe(001) junction. The maximum TMR ratio of ≈1000% occurs for a junction with seven to nine atomic planes of MgO sandwiched between two Au interlayers, each consisting of eight atomic planes. The insertion of two Au interlayers between the Fe electrodes and the MgO barrier should prevent oxidation of the Fe electrodes, which is detrimental to the TMR. We also found that the TMR oscillates as a function of the Au layer thickness with a period determined by the spanning vector of the Au FS belly. The underlying physics can be understood in terms of a simple parabolic band model of resonant tunneling for a barrier sandwiched between two quantum wells. This model explains well the origin of a high conductance of minority-spin eletrons in the Fe/Au/MgO/Au/Fe(001) junction and oscillations of the TMR in this system. It also predicts correctly the oscillation period and the qualitative behavior of the TMR as a function of MgO thickness.

The principal message of this theoretical study is, therefore, that the properties of MgO-based MTJs can be tuned to suit applications by the insertion of nonmagnetic interlayers without a significant reduction in their TMR. This holds provided the junction remains epitaxial so that tunneling is coherent and there is a large mismatch between the bands of the ferromagnet and nonmagnet in one spin channel but good match in the other spin channel.

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