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# Magnetoresistance of magnetic tunnel junctions in the presence of a nonmagnetic layer

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A quantum-statistical theory of electron tunneling in magnetic junctions of the form $F/P/O/F$, where $F$ and $P$ are, respectively, a magnetic and a paramagnetic metal is presented. The effect of scattering at the $P/O$ interface is discussed. The relatively slow decay of tunnel magnetoresistance experimentally observed when $P$ is Cu as a function of the Cu layer thickness is explained by considering the particular shape of the Fermi surface in the (111) direction. For other metals, our model predicts a much more rapid decay, in agreement with experimental observations.

## I. INTRODUCTION

In a recent paper,[1] we investigated a model of magnetic tunnel junction comprising one (or two) nonmagnetic layer(s), inserted between the ferromagnetic electrodes and the insulating layer. It was shown that in such junctions, the nonmagnetic layer can constitute a spin-dependent quantum well. This is the case in particular if a Cu spacer layer is inserted at the interface between a Co electrode and $Al_2O_3$. Indeed, in the theory of oscillatory coupling in (Co/Cu) multilayers, it was shown that a good electron band matching occurs for minority electrons at the Co/Cu interface whereas the Cu layers constitute quantum wells for majority electrons.[2] The oscillatory coupling then results from the existence of spin-dependent coefficients of reflection of electrons at the Co/Cu interfaces. Similarly in Co/Cu/$Al_2O_3$/ferromagnet junctions, the conductance through the barrier and the tunnel magnetoresistance (TMR) oscillates with the thickness of the paramagnetic layer $a$, exhibiting resonances, due to the existence of spin-dependent quantum-well states in the paramagnetic layer. Furthermore, it was shown that large enhancements in the TMR amplitude could be observed when two paramagnetic layers of the same thickness are introduced on both sides of the barrier. In such a situation, the amplitude and average value of the TMR oscillations slowly decrease and are still of the order of a few % for $a$ of the order of a few nanometers. In this calculation,[1] bulk scattering in the metallic layers was taken into account. Experimental attempts to observe this effect in junctions with Cu spacer on both sides of the barrier were carried out in Co/Cu/$Al_2O_3$/Cu/NiFe junctions.[3] However, all of the junctions with Cu underneath the barrier were short circuited. Furthermore, the requirement of keeping the thickness of the two paramagnetic layers equal is quite difficult to achieve experimentally so that our theoretical prediction could not be tested. In the case of an asymmetric junction, it was observed experimentally that the insertion of an interfacial paramagnetic layer on one side of the barrier only, in most cases, very rapidly reduces the TMR amplitude. For instance, a few monolayers of metallic Al between the ferromagnetic electrode and the $Al_2O_3$ barrier are sufficient to strongly reduce the TMR amplitude. In contrast, in the case of Cu, Sun and Freitas[3] showed that the TMR decays much more slowly with the copper layer thickness than for other metals. For a Cu thickness of 50 Å, a TMR of 1% at 300 K is still observed. However, it seems that this result varies significantly from one group to another and therefore depends on the details of the preparation method.[4] In a recent paper,[5] Zhang and Levy explained the rapid drop of TMR observed with most paramagnetic layers by considering the loss of coherence in transmission through the paramagnetic layer due to fluctuations in the thickness of the inserted layer or to the interaction between modes with different in-plane momentum $k$ of electrons transmitted through the barrier. However, we think that the situation is not described completely in Ref. 5. First, the fluctuations in the thickness of the inserted layer can have quite different length scales in the plane of the structure. On one hand, the layers can form large terraces with thickness fluctuating by plus or minus one monolayer. These terraces may have lateral dimension $D$ much larger than $k_F^{-1}$ ($k_F$ is Fermi momentum). In that case, the electron transmissions over different terraces can be considered as independent. The observed value of tunnel current and consequently of TMR is then an average over the distribution of the layer thickness. On the other hand, if the roughness in the metal/insulator interface is caused by intermixing

 

at the atomic scale, the characteristic length of the roughness in the plane is of the order of $k_F^{-1}$. To take into account the influence of such roughness on the tunnel conductivity, one has to solve the problem of electron tunneling with scattering at the rough interfaces of the barrier. We therefore decided to take into account both types of interfacial defects and to investigate their influence on the observed value of TMR. In Ref. 5 the expression obtained for the conductance versus the thickness of the inserted layer $a$ (supposed in the calculation to be perfectly flat) was integrated over values of $a$ in the limits $[a-a_0/2, a+a_0/2]$, where $a_0$ indicates the lattice parameter. As we mentioned above, we consider that in such a situation the proper way to take into account such a roughness is to consider the electron scattering at the interface. Furthermore, for perfectly flat interfaces, we obtained the asymptotic expression of the conductivity for large thickness $a$ of the inserted layer ($ak_F \gg 1$) and showed that the TMR amplitude drops as the thickness $a$ increases as

$$\frac{ak_F \exp(-a/l)}{(bq)^2+(ak_F)^2},$$

where $b$ and $q$ represent, respectively, the thickness of the barrier and the damping wave vector of electrons in the barrier, and $l$ is the electron mean-free path in the paramagnetic layer.[1] The critical length of decay in the TMR amplitude $a_c=\pi bq/k_F$ introduced in Ref. 5 corresponds to a balance between the two terms in the denominator of the above expression.

## II. MODEL

We consider a magnetic tunnel junction consisting of two ferromagnetic electrodes separated by an insulating barrier with additional paramagnetic layer placed between one of the ferromagnetic electrodes and the barrier. On the interface between this paramagnetic layer and the insulator we assume the presence of a random distribution of scattering centers (impurities) with short-range potential $v_n^i(z,\rho)=\delta_n^i\delta(z-z_0)\delta(\rho-\rho_n)$, with $\overline{\delta_n^i\delta_m^i}-\overline{\delta_n^i}*\overline{\delta_m^i}=((\overline{\delta_n^{i^2}}-\overline{\delta_n^i})^2)\Delta_{nm}$, where $i=\uparrow(\downarrow)$ refers to the spin index, $z$ is the axis perpendicular to the plane of the layers, $z_0$ is the coordinate of the paramagnetic/insulator interface, $\rho$ and $\rho_n$ are the electron's and impurities' coordinates in plane of the interface, the overbar denotes the average over the distribution of impurities, and $\Delta_{nm}$ is the Kroneker symbol.

To calculate the two-point conductivity $\sigma(z,z')$, the Kubo formula in the mixed real-space momentum representation[1] is used in the limit of low-bias voltage as compared to the height of the barrier, which is typically of the order of 2 eV (linear response):

$$\sigma(z,z')=\frac{4e^2}{\pi\hbar}\sum_{k,k',i} A_{kk'}^i \vec{\nabla}_z \vec{\nabla}_{z'} A_{kk'}^i, \tag{1}$$

where $\nabla_z=(1/2)(\vec{\nabla}_z-\overleftarrow{\nabla}_z)$ is the antisymmetric gradient, and $A_{kk'}^i=(1/2)[G_{kk'}^{ret,i}-(G_{kk'}^{ret,i})^*]$ and $G_{kk'}^{ret,i}$ are the retarded Green functions of electrons with spin-index $i$. To calculate the average over impurity configurations in expression (1), the coherent potential approximation (CPA) was used.[6] The difference with respect to the usual situation of bulk scattering is that the initial Green functions must be calculated here in the system with a barrier. In this case, it is important to point out that in contrast to the homogeneous case, the vertex corrections are not equal to zero. They were calculated in CPA ladder approximation.[7] They give large contributions to the total tunnel conductivity. As an example, the final expression for the conductance for spin $\uparrow$ channel for antiferromagnetic configuration of magnetic moments in ferromagnetic layers can be written as

$$\begin{aligned}\sigma_{AP}^{\uparrow}=&\frac{e^2}{\pi^2\hbar}\int \kappa\, d\kappa\\ &\times\frac{c_2[(c_1^2+c_2^2)\sinh 2d_2a+2c_1c_2\cosh 2d_2a]}{|Den|^2}\\ &\times\left\{\frac{q^2\exp(-2qb)c_3}{q^2+c_3^2}\right.\\ &+\frac{2a_0^2}{\pi}\Gamma^{\uparrow}\int \tilde{\kappa}\, d\tilde{\kappa}\frac{\tilde{c}_3\tilde{q}^2\exp(-2\tilde{q}b)}{q^2+c_3^2}\\ &\left.\times\left(\frac{|\exp(i\tilde{k}_2a)(\tilde{c}_1-\tilde{c}_2)-\exp(-i\tilde{k}_2a)(\tilde{c}_1+\tilde{c}_2)|}{|D\tilde{e}n|}\right)^2\right\},\end{aligned} \tag{2}$$

where

$$\begin{aligned}Den=&\exp(ik_2a)(c_1-c_2)(ic_2+q+\Sigma^{\uparrow})\\ &-\exp(-ik_2a)(c_1+c_2)(ic_2-q-\Sigma^{\uparrow}).\end{aligned} \tag{3}$$

In these expressions, $a_0$ represents the lattice parameter, $\hbar q_0=\sqrt{2m(U_0-\epsilon_F)}$. $b$ and $U_0$ indicate, respectively, the thickness and the height of the barrier. $\epsilon_F$ is the Fermi energy. $q=\sqrt{q_0^2+\kappa^2}$, $\kappa$ is the modulus of electron momentum in the plane of the structure. $k_i=c_i+id_i=\sqrt{(k_F^{(i)})^2-\kappa^2+2ik_F^{(i)}/l_i}$, the $l_i$'s indicate the mean-free paths, and $k_F^{(i)}$ is the Fermi momentum for spin $\uparrow$ ($\downarrow$) electrons when $i=1$ (3) (in ferromagnetic layers) and for electrons in the paramagnetic layer when $i=2$. Similar expressions give $\tilde{q}_i$ and $\tilde{k}_i$ as a function of the integration variable $\tilde{\kappa}$. $\Sigma^{\uparrow}$ is the coherent potential at the interface for spin $\uparrow$ electrons. Its value is the solution of the usual CPA equation $\overline{T\{\Sigma^{\uparrow}\}}=0$, where $T$ is the scattering matrix. Furthermore, $\Gamma^{\uparrow}$ is the vertex correction in conductivity. It is given by the expressions

$$\Gamma^{\uparrow}=\frac{\overline{|T^{\uparrow}|^2}}{1-\overline{|T^{\uparrow}|^2}D^{\uparrow}}, \tag{4}$$

$$\begin{aligned}\overline{|T^{\uparrow}|^2}=&c_A\left(\frac{\delta^A-\Sigma^{\uparrow}}{1-(\delta^A-\Sigma^{\uparrow})(1/N)\Sigma_{\kappa}G_{\kappa}^{\uparrow}}\right)^2\\ &+c_B\left(\frac{\delta^B-\Sigma^{\uparrow}}{1-(\delta^B-\Sigma^{\uparrow})(1/N)\Sigma_{\kappa}G_{\kappa}^{\uparrow}}\right)^2\end{aligned} \tag{5}$$

$$D^{\uparrow}=\frac{1}{N}\sum_{\kappa}|G_{\kappa}^{\uparrow}(z_0,z_0)|^2-\left|\frac{1}{N}\sum_{\kappa}G_{\kappa}^{\uparrow}(z_0,z_0)\right|^2, \tag{6}$$

where $c_{A(B)}$ and $\delta_{A(B)}=\alpha[(k_F^{(2)})^2+q_0^2]$ represent, respectively, the concentration and the scattering potential of atoms of type $A(B)$ in the interfacial alloy $A_xB_{1-x}$ resulting from the intermixing between the paramagnetic layer and the oxide barrier. In the present model, for simplicity, $\delta_A$ and $\delta_B$ are not spin dependent. $G_\kappa^{\uparrow}(z_0,z_0)$ is the Green function at the interface:

$$G_\kappa^{\uparrow}(z_0,z_0)=-\frac{\exp(ik_2a)(c_1-c_2)-\exp(-ik_2a)(c_1+c_2)}{Den}. \tag{7}$$

The expressions for spin ↓ electron conductance and for ferromagnetic alignment of magnetizations may be obtained from Eq. (2) by appropriate change of indexes $i$.

## III. RESULTS AND DISCUSSION

The important conclusions which can be derived from expressions (2)–(7) are the following: First of all, in the absence of interfacial scattering, all $\Sigma$'s and $\Gamma$'s in expression (2) are equal to zero. The results obtained in Refs. 1 and 5 are then recovered. We can estimate the conductance in the asymptotic limit of large thicknesses $k_Fa$ and $q_0b\gg1$. In this limit, the expression of the conductivity can be written as

$$\sigma_{AP}^{\uparrow}=\sigma_{\text{bulk}}\left\{1+\left[1+\left(\frac{c_1-c_2}{c_1+c_2}\right)\exp\left(-4\frac{a}{l}\right)\right]\left(\frac{c_1-c_2}{c_1+c_2}\right)\right.$$
$$\times\exp\left(-2\frac{a}{l}\right)\left[\frac{(k_F^{(2)}a)\cos(2k_F^{(2)}a+\phi)}{(k_F^{(2)}a)^2+(q_0b)^2}\right.$$
$$\left.\left.+\frac{(q_0a)\sin(2k_F^{(2)}a+\phi)}{(k_F^{(2)}a)^2+(q_0b)^2}\right]\right\}, \tag{8}$$

where

$$\arctan\phi=\frac{2q_0k_F^{(2)}}{q_0^2-(k_F^{(2)})^2}$$

and $\sigma_{\text{bulk}}$ is the conductance of the system for infinitely thick paramagnetic layers.

As is clear from expression (8), the ferromagnetic layer adjacent to the paramagnetic spacer influences the total conductivity and correlatively produces a TMR effect through the second term in Eq. (8) which is an oscillating term. The amplitude of these oscillations decreases with the thickness of the paramagnetic layer. The real characteristic length of this decrease is $l$—electron's mean-free path in the paramagnetic layer. However, in the case where $l\gg a$, the TMR decreases approximately as $(k_F^{(2)}a)^{-1}$ so that the value $a_c=\pi bq_0/k_F^{(2)}$ may be considered in some sense as a characteristic length, but not as a characteristic length of an exponential decay.

Let us now consider the situation where the interfacial scattering is taken into account. In expression (2), an additional term $\Sigma^{\uparrow}$ appears in the denominator (3) which describes the influence of interfacial scattering on the transmission of electrons through the structure. This denominator is an oscillating function of the product $c_2a$. For definite values of $c_2a$, the real part of the expression (3) is equal to zero. This results in resonances (sharp oscillations) in conductance

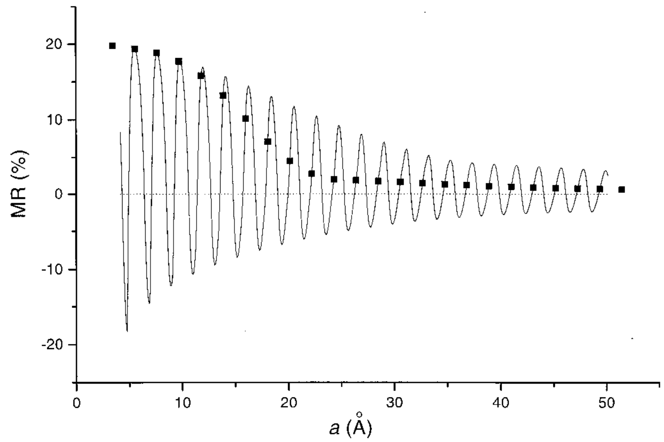


FIG. 1. Calculated MR of Co/Cu[111]/$Al_2O_3$/Co magnetic junction vs thickness of the paramagnetic layer $a$. Square points indicate the values of MR for thicknesses corresponding to an integer number of monolayers. The parameters are $k_F^{(1)}=1.09\,\text{Å}^{-1}$, $k_F^{(2)}=1.51\,\text{Å}^{-1}$, $k_F^{(3)}=0.42\,\text{Å}^{-1}$, $a_0=2.08\,\text{Å}$, $q_0=0.9\,\text{Å}^{-1}$, $b=10\,\text{Å}$, $\alpha=0$.

and consequently in TMR, associated with the formation of quantum-well states in the paramagnetic layer. This is illustrated in Figs. 1 and 2, which show the calculated TMR versus the thickness ($a$) of the paramagnetic layer, respectively, Cu (111) for Fig. 1 and Al for Fig. 2. From earlier studies on (Co/Cu) multilayers, it is known that a layer-by-layer (111) growth of Cu can be achieved on fcc(111) Co by sputtering or molecular-beam epitaxy (MBE). The resulting Cu layer can either be single crystal if grown by MBE or with a strong (111) texture if grown by sputtering. If the Cu layer is grown above the tunnel barrier, i.e., on amorphous alumina, it is likely that a (111) texture is obtained since the (111) planes are the dense planes in fcc crystal. In these cases, the interatomic spacing along the growth direction is $a_0=2.08\,\text{Å}$ and the Fermi wave vector in this direction is exactly equal to $\pi/a_0=1.51\,\text{Å}^{-1}$.[2] Much less is known about the growth of Al on Co or other ferromagnetic transition metals. The thin Al layer which is deposited on the

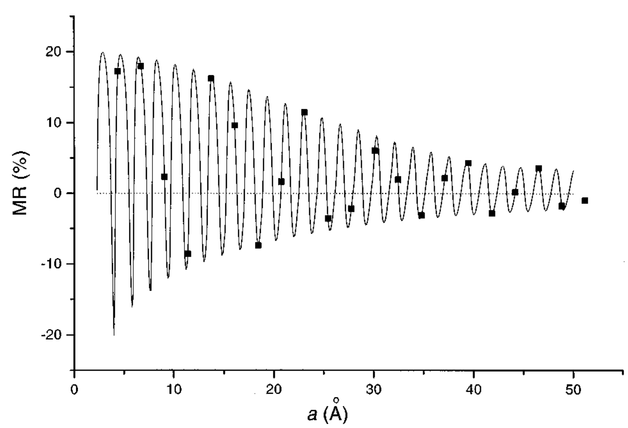


FIG. 2. Calculated MR of Co/Al/$Al_2O_3$/Co magnetic junction vs thickness of the paramagnetic layer $a$. Square points indicate the values of MR for thicknesses corresponding to an integer number of monolayers. The parameters are $k_F^{(1)}=1.09\,\text{Å}^{-1}$, $k_F^{(2)}=1.75\,\text{Å}^{-1}$, $k_F^{(3)}=0.42\,\text{Å}^{-1}$, $a_0=2.34\,\text{Å}$, $q_0=0.9\,\text{Å}^{-1}$, $b=10\,\text{Å}$, $\alpha=0$.

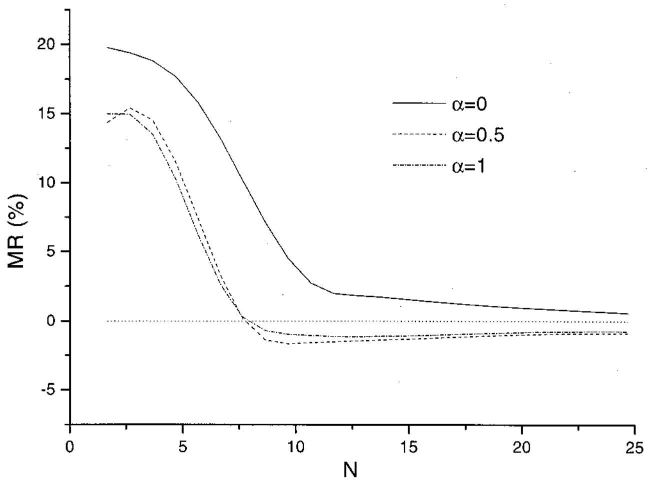


FIG. 3. Averaged MR amplitude [see text, expression (9)] of Co/Cu[111]/$Al_2O_3$/Co junction vs the number of Cu monolayers $N$ for different values of interfacial scattering amplitude. The parameters are the same as for Fig. 1.

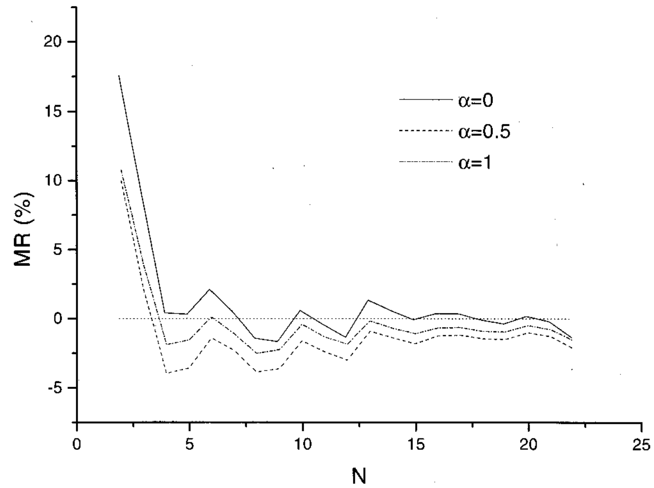


FIG. 4. Averaged MR amplitude [see text, expression (9)] of Co/Al/$Al_2O_3$/Co junctions vs the number of Cu monolayers $N$ for different values of $\alpha$. The parameters are the same as for Fig. 2.

bottom ferromagnetic electrode prior to oxidation is likely to be polycrystalline considering the large lattice mismatch between, for instance, Co, and Al. For Al, which is also fcc, we therefore chose a lattice parameter equal to 2.34 Å in the direction perpendicular to the interface. This corresponds to the lattice parameter in the (111) direction. However, as we will show further, the exact value chosen for this parameter is not important in the following discussion. The only important feature is that the Fermi wave vector in Al perpendicular to the interface is significantly smaller than $\pi/a_0$. This is actually the most common situation except in a few particular cases such as Cu (111). In Figs. 1 and 2, the positions and amplitudes of the resonances depend not only on the value of $k_F^{(2)}$, but also on the value of the Fermi momentum of spin $\uparrow$ and $\downarrow$ electrons in the adjacent ferromagnetic layer. However, if the electrons are strongly scattered at the interface between the paramagnetic metal and the insulator and if the amplitude of the scattering potential is not spin dependent, the resonances become much broader and the TMR decreases. This is illustrated in Figs. 3 and 4, where we plotted the averaged TMR amplitude [see further the expression (9) versus thickness (number of monolayers)] of paramagnetic layer for several values of the scattering potential amplitude. In that sense, the scattering on the barrier interface produces the same decrease in TMR amplitude than the scattering in the bulk of the paramagnetic layer discussed in Ref. 1.

Let us now discuss the effect of the presence of large terraces at the interfaces of the paramagnetic layer. As an example, we assume that the thickness of this layer changes by plus or minus one atomic distance around its average value with equal weight for each thickness. In this case, the conductance must be averaged as follows:

$$\sigma(a_n)=\frac{\sigma(a_n-a_0)+\sigma(a_n)+\sigma(a_n+a_0)}{3}. \tag{9}$$

Furthermore, in a very similar way as the aliasing effect introduced to explain the long-period oscillation of the oscillatory coupling in magnetic multilayers,[8] we have to consider that the thickness of the layer can only vary by an integer number of atomic planes. Therefore, in Figs. 1 (for Cu) and 2 (Al), we denoted by squares the values of TMR at points where $a=na_0$ ($n=0,1,2\ldots,a_0$—lattice parameter). These TMR values, averaged according to Eq. (9), have been replotted in Figs. 3 and 4, respectively, for Cu(111) and Al. It is clear in Fig. 1 that in the case of copper paramagnetic layer, the TMR amplitude decays relatively slowly as a function of the number of monolayers. The TMR still exhibits a significant amplitude ($\geqslant 1\%$) for a Cu thickness of the order of 50 Å, in agreement with the experimental observation of Sun and Freitas.[3] This slow decay can be explained by the properties of the electronic structure of Cu.[9] One can see from the cross section of the Fermi surface for a fcc (001) spacer [see Fig. 1(b) in Ref. 9] that along the (111) direction the value of Fermi momentum for Cu is equal to $\pi/a_0$ ($a_0$ is the distance between layers) which almost corresponds to the period of oscillations in expression (2). We also point out that the averaging procedure (9) almost does not change the values of TMR amplitude in that particular case. For other Cu growth direction, $k_F$ is smaller than $\pi/a_0$, resulting in a much steeper decay of TMR amplitude versus Cu thickness. Consequently, different Cu growth directions due to different deposition conditions may explain the discrepancy in the results obtained by various groups concerning the effect of introducing Cu at the interface between Co and $Al_2O_3$.[3,4] We also point out that as for the oscillatory coupling in ($F$/Cu) multilayers, where $F$ is a ferromagnetic transition metal,[2] changing the nature of the ferromagnetic metal can change the phase of the oscillations of TMR amplitude versus the thickness of the paramagnetic layer $a$ (Fig. 1). Due to the aliasing effect, this can lead to a very significant change not on the rate of decay in the TMR amplitude versus $a$ but on the TMR amplitude itself.

We also considered the case where Al is introduced as the paramagnetic layer. This is actually a rather common experimental situation since the Al layer initially deposited to prepare the $Al_2O_3$ (Refs. 3, 4) barrier may only be partially oxidized. In this case, the TMR amplitude decreases very rapidly and disappears for only a few monolayers (Fig. 4).

Finally, the second term in expression (2) (proportional to $\Gamma^{\uparrow}$) shows that the interfacial scattering assists the tunneling. This effect was discussed, for example, in Refs. 10 and 11 about semiconductor structures comprising a double barrier. However, since the scattering potentials $\delta_A$ and $\delta_B$ are not spin dependent in our model [Eq. (5)], this second term does not give a significant contribution to the TMR amplitude. It actually tends to decrease the relative TMR amplitude by increasing a spin-independent part in the conductivity.

In conclusion, we proposed a model to explain the relatively slow decay in tunnel magnetoresistance in Co/Cu/$Al_2O_3$/NiFe magnetic junctions as a function of the thickness of the Cu layer. This slow decay is explained in terms of the aliasing effect by the particular shape of the Cu Fermi surface in the (111) direction for which $k_F = \pi/a_0$. For Cu in other growth directions or for other paramagnetic metals inserted between the ferromagnetic electrode and the barrier, the decay is expected to be much more rapid, in agreement with experimental observations. It would be interesting to check experimentally that the different decay rates in the TMR amplitude in Co/Cu/$Al_2O_3$/NiFe junctions as a function of the Cu layer thickness are associated with various growth directions of the Cu layer, the slowest decay being associated with the largest Cu (111) texture.

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