Quantum oscillation of magnetoresistance in tunneling junctions with a nonmagnetic spacer

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We make a theoretical study of the quantum oscillations of the tunneling magnetoresistance (TMR) as a function of the spacer layer thickness. Such oscillations were recently observed in tunneling junctions with a nonmagnetic metallic spacer at the barrier-electrode interface. It is shown that momentum selection due to the insulating barrier and conduction via quantum well states in the spacer, mediated by diffusive scattering caused by disorder, are essential features required to explain the observed period of oscillation in the TMR ratio and its asymptotic value for thick nonmagnetic spacer.

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Large magnetoresistance observed in ferromagnetic tunneling junctions such as Fe/Al₂O₃/Fe and Co/Al₂O₃/CoFe currently attracts much interest due to the possibility of its application to magnetic sensors and MRAM elements. Because the tunneling magnetoresistance (TMR) ratio is related to the spin polarization of the ferromagnetic leads, attempts have been made to fabricate junctions with more highly spin-polarized ferromagnets. Realistic calculations, on the other hand, have given much higher TMR ratios than the observed values, which is probably due to their assumption of epitaxial structures. Recent experiments on TMR using epitaxial junctions, however, were unsuccessful in producing TMR ratios as high as expected. Thus, our understanding of the relationship between the electronic structure of the ferromagnets and the TMR ratio is far from complete.

The most important factor governing the TMR ratio may be the electronic structure at junction interfaces. In order to clarify its role, several experiments have been performed to measure the dependence of TMR ratio on the thickness of a nonmagnetic metal layer inserted at the interface. The observed TMR ratios show almost monotonic decrease with increasing thicknesses of inserted layers of Au, Cu, or Cr, contrary to a theoretical study for clean junctions which shows clear oscillations of the TMR ratio as a function of the nonmagnetic layer thickness. Zhang and Levy have successfully explained this decrease in TMR ratio in terms of the decoherence of electron propagation across a nonmagnetic layer. However, recent experiments by Yuasa et al. show clear oscillations of the TMR ratio as a function of Cu layer thickness for high quality NiFe/Al₂O₃/Cu/Co junctions in which the Co/Cu electrode is a single crystal. In their experiments, two characteristic features of the oscillations have been observed: (i) the average TMR ratio decays to zero with increasing nonmagnetic layer thickness; (ii) the period of the oscillations is determined solely by the perpendicular Fermi wave vector kₚ of Cu. The observed period agrees quite well with that of the oscillations of photoemission spectra caused by quantum well states in Co/Cu multilayers. From the theoretical point of view this is confusing, since in addition to the Fermi wave vector, another wave vector, i.e., the cutoff k-point kₚ, given by the depth of the quantum well, is also known to contribute to the conductance oscillations. This wave vector dominates the predicted oscillations of CPP-GMR in a Co/Cu/Co trilayer. In fact, the calculated oscillations of TMR for a clean junction cannot be explained by a single period determined by kₚ only. Furthermore, the asymptotic value of the TMR ratio calculated for a thick spacer layer is finite, which disagrees with the observed results. The purpose of the present work is to reconcile the theoretical results with the observed ones and thus deepen our understanding of the TMR effect.

In this Letter we will show that the combined effects of barrier thickness and disorder can explain the experimental results. In particular, we will demonstrate that i) increasing barrier thickness increases the amplitude of the kₚ oscillation period relative to the kₚ oscillation period, ii) disorder introduced in the barrier also weakens the amplitude of the kₚ oscillation period, and iii) the disorder decreases the asymptotic value of the TMR ratio. These results are interpreted in terms of the momentum selection of electrons incident on the barrier interface and in terms of the diffusive scattering due to disorder which opens additional conduction channels via quantum well states. In the first part of this Letter, these effects will be demonstrated by numerical calculation for a single-orbital tight-binding model. We will then demonstrate
that the calculated results can be reproduced by the stationary phase approximation [24]. This implies that this technique is applicable to a realistic multi-orbital tunneling junction, where a purely numerical calculation would be unfeasible. The results indicate that \( k_F \) of Cu is really responsible for the oscillation period observed.

Let us consider a FM/I/NM/FM junction on a simple cubic lattice with lattice spacing \( a \), where FM, I, and NM denote a ferromagnetic electrode, an insulating barrier, and a nonmagnetic metallic spacer, respectively. Initially we adopt a single-orbital tight-binding Hamiltonian in order to model a Co/Al\(_2\)O\(_3\)/Cu/Co junction:

\[
H = -t \sum_{\langle i,j \rangle, \sigma} c_{i \sigma}^\dagger c_{j \sigma} + \sum_{i, \sigma} V_{i \sigma} c_{i \sigma}^\dagger c_{i \sigma},
\]

where \( c_{i \sigma}^\dagger, c_{i \sigma} \) is the annihilation (creation) operator of an electron with spin \( \sigma \) at site \( i \), \( t \) the hopping integral between nearest neighbor sites, and \( V_{i \sigma} \) the on-site potential for an electron with spin \( \sigma \) at site \( i \). Since the majority (+) spin band of Co is similar to the Cu band, we assume that \( V_{\text{FM}+} \) is equal to \( V_{\text{NM}} \). Figure 1 shows the potential profile of the system. Quantum well states are formed in NM only for electrons with minority (−) spin in the right FM. Since the insulating Al\(_2\)O\(_3\) barrier is amorphous in real junctions, we introduce disorder in the barrier by requiring that \( V_{\text{NM}} \) takes \( V_i + \Delta V \) or \( V_i - \Delta V \) values randomly depending on the site in the barrier.

The Kubo formula and a recursive Green’s function method are used to calculate the tunneling conductances \( G_{++}, G_{--}, G_{+-}, \) and \( G_{-+} \), where \( G_{++} \) and \( G_{--} \) are the conductances in parallel alignment for \( \uparrow \) and \( \downarrow \)-spin electrons, respectively, and \( G_{-+} \) and \( G_{+-} \) are those in antiparallel alignment for \( \uparrow \) and \( \downarrow \)-spin electrons, respectively. The conductance is given by

\[
G_{\sigma \sigma'} = \frac{e^2}{h} \sum_{\mathbf{k}_i, \mathbf{k}'_i} t_{\sigma \sigma'}(\mathbf{k}_i \rightarrow \mathbf{k}'_i),
\]

where \( t_{\sigma \sigma'}(\mathbf{k}_i \rightarrow \mathbf{k}'_i) \) is the transmission coefficient for an electron incident from the left FM with \( \mathbf{k}_i \) and scattered to the right FM with \( \mathbf{k}'_i \). TMR ratio is evaluated from the conductances in the parallel and antiparallel alignments as \( TMR \equiv 1 - (G_{+-} + G_{-+})/(G_{++} + G_{--}) \).

In order to treat the disorder introduced in the insulating barrier, we use the single-site coherent potential approximation (CPA). The vertex correction to the conductance, which describes diffusive scattering, is calculated consistently with the coherent potential (self-energy) so that the current conservation is satisfied [24]. We have also performed numerical simulations [25] for finite-size clusters and checked that the results obtained by the two methods agree.

In these numerical calculations, we use \( V_{\text{FM}+} = V_{\text{NM}} = 2.382t \), \( V_{\text{FM}−} = 5.382t \), and Fermi energy \( E_F \) = 0.0. The choice of these parameters gives commensurate periods of oscillation as shown below. As for the insulating barrier, parameters \( V_i = 9.0t \) and \( \Delta V = 0 \) are used for clean junctions, and \( V_i = 9.0t \) and \( \Delta V = 0.5t \) are used for disordered junctions. We only show the calculated results for disorder within the insulating barrier. However, we have checked that the results are not changed qualitatively even when we introduce disorder at the interface between the nonmagnetic spacer and the ferromagnetic electrode.

Figures 2(a) and 2(b) show, respectively, the spin-dependent conductances and TMR ratios of junctions without disorder. It can be seen that \( G_{--} \) and \( G_{+-} \) oscillate with the NM layer thickness \( L_{\text{NM}} \) due to interference effects caused by the quantum well. These oscillations show more than one period. The Fermi wave vector \( k_F \) and the cut-off k-point \( k_\text{cp} \) of NM are given by \( 2t \cos(k_Fa) = V_{\text{NM}} - E_F - 4t \) and \( 2t \cos(k_\text{cp}a) = V_{\text{VM}−} - V_{\text{NM}} \), respectively [26]. Therefore, the periods of oscillation estimated from \( k_F = 4\pi/5a \) and \( k_\text{cp} = 2\pi/3a \) are 5\( a \) and 3\( a \), respectively. The periods of oscillation in \( G_{--} \) and \( G_{+-} \) shown in Fig. 2(a) may be interpreted as a superposition of these two periods as discussed later. The situation is analogous to that of CPP-GMR in a Co/Cu/Co trilayer [27]. The TMR ratio shown in Fig. 2(b) oscillates with the same periods as the conductance and has a finite asymptotic value for large NM thicknesses. These results are consistent with the previous results [17] where the effect of disorder was ignored.

Figure 3 shows the dependence of oscillations in \( G_{--} \) on the barrier thickness \( L_1 \). Here the conductances are normalized to the asymptotic values \( G_{--}^\infty \) obtained for \( L_{\text{NM}} \rightarrow \infty \). It can be seen that the oscillation period tends to 5\( a \) with increasing barrier thickness. This result is explained as follows. The wave vector \( k_F \) parallel to the interface is conserved in the system without disorder, that is, \( t_{\sigma \sigma'}(\mathbf{k}_i \rightarrow \mathbf{k}_i) = t_{\sigma \sigma'}(\mathbf{k}_i) \delta_{\mathbf{k}_i, \mathbf{k}_i} \). The transmission coefficient depends strongly on the angle of incidence of electrons tunneling across the barrier, and the normal incidence contributes most to the conductance. It follows that, as the barrier thickness increases, the oscillation given by cut-off k-points, i.e., \( k_F \neq 0 \), becomes progressively weakened compared to that given by the Fermi wave vector of NM, i.e., \( k_F = 0 \). As for \( G_{+-} \), we could not see the increase in oscillation period from 3\( a \) to 5\( a \) unless we increase \( L_1 \) further. This might be due
to the fact that $t_{+-}(k_{||} = 0)/\sum_{k_{||}} t_{+-}(k_{||})$ is smaller than $t_{--}(k_{||} = 0)/\sum_{k_{||}} t_{--}(k_{||})$: that is, the contribution of normal incidence to the conductance in $G_{+-}$ is less than that in $G_{--}$. As a result, the oscillation period of the TMR ratio is not quite 5$a$ for the present barrier thickness.

We now introduce disorder into the insulating barrier and show the calculated results of the spin-dependent conductances and TMR ratios in Figs. 4(a) and 4(b), respectively. It can be seen that the conductance $G_{+-}$ is enhanced by disorder whereas the other conductances $G_{++}, G_{--},$ and $G_{-+}$ are hardly affected. $G_{+-}$ now oscillates almost exclusively with period 5$a$, (i.e. $k_{p}$ period) about $G_{++}$ (see Fig. 4(a)). This results in a TMR ratio which is decreased and oscillates around zero with period 5$a$. This should be contrasted with the ordered case in which the TMR ratio oscillates with a mixed period about a constant background (cf. Fig. 4(b) and Fig. 2(b)). The asymptotic values of the TMR ratio as $L_{NM} \to \infty$ are shown in the inset of Fig. 4(b) as functions of the barrier thickness. Both the cases with and without disorder are shown. The asymptotic value of the TMR ratio of junctions without disorder decreases slowly with increasing $L_{1}$, whereas that of junctions with disorder decreases rapidly and becomes zero for large $L_{1}$.

To gain a better understanding of the effects of disorder on the magnitude of $G_{+-}$, and on the period of oscillations, we have calculated the dependence of the transmission coefficient on $k_{||}$. Figures 5(a) and 5(b) show the transmission coefficients $T_{+-}(k_{||})$, where $T_{\sigma\sigma'}(k_{||}) \equiv \sum_{k_{j}} t_{\sigma\sigma'}(k_{||} \to k'_{||})$ is the transmission coefficient for an electron incident from the left FM with momentum $k_{||}$ on the barrier without and with disorder, respectively. When there is no disorder, the contribution to $T_{+-}$ in the momentum space is concentrated near $k_{||} = (0, 0)$. However, inclusion of disorder gives rise to additional contributions to $T_{+-}$ of momenta outside this area. This is due to the fact that $k_{||}$ need not be conserved in diffusive scattering. In the absence of disorder, only $k_{||}$ points on the Fermi surfaces, that satisfy the $k_{||}$ conservation, may contribute to the conductance. For diffusive scattering, on the other hand, the entire set of $k_{||}$ points on the Fermi surface contributes to the transport.
picted by solid curves in fig. 4(a) and 4(b), are in excellent
determination of disorder, as scattering cannot open
this hypothesis, we use the stationary phase method (see Ref. 23 and references therein), which is able to deter-
magnitude 
\[ G \]
from states in the region of \( k \) states contributing to \( T_{+\pm} \) is weakened by disorder. The
other transmission coefficients are not greatly affected by the disorder increases the conductance in antiparallel alignment by opening new conductance channels via quantum well states and results in the oscillation of the TMR ratio around an averaged value close to zero. The success of the stationary phase approximation in reproducing the numerical results indicates that the oscillation period observed in realistic tunneling junctions can be explained in terms of \( k_F \) of Cu spacer.

In summary, the period of oscillation determined by \( k_F \) of the spacer is dominant in TMR due to the \( k|| \)-selection by the insulating barrier and the deconfinement of the quantum well states by disorder. The diffusive scattering caused by the disorder increases the conductance in antiparallel alignment by opening new conductance channels via quantum well states and results in the oscillation of the TMR ratio around an averaged value close to zero. The success of the stationary phase approximation in reproducing the numerical results indicates that the oscillation period observed in realistic tunneling junctions can be explained in terms of \( k_F \) of Cu spacer.

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More precisely, \( k|| \) points corresponding to these quantum well states contribute to the conductance. These \( k|| \) points appear as spiky peaks in Fig. 5(b) and they fall on concentric rings in the \( k|| \) space.

It is clear from Fig. 5(b) that the number of open \( k|| \) channels contributing to \( T_{+\pm} \) is the same as that contributing to \( T_{+\pm} \). This explains the increase in the constant part of the conductance \( G_{++} \) to a value of approximately \( G_{++} \). In addition, the introduction of diffusive scattering has almost eliminated the sharp momentum cut-off observed in fig. 5(a), which explains why the \( k_{cp} \) oscillation period of \( G_{++} \) is weakened by disorder. The other transmission coefficients are not greatly affected by the introduction of disorder, as scattering cannot open new \( k|| \) channels for these cases. This explains why the introduction of disorder has little affect on the conductances \( G_{++}, G_{+-}, \) and \( G_{-+} \).

We therefore expect that in the presence of disorder, the oscillatory part of the conductance is derived entirely from states in the region of \( k|| = (0, 0) \). In order to check this hypothesis, we use the stationary phase method (see Ref. 23 and references therein), which is able to determine the oscillatory contributions from isolated regions of the Brillouin zone, for thick spacers. The results, depicted by solid curves in fig. 4(a) and 4(b), are in excellent agreement with the numerical calculations for \( L_{NM} > 5 \). This fact indicates that the oscillation period observed in the experiments may be determined in the stationary phase approximation for realistic systems. The experimental finding [13] that the oscillation period is determined by \( k_F \) of Cu spacer is thus naturally explained. Realistic calculation for the TMR oscillation and its bias dependence is in progress.

In summary, the period of oscillation determined by \( k_F \) of the spacer is dominant in TMR due to the \( k|| \)-selection by the insulating barrier and the deconfinement of the quantum well states by disorder. The diffusive scattering caused by the disorder increases the conductance in antiparallel alignment by opening new conductance channels via quantum well states and results in the oscillation of the TMR ratio around an averaged value close to zero. The success of the stationary phase approximation in reproducing the numerical results indicates that the oscillation period observed in realistic tunneling junctions can be explained in terms of \( k_F \) of Cu spacer.

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