

Quantum oscillations of tunneling magnetoresistance in magnetic tunnel junctions

Jun Yang,^{1,2} Jun Wang,¹ Z. M. Zheng,¹ D. Y. Xing,^{1,*} and C. R. Chang³

¹National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China

²Institute of Sciences, PAL University of Science and Technology, Nanjing, 210007, China

³Department of Physics, National Taiwan University, Taipei 106, Taiwan

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Taking into account a tunneling current with ballistic and diffusive components, we study the tunneling magnetoresistance (MR) in FM/I/NM/I/FM double tunnel junctions where FM is the ferromagnet, NM the normal metal, and I the insulating barrier. The ballistic component results in oscillations of the MR with a single period, while the diffusive one leads to their decay with thickness of the NM layer. It is shown that the experimental results observed in NiFe/Al₂O₃/Cu/Co junctions by Yuasa, Nagahama, and Suzuki [Science **297**, 234 (2002)] are intrinsic features, which can be reproduced by the present calculations.

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Spin-dependent tunneling and large tunneling magnetoresistance¹⁻³ (TMR) in magnetic tunnel junctions FM/I/FM [where FM is the ferromagnetic metal (electrode) and I the insulating layer (tunnel barrier)], have recently attracted much interest due to their wide applications in spintronics. Magnetic double tunnel junctions with the more complicated structure FM/I/A/I/FM have more interesting physics, where A stands for a normal metal (NM),⁴⁻⁷ or a semiconducting,⁸ or a superconducting layer.^{9,10} For such a double-barrier structure, if region A between two tunnel barriers is a few nanometers in length, it is a typical mesoscopic system and coherent tunneling plays an important role in the mesoscopic transport. For thickness L of region A shorter than the mean free path ℓ_p of electrons due to elastic scattering, the coherent transport is in the ballistic regime; while for L longer than ℓ_p , the transport is in the diffusive regime. Adding the spin degree of freedom to conventional charge-based transport in mesoscopic systems will result in additional effects and applications.

For a FM/I/NM/I/FM double tunnel junction, theories predicted an oscillation of the TMR effect as a function of the NM layer thickness because the spin polarization of the tunneling electron oscillates as a result of the resonant tunneling.⁴⁻⁶ In early experiments,¹¹⁻¹⁴ however, the observed TMR ratios of the magnetic double tunnel junctions showed an almost monotonic decrease with NM layer thickness, no oscillation of the TMR being observed. Such a monotonic decrease of TMR has been explained by Zhang and Levy¹⁵ in terms of decoherent electron propagation across the NM layer. Recently, Yuasa *et al.*¹⁶ performed an elegant experimental study in high-quality NiFe/Al₂O₃/Cu/Co junctions and observed clear oscillations of the TMR, indicating a spin-polarized resonant tunneling. Three characteristic features of the oscillations were reported: (i) the TMR oscillation is well fitted by a damped oscillation function with an exponential decay; (ii) there is zero average value of the oscillating TMR so that the sign of the TMR ratio alternates; (iii) the single period of the TMR oscillations with Cu thickness is determined by $2\pi/q_1$ where q_1 is the scattering vector for the Cu quantum-well states along the (001) direction. Only a few theories have been proposed to explain these features observed in the experi-

ment. Itoh *et al.*¹⁷ stressed the combined effects of barrier thickness and disorder, and suggested that the disorder in the barrier decreases the asymptotic value of the TMR ratio to zero. Mu *et al.*¹⁸ supposed a relatively complicated barrier structure for the Al₂O₃/Cu interface, in which the width of the main barrier was assumed proportional to the NM layer thickness L so that the oscillations of the tunneling conductance decay in amplitude with increasing L . Recently, the exponential decay of the TMR oscillation was attributed to the finite mean free path of electrons in the NM by Itoh *et al.*¹⁹

In this paper we propose that the tunneling current in the system includes ballistic and diffusive components. In the presence of scattering processes in the NM, only a fraction of the electrons transmit ballistically while the remainder get scattered inside the well and effectively leak out of the ballistic stream. The ballistic component of the spin-polarized electrons results in the oscillations of the TMR, while the diffusive component leads to their decay with increasing L . The zero average value of TMR oscillations is found to stem from the high asymmetry in barrier strength in the present composite junction: a tunnel junction in one side and an Ohmic-contact one in the other side. It is an intrinsic feature of a FM/I/NM/FM junction, independent of properties of the potential barrier. Even though the simplest δ -type barrier is used, the zero average value of TMR will be reproduced. If both junctions are of tunnel type, the average value of oscillations of the TMR will be finite. In order to clarify the physical origin of the experimental results, we do not want to make more complicated band calculations for the realistic system. Instead, we wish to capture essential physical factors and perform a model calculation as simple as possible. It is found from our calculations that, for parallel (P) and antiparallel (AP) magnetization configurations, the oscillations of tunneling conductances have the same period and the same average value, but different amplitudes, making the sign of the TMR alternate and the average value vanish. Further, this result can be analytically verified under the assumption that the transmission probability of the left FM/I/NM junction is much smaller than that of the right NM/FM Ohmic contact. This assumption is undoubtedly reasonable in the NiFe/Al₂O₃/Cu/Co junctions.

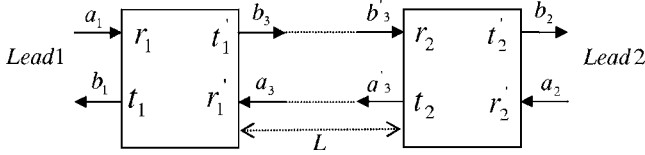


FIG. 1. Two potential barriers as shown are placed in series with coherence distance L . The problem is to find the S matrix of the composite structure.

We first consider the ballistic tunneling of electrons for current flow through an FM/I/NM/I/FM double-barrier structure. As shown in Fig. 1, for the left barrier, let a_1 and a_3 indicate the incoming wave amplitudes from both sides, and b_1 and b_3 the outgoing wave amplitudes. They satisfy the following relation:

$$\begin{pmatrix} b_1 \\ b_3 \end{pmatrix} = \begin{pmatrix} r_1 & t_1' \\ t_1 & r_1' \end{pmatrix} \begin{pmatrix} a_1 \\ a_3 \end{pmatrix}, \quad (1)$$

where r_1 and r_1' are the reflection amplitudes for the left barrier while t_1 and t_1' are the corresponding transmission amplitudes. For the right barrier, similarly, we have

$$\begin{pmatrix} a_3' \\ b_2 \end{pmatrix} = \begin{pmatrix} r_2 & t_2' \\ t_2 & r_2' \end{pmatrix} \begin{pmatrix} b_3' \\ a_2 \end{pmatrix}. \quad (2)$$

Since the electron transport in the NM is ballistic and the electronic wave vector remains unchanged, $b_3(a_3)$ is related to $b_3'(a_3')$ by a phase difference, i.e.,

$$b_3' = b_3 \exp(ik_N L), \quad a_3' = a_3 \exp(-ik_N L), \quad (3)$$

where k_N is the perpendicular component of the Fermi wave vector of electrons in the NM, along the (001) direction. It is straightforward to eliminate $a_3, a_3', b_3,$ and b_3' from Eqs. (1)–(3) to obtain

$$\begin{pmatrix} b_1 \\ b_2 \end{pmatrix} = \begin{pmatrix} r & t' \\ t & r' \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}, \quad (4)$$

where $t = t_1 t_2 \exp(ik_N L)/D$, $t' = t_1' t_2' \exp(ik_N L)/D$, $r = r_1 + t_1 t_1' r_2 \exp(i2k_N L)/D$, and $r' = r_2' + r_1' t_2' t_2 \exp(i2k_N L)/D$ with $D = 1 - r_1' r_2 \exp(i2k_N L)$. If the tunnel barrier has bilateral symmetry, we have $t_i = t_i'$ and $r_i = r_i'$ with $i=1,2$. In this case, squaring $|t_i|^2 = T_i$ and $|r_i|^2 = R_i$, we obtain the transmission probability in the ballistic regime,²⁰

$$T^c(\theta) = \frac{T_1 T_2}{1 + R_1 R_2 - 2\sqrt{R_1 R_2} \cos \theta}, \quad (5)$$

and the reflection probability $R^c(\theta) = 1 - T^c(\theta)$, where $\theta = q_1 L$ with q_1 the scattering vector of quantum states for electrons confined in the (001) direction. Both $T^c(\theta)$ and $R^c(\theta)$ are oscillating functions of θ , where the relation $\cos(2k_N L) = \cos(K - q_1)L = \cos(q_1 L)$ has been used with K the reciprocal lattice vector in the (001) direction. It was pointed out by Yuasa, Nagahama, and Suzuki,¹⁶ and will also be shown below, that conduction electrons with wave vector \mathbf{k} normal to the tunnel barrier ($\mathbf{k}_{\parallel} = \mathbf{0}$) make the main contributions to the tunneling current in an ideal tunnel junction. The oscillation period of the conductance, regardless of the rela-

tive orientation of the magnetizations of the two FM electrodes, is equal to $2\pi/q_1$. The oscillation is around an average value, which is determined by averaging T^c in an oscillation period, yielding $\langle T^c(\theta) \rangle = T_1 T_2 / (1 - R_1 R_2)$. If $T_1 \ll T_2$, we have $\langle T^c(\theta) \rangle \approx T_1$.

We wish to point out that the average of the oscillating transmission probability $T^c(\theta)$ in the ballistic regime is approximately equal to the constant transmission probability T^s in the diffusive regime. Since the diffusive scattering may cause a change in parallel wave vector k_{\parallel} , we introduce the averaging transmission probability $\bar{T}_1(\bar{T}_2)$ for the left (right) barrier as the average of $T_1(T_2)$ over k_{\parallel} , the latter being a function of k_{\parallel} . The average reflection probability is given by $\bar{R}_1 = 1 - \bar{T}_1$ ($\bar{R}_2 = 1 - \bar{T}_2$). T^s can be approximately obtained by summing the probabilities for electronic transmission with zero reflection, with two reflections, with four reflections, and so on: $T^s = T_1 \bar{T}_2 + T_1 \bar{T}_2 \bar{R}_1 \bar{R}_2 + T_1 \bar{T}_2 \bar{R}_1^2 \bar{R}_2^2 + \dots$,²⁰ yielding

$$T^s = \frac{T_1 \bar{T}_2}{1 - \bar{R}_1 \bar{R}_2}. \quad (6)$$

Alternatively, Eq. (6) can be obtained by remaking Eqs. (1)–(4). In Eqs. (1), (2), and (4), each component of the column matrices is redefined as an incoming or outgoing electron flow, and the S -matrix element s_{ij} is replaced by either average reflection or transmission probability $|s_{ij}|^2$. Instead of Eq. (3), electron flow continuity conditions in the NM yield $a_3' = a_3$ and $b_3' = b_3$. Equation (6) then follows. Such a semiclassical result is the same as Eq. (59) of Ref. 21.

In what follows we consider a combination of the ballistic with the diffusive transport. For a fraction of the current due to the diffusive transport, an electron first tunnels into the NM and then, after changing its wave vector due to the elastic scattering, tunnels out of the NM. Introducing the mean free path ℓ_p of electrons in the NM and taking care to insert a factor $\exp(-L/\ell_p)$,²⁰ we then obtain

$$T^c = \frac{T_1 T_2 \exp(-2L/\ell_p)}{1 + R_1 R_2 \exp(-4L/\ell_p) - 2\sqrt{R_1 R_2} \exp(-2L/\ell_p) \cos \theta}, \quad (7)$$

$$R^c = \frac{R_1 + R_2 \exp(-4L/\ell_p) - 2\sqrt{R_1 R_2} \exp(-2L/\ell_p) \cos \theta}{1 + R_1 R_2 \exp(-4L/\ell_p) - 2\sqrt{R_1 R_2} \exp(-2L/\ell_p) \cos \theta}. \quad (8)$$

In this case, $T^c + R^c < 1$, so that $T_1^s = 1 - T^c - R^c$ is the scattering probability in the NM region. As shown in Fig. 2, T_1^s is just the diffusive transport part of arriving at the right NM/FM interface. Evidently, T_1^s increases with L . In the large- L limit, $T^c = 0$, $R^c = R_1$, and $T_1^s = 1 - R_1$, corresponding to a completely diffusive case. It then follows that the diffusive transport part of the whole double-barrier structure is given by $T^s = T_1^s \bar{T}_2 / (1 - \bar{R}_1 \bar{R}_2)$. The total tunneling probability is the sum of the ballistic and the diffusive components, yielding

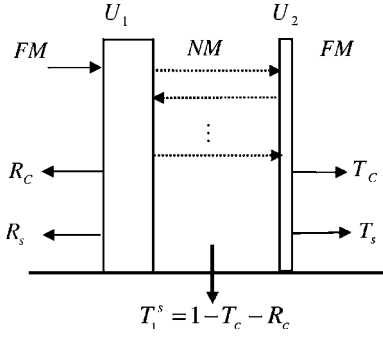


FIG. 2. Scattering processes in the NM cause electrons to leak out of the ballistic stream and result in the diffusive component of current in a FM/I/NM/I/FM double tunnel junction.

$$T = T^c + \frac{(1 - T^c - R^c)\bar{T}_2}{1 - \bar{R}_1\bar{R}_2}. \quad (9)$$

Equation (9) together with Eqs. (7) and (8) is a general result for the tunneling probability in the double-barrier structure. It is not only suitable to various bilateral-symmetric barriers, but also readily extended to other asymmetric barriers. The tunneling probability T should be spin dependent in magnetic tunnel junctions; its spin dependence is not explicitly given in Eq. (9) and will be discussed below.

For a FM/NM tunnel junction with a rectangular potential barrier, the tunneling probability of an electron at the Fermi level in the majority-spin ($\sigma = \uparrow$) or minority-spin ($\sigma = \downarrow$) band is given by²²

$$T_{i\sigma}(k_{\parallel}) = \frac{4k_{F_{i\sigma}}k_N\kappa^2}{(\kappa^2 + k_{F_{i\sigma}}^2)(\kappa^2 + k_N^2)\sinh^2(\kappa b) + \kappa^2(k_{F_{i\sigma}} + k_N)^2}, \quad (10)$$

with $i=1$ and 2 . Here $k_{F_{i\uparrow(\downarrow)}}$ is the perpendicular component of the electronic Fermi wave vector with majority (minority) spin in the FM, $\kappa = \sqrt{2m(U - E_F) + k_{\parallel}^2}$ with U the barrier height, and b the barrier width. We choose a free-electron model for a NiFe/Al₂O₃/Cu/Co junction, similar to that used in Ref. 17. In the present model, however, no disorder effect is taken into account in the tunnel barrier, and NiFe and Co have different exchange energies Δ_1 and Δ_2 , respectively. The potential profile of the model system is shown in Fig. 3, where Γ is the energy difference in the band bottom between the majority-spin band of NiFe and the Cu band. If Γ is taken to be zero and $\Delta_1 = \Delta_2$, the band model will be exactly the same as that in Ref. 17. From Fig. 3, it follows

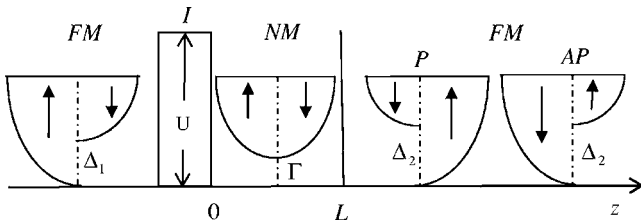


FIG. 3. Schematic representation of energy bands and potential profile in the P and AP alignments in a FM/I/NM/FM junction.

that $k_{F_{i\uparrow}} = \sqrt{2mE_F - k_{\parallel}^2}$, $k_{F_{i\downarrow}} = \sqrt{2m(E_F - \Delta_i) - k_{\parallel}^2}$, and $k_N = \sqrt{2m(E_F - \Gamma) - k_{\parallel}^2}$. For the left NiFe/Al₂O₃/Cu tunnel junction, since $\kappa b \gg 1$, Eq. (10) reduces to

$$T_{1\sigma}(k_{\parallel}) = \frac{16k_{F_{1\sigma}}k_N\kappa^2 \exp(-2\kappa b)}{(\kappa^2 + k_{F_{1\sigma}}^2)(\kappa^2 + k_N^2)}. \quad (11)$$

For the right Cu/Co Ohmic contact, we have

$$T_{2\sigma}(k_{\parallel}) = \frac{4k_{F_{2\sigma}}k_N}{(k_{F_{2\sigma}} + k_N)^2}. \quad (12)$$

Evidently, $T_{1\sigma}(k_{\parallel})$ is much smaller than $T_{2\sigma}(k_{\parallel})$ and $T_{1\sigma}(k_{\parallel})$ is maximal at $k_{\parallel} = 0$. From the two-channel current model, the tunneling conductance is the sum of those in the two spin channels and depends on the magnetization configuration of the two FM electrodes. For the P configuration, $G_P = G_{\uparrow\uparrow} + G_{\downarrow\downarrow}$; while for the AP configuration, $G_{AP} = G_{\uparrow\downarrow} + G_{\downarrow\uparrow}$, with

$$G_{\sigma\sigma'} = \frac{e^2 k_{F\sigma}^2}{(2\pi)^2} \int_0^{\phi_c} d\phi T_{\sigma\sigma'}(\phi) \cos \phi \sin \phi. \quad (13)$$

Here ϕ is the angle between the electronic wave vector and the barrier normal, and ϕ_c is the critical incident angle of an electron with spin σ in the FM electrode. To guarantee all the wave vectors ($k_{F_{i\sigma}}$ and k_N) appearing in the integral to be real variables, we get $\phi_c = \sin^{-1}(k_{\sigma, \min}/k_{F_{1\sigma}})$ where $k_{\sigma, \min}$ is the minimum among the electronic wave vectors for the spin- σ channel in the three regions: the left FM, the middle NM, and the right FM. Taking the spin-up channel for example, $k_{F_{1\sigma}} = k_{F_{1\uparrow}}$, $k_{\sigma, \min}$ is the minimum among $k_{F_{1\uparrow}}$, k_N , and $k_{F_{2\downarrow}}$ ($k_{F_{2\downarrow}}$) in the P (AP) alignment. According to Eqs. (7)–(9), T is a function of T_1, R_1, T_2 , and R_2 , but their spin dependences are not explicitly given there. In consideration of spin dependence of $T_{\sigma\sigma'}$, T_1 and R_1 in Eqs. (7)–(9) should be replaced by $T_{1\sigma}$ and $R_{1\sigma}$, respectively, and T_2 and R_2 replaced by $T_{2\sigma'}$ and $R_{2\sigma'}$. The TMR ratio is given by $(G_P - G_{AP})/G_{AP}$.

From Eq. (13) together with Eqs. (7)–(10), we have evaluated numerically the conductance as a function of thickness of the NM layer for P and AP magnetization configurations. The parameters used in the present calculations are as follows: $E_F = 4.2$ eV, $\Gamma = 0$, and $\ell_p = 1.5$ nm for the Cu layer, $\Delta_1/E_F = 0.42$ and $\Delta_2/E_F = 0.82$ for the NiFe and Co electrodes, respectively, and $U_1 = 5$ eV and $b = 2$ nm for the Al₂O₃ barrier. Figure 4 shows a rapid drop of the transmission probability $T_{\sigma\sigma'}$ with incident angle ϕ of the electron. It is found that $T_{\sigma\sigma'}$ reduces its magnitude by a factor of almost 10, respectively, at $\phi = 0.23$ for the spin-up channel and at $\phi = 0.32$ for the spin-down channel, indicating that $T_{\sigma\sigma'}(\phi)$ in the integrand in Eq. (13) may be approximately replaced by $T_{\sigma\sigma'}(\phi = 0)$. This numerical result may be understood by the following analytic argument. According to Eq. (11), the transmission probability is proportional to $\exp(-2\kappa b)$, which is approximately equal to $\exp(-2\kappa_0 b) \exp(-\beta \sin^2 \phi)$ where $\kappa_0 = \kappa(k_{\parallel} = 0) = \sqrt{2m(U - E_F)}$, $\beta = \kappa_0 b E_F / (U - E_F)$ for the spin-up channel and $\beta = \kappa_0 b (E_F - \Delta_1) / (U - E_F)$ for the spin-down channel. The transmission probability decreases exponentially with $\sin^2 \phi$. The same argument has been made^{16,23,24} that the conduction electrons with wave vector \mathbf{k}

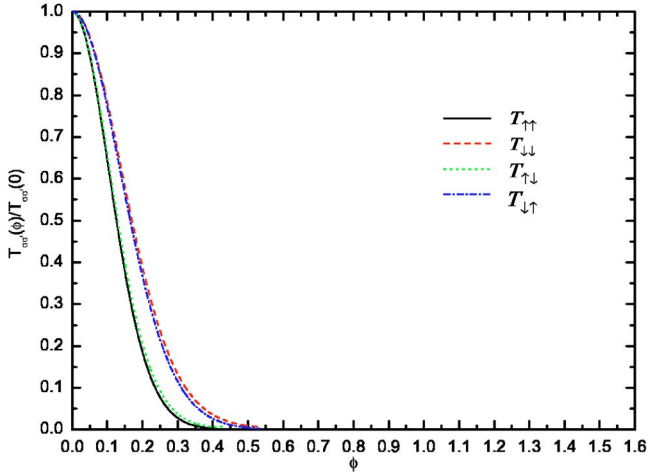


FIG. 4. (Color online) Transmission probability $T_{\sigma\sigma'}$ as a function of incident angle ϕ of electrons.

normal to the barrier ($\mathbf{k}_{\parallel}=\mathbf{0}$) make the main contributions to the tunneling current. This is the reason why $\cos\theta = \cos 2k_N L$ in Eqs. (5), (7), and (8) can be replaced by $\cos q_1 L$, resulting in the single period $2\pi/q_1$ of the TMR oscillations.

Figure 5 shows the L dependence of the TMR ratio, which reproduces essential features observed in the experiment.¹⁶ It is easily understood that the TMR oscillations arise from the ballistic conduction of electrons and quantum interference in the NM, while the decay comes from the existence of ℓ_p and the contribution of the diffusive transport. The alternation of the positive and negative TMR can be understood by the fact that the tunneling conductances for the P and AP configurations have the same oscillation period, but the oscillating amplitude for the former is greater than that for the latter, as shown in Fig. 6. As far as the zero average of TMR oscillations is concerned, we examine the average transmission probability in the ballistic regime. The average transmission probability $\langle T^c(\theta) \rangle = T_1 T_2 / (1 - R_1 R_2)$ may be rewritten as

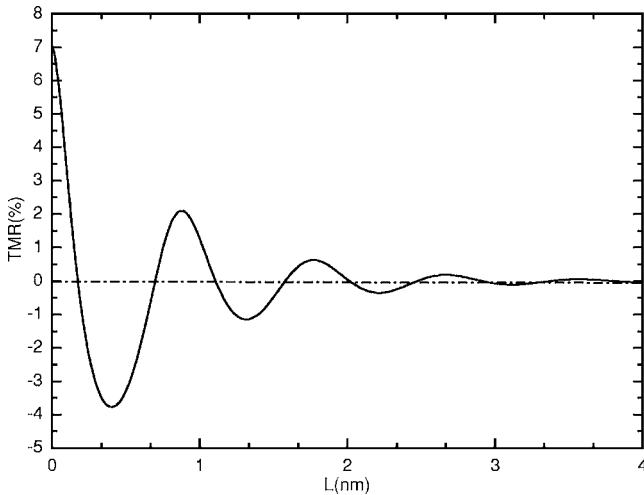


FIG. 5. The TMR ratio as a function of thickness of the NM layer (solid line). In the diffusive case, the TMR vanishes as shown by the dashed line.

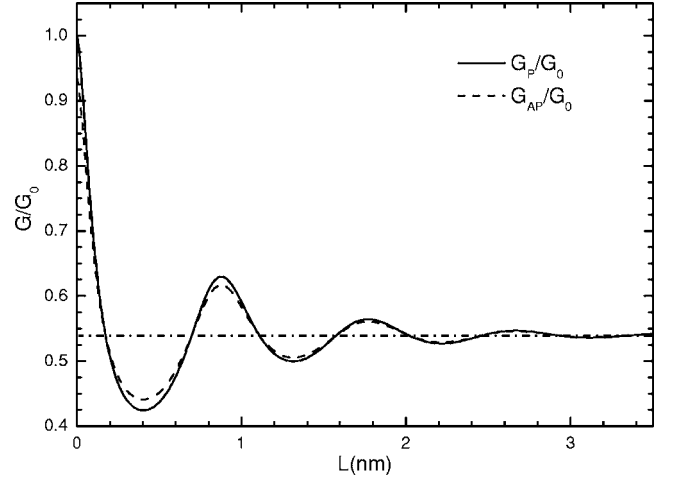


FIG. 6. Tunneling conductance as a function of thickness of the NM layer for P and AP magnetization configurations with $G_0 = G_P(L=0)$. In the diffusive case, $G_P = G_{AP}$ is constant, as shown by the horizontal line.

$$\langle T_{\sigma\sigma'}^c(\theta) \rangle^{-1} = T_{1\sigma}^{-1} + T_{2\sigma'}^{-1} - 1. \quad (14)$$

Since $T_{1\sigma}^{-1} \gg T_{2\sigma'}^{-1} \gg 1$ in the present system, we have $\langle T_{\sigma\sigma'}^c(\theta) \rangle \approx T_{1\sigma}$. It depends only on $T_{1\sigma}$ of the NiFe/Al₂O₃/Cu junction, but is independent of $T_{2\sigma'}$, indicating that $G_{\sigma\sigma'}$ does not depend on whether the magnetization configuration is P or AP. It then follows that the average of the coherent TMR oscillations is zero, which stems from the highest asymmetry of two potential barriers. For a FM/I/NM/I/FM double tunnel junction with higher symmetry, there will be a finite average value of the TMR oscillations. The present mechanism is quite different from that in Ref. 17. In that work the key point is the presence of disorder in the barrier, which breaks the \mathbf{k}_{\parallel} conservation in tunneling processes and induces new conductance channels via quantum-well states for the AP alignment. The new conductance channels increase the conductance of the AP alignment to approximately that of the P alignment so that the average MR ratio decreases to almost zero.

Finally, we wish to briefly discuss the decaying behavior of the TMR oscillation amplitude by comparing the present approach with that in Ref. 17. To explain the experimental data of TMR oscillations,¹⁶ the diffusive scattering has been taken into account in both the approaches. The main difference between them is that the diffusive scattering was assumed within the tunnel barrier in Ref. 17, while it is in the NM in the present work. Such a difference gives rise to qualitatively different results for decaying oscillations of the TMR. For diffusive scattering in the barrier, the TMR oscillations decay inversely proportional to L ; while in the present work, they decay exponentially as $\exp(-2L/\ell_p)$, much faster than $1/L$. The present result is similar to that in a recent theory of Itoh *et al.*,¹⁹ both of them being consistent with the experimental data.¹⁶ In the present approach to the diffusive transport, we have made the approximation of replacing the k_{\parallel} -dependent $T_1(T_2)$ with its average $\bar{T}_1(\bar{T}_2)$ over k_{\parallel} . Although this approximation is very suitable for the

present system of $T_1 \ll T_2$, its improvement would be necessary for a complete theory, which merits further study.

In summary we have presented a theoretical approach to the tunneling conductance and TMR in magnetic double tunnel junctions with both ballistic and diffusive components. The former results in oscillations of the MR with single period $2\pi/q_1$, while the latter leads to their decay exponentially with thickness of the NM layer. The average value of MR oscillations depends to a great extent on the two poten-

tial barriers. For the present FM/I/NM/FM junction with high asymmetry, the average tends to zero. The single period arises from the fact that the normal incidence ($k_{\parallel}=0$) dominates the tunneling conductance as long as the potential barrier is wide enough.

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*Email address: dyxing@nju.edu.cn

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