Tunneling magnetoresistance in mixed-valence manganite tunnel junctions

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An effective tunneling Hamiltonian including spin-flip effect is proposed to account for the tunneling magnetoresistance (TMR) in mixed-valence manganite tunnel junctions and the current-perpendicular-to-plane TMR in layered manganite crystals. It is found that the electron spin-flip tunneling plays an important role in diminishing the amplitude of the TMR ratio. The theoretical results are in agreement with the recent experimental observations in the magnetic trilayer junction structure $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.67}Sr_{0.33}MnO_3$ and the bulk crystals $La_{1.4}Sr_{1.6}Mn_2O_7$. It is suggested that reducing electron spin-flip tunneling is a potential way to gain larger TMR in the manganite tunnel junctions. [S0163-1829(98)02225-5]

The recent discovery of colossal magnetoresistance (CMR) in Mn oxides $La_{1-x}A_xMnO_3$ with $A=$Sr, Ca, or Ba (Ref. 1) has triggered renewed interest in these perovskites.[2–6] The properties of $La_{1-x}A_xMnO_3$ are usually explained by a simple double-exchange (DE) mechanism.[7–9] In order to completely understand the transport phenomena, the Jahn-Teller electron-phonon coupling,[4] complex hopping amplitude and electron-electron interaction,[5] and nonmagnetic randomness[6] have been proposed as necessary extensions of the DE mechanism.

In the interesting doping range $0.2\lesssim x\lesssim 0.5$, $La_{1-x}A_xMnO_3$ is a metallic ferromagnet at low temperatures, where it is associated with the simultaneous presence of Mn^{3+} and Mn^{4+}. The Mn^{3+} ions have three electrons in the t_{2g} state forming a local $S=3/2$ spin, and one electron in the e_g state which hops between nearest-neighbor Mn ions, with double occupancy suppressed by the strong Hund coupling between the local spin and the itinerant e_g electron. The widely used Hamiltonian containing this physics is

$$\mathcal{H}=-t\sum_{\langle ij\rangle\sigma}(c^{\dagger}_{i\sigma}c_{j\sigma}+\mathrm{H.c.})-J\sum_i \mathbf{S}_i\cdot\sigma_i\,, \tag{1}$$

where the first term represents the e_g electron transfer between nearest-neighbor Mn ions at sites i and j, while the second term stands for the Hund coupling between the $S=3/2$ localized spin $\mathbf{S}_i$ and the mobile electron with spin σ_i with $J>0$ and $J\gg t$.

It is well known that the strong Hund coupling dominates the basic physics of the mixed-valent manganite systems. The e_g electron spin combines with the located spins to form two configurations of the total spin: $S+\frac{1}{2}$ and $S-\frac{1}{2}$, respectively, with energies $-JS$ and $J(S+1)$. In sufficiently large J limit, the $S-\frac{1}{2}$ configuration and the doubly occupied state are forbidden, and there only exists the $S+\frac{1}{2}$ configuration. Also, the large J means that the hopping of an e_g electron from the Mn^{3+} to the Mn^{4+} ions is sensitive to the relative orientation of their located spins. An important fact should be noted that in the mangnites the conduction electrons are almost fully spin polarized well below the critical temperature, which is quite different from ordinary ferromagnetic transition metals such as Fe, Co, and Ni.[10]

Recently reducing the field scale of the magnetotransport in manganites has been a major goal of research motivated by the latent application of the CMR effect for devices. One approach is to incorporate the metallic ferromagnet $La_{1-x}A_xMnO_3$ into magnetic tunnel junctions.[11,12] The epitaxial tunnel junction has been fabricated in the form of $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.67}Sr_{0.33}MnO_3$, and a large tunneling magnetoresistance (TMR) as high as 83% was observed at low magnetic fields of only tens of Oe at 4.2 K.[11] Another approach is to synthesize the layered manganite crystal $La_{2-2x}Sr_{1+2x}Mn_2O_7$,[13,14] in which the ferromagnetic-metallic MnO_2 bilayers are separated by nonmagnetic $(La,Sr)_2O_2$ insulating layers, forming a virtually infinite array of ferromagnet/insulator/ferromagnet junctions. In the low-temperature case at 4.2 K, the current-perpendicular-to-plane (CPP) resistivity drastically decreases in the low magnetic field region during the magnetization process, and becomes constant when the field exceeds its saturation value about 5 kOe. The field-sensitive tunneling process gives rise to a low-field (<1kOe) CPP TMR as large as 240%.[13] Very recently, it was reported that the magnitude of this TMR can be drastically enhanced up to $\sim$4000% by applying pressure of $\sim$10 kbar at 4.2 K.[15]

Previous spin-polarized tunneling models[16–18] are suitable to the magnetic tunnel junctions where two metallic electrodes are the ferromagnetic transition metals.[19] Due to the almost full spin polarization in the manganites, the tunneling probability in a manganite tunnel junction is much more sensitive to the relative alignment of the moments of two ferromagnetic electrodes than that in ordinary magnetic tunnel junctions. Therefore, it is highly desired to develop a tunneling theory suitable for the manganite tunnel junctions by taking into account the basic physics of the manganites. It is found that the electron spin-flip tunneling plays an important role in determining the amplitudes of TMR. Its consideration will be conducive to the explanation of TMR observed both in the $La_{1-x}A_xMnO_3$ tunnel junctions and in the layered manganite crystals.

Consider a magnetic tunnel junction of which both electrodes are the ferromagnetic-metallic $La_{1-x}A_xMnO_3$ layers with perovskite structure. Electron tunneling removes an electron with spin σ from a Wannier state at Mn site j of one side and creates an electron in a Wannier state at Mn site i of the other side with spin σ for spin conservation or $-\sigma$ for spin flipping. As a generalization of the DE Hamiltonian (1), the tunneling Hamiltonian describing such an electron tunneling process is

$$H_{ij}=\sum_{\sigma}\left(T_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}+iT'_{ij}c_{i\sigma}^{\dagger}c_{j,-\sigma}+\text{H.c.}\right)-J(\mathbf{S}_i\cdot\sigma_i+\mathbf{S}_j\cdot\sigma_j), \tag{2}$$

where T_{ij} and T'_{ij} are the spin-conserving and spin-flip tunneling matrix elements, respectively, and the Hund coupling terms are special for the manganite electrodes.

In order to derive an effective tunneling Hamiltonian, the semiclassical approach[8] is applied to Eq. (2). Take the local spin quantization axis at Mn site $i(j)$ to be along $\mathbf{S}_i(\mathbf{S}_j)$. The electron states with spins parallel and antiparallel to $\mathbf{S}_i$ are labeled with α (spin up) and β (spin down), respectively, and those parallel and antiparallel to $\mathbf{S}_j$ labeled with α' (spin up) and β' (spin down). The eigenstates on Mn site $i(j)$ are $\mathbf{S}_i\alpha$ and $\mathbf{S}_i\beta(\mathbf{S}_j\alpha'$ and $\mathbf{S}_j\beta')$. The spinor transformation is well known,

$$\begin{pmatrix}\alpha\\ \beta\end{pmatrix}=\begin{pmatrix}\cos\left(\frac{\theta}{2}\right) & \sin\left(\frac{\theta}{2}\right)\\ -\sin\left(\frac{\theta}{2}\right) & \cos\left(\frac{\theta}{2}\right)\end{pmatrix}\begin{pmatrix}\alpha'\\ \beta'\end{pmatrix}, \tag{3}$$

where θ is the angle between $\mathbf{S}_i$ and $\mathbf{S}_j$. We can write down the secular equation $|H_{ij}-E|=0$ with H_{ij} given by

$$\begin{array}{c|cccc} & \mathbf{S}_i\alpha & \mathbf{S}_i\beta & \mathbf{S}_j\alpha' & \mathbf{S}_j\beta' \\ \hline \mathbf{S}_i\alpha & -JS & 0 & T_{ij}\cos\frac{\theta}{2}+iT'_{ij}\sin\frac{\theta}{2} & T_{ij}\sin\frac{\theta}{2}+iT'_{ij}\cos\frac{\theta}{2} \\ \mathbf{S}_i\beta & 0 & J(S+1) & -T_{ij}\sin\frac{\theta}{2}+iT'_{ij}\cos\frac{\theta}{2} & T_{ij}\cos\frac{\theta}{2}-iT'_{ij}\sin\frac{\theta}{2} \\ \mathbf{S}_j\alpha' & T_{ij}\cos\frac{\theta}{2}-iT'_{ij}\sin\frac{\theta}{2} & -T_{ij}\sin\frac{\theta}{2}-iT'_{ij}\cos\frac{\theta}{2} & -JS & 0 \\ \mathbf{S}_j\beta' & T_{ij}\sin\frac{\theta}{2}-iT'_{ij}\cos\frac{\theta}{2} & T_{ij}\cos\frac{\theta}{2}+iT'_{ij}\sin\frac{\theta}{2} & 0 & J(S+1) \end{array}. \tag{4}$$

The secular equation reduces to

$$\left\{\left(\frac{J}{2}-E\right)^2-\left[J\left(S+\frac{1}{2}\right)+A_{ij}(\theta)\right]^2-B_{ij}^2(\theta)\right\}\left\{\left(\frac{J}{2}-E\right)^2-\left[J\left(S+\frac{1}{2}\right)-A_{ij}(\theta)\right]^2-B_{ij}^2(\theta)\right\}=0,$$

with

$$A_{ij}(\theta)=|T_{ij}\cos(\theta/2)+iT'_{ij}\sin(\theta/2)| \tag{5}$$

and $B_{ij}(\theta)=|T_{ij}\sin(\theta/2)+iT'_{ij}\cos(\theta/2)|$. In the strong Hund coupling case ($J\gg T_{ij}$ and $J\gg T'_{ij}$), we can ignore the eigenvalues with spin down having very high energies and obtain the approximate eigenvalues with spin up as $E\simeq -JS \pm A_{ij}(\theta)$. It then follows that Eq. (2) can be replaced by the effective Hamiltonian $H_{ij}^{\text{eff}}=[T_{ij}\cos(\theta/2)+iT'_{ij}\sin(\theta/2)]d_i^{\dagger}c_j+\text{H.c.}$ where $d_i(c_j)$ is the electron operator with spin parallel to the local quantization axis $\mathbf{S}_i(\mathbf{S}_j)$. Such an effective Hamiltonian includes both the spin-conserving and spin-flip tunneling. Owing to the strong Hund couling and the DE conduction of electrons, each of two manganite electrodes is a ferromagnet with almost full spin polarization and all the local spins on each manganite electrode are parallel to each other at zero temperature. As a result, for a manganite tunnel junction, the total tunneling Hamiltonian can be written as

$$H_{\text{T}}=\sum_{ij}\{[T_{ij}\cos(\theta/2)+iT'_{ij}\sin(\theta/2)]d_i^{\dagger}c_j+\text{H.c.}\}, \tag{6}$$

where the angle θ is the relative orientation between the moments of the two manganite electrodes.

In the momentum representation, the system Hamiltonian is given by $H=H_{\text{L}}+H_{\text{R}}+H_{\text{T}}$ where $H_{\text{L}}=\Sigma_{\mathbf{p}}\varepsilon_{\mathbf{p}}c_{\mathbf{p}}^{\dagger}c_{\mathbf{p}}$ and $H_{\text{R}}=\Sigma_{\mathbf{k}}\varepsilon_{\mathbf{k}}d_{\mathbf{k}}^{\dagger}d_{\mathbf{k}}$ are the spin-up electron Hamiltonians in the left and right electrodes of the manganite tunnel junction, respectively, and

$$H_{\text{T}}=\sum_{\mathbf{k,p}}\{[T_{\mathbf{kp}}\cos(\theta/2)+iT'_{\mathbf{kp}}\sin(\theta/2)]d_{\mathbf{k}}^{\dagger}c_{\mathbf{p}}+\text{H.c.}\} \tag{7}$$

is the tunneling Hamiltonian. The electrons that participate in the tunneling current have their wave vectors very near the Fermi wave vectors $\mathbf{k}_F$ and $\mathbf{p}_F$. It is an adequate approximation to treat the transfer rates $|T_{\mathbf{kp}}|^2$ and $|T'_{\mathbf{kp}}|^2$ as their average values $|T|^2$ and $|T'|^2$, respectively. Using the standard Green's function method,[20] the θ-dependent tunneling conductance $G(\theta)$ is obtained at zero bias as

$$G(\theta)=2\pi e^2N_{\text{L}\uparrow}(\varepsilon_{\text{F}})N_{\text{R}\uparrow}(\varepsilon_{\text{F}})|T|^2[\cos^2(\theta/2)+\gamma\sin^2(\theta/2)], \tag{8}$$

where $N_{\text{L}\uparrow}(\varepsilon_{\text{F}})$ and $N_{\text{R}\uparrow}(\varepsilon_{\text{F}})$ are the densities of states of spin-up electrons at Fermi energies on the left and right sides of the tunneling junction, respectively. We have also intro-

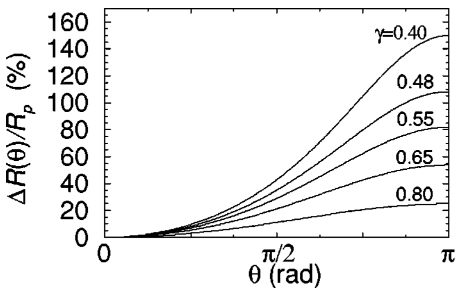

FIG. 1. Angle dependence of the TMR ratio for several values of γ in manganite tunnel junctions.

duced a parameter $\gamma = |T'|^2/|T|^2$ to character the spin-flip effects in the tunneling process. This parameter is usually smaller than unity and increases with temperature. A tunnel junction can be regarded as a simple resistor,[20] with the resistance given by $R(\theta) = 1/G(\theta)$.

We first focus our attention on the TMR at low temperatures. It is well known that there exist two different definitions for the TMR ratio: $\Delta G(\theta)/G_p = [G(0) - G(\theta)]/G(0)$ (Refs. 16,18), and $\Delta R(\theta)/R_p = [R(\theta) - R(0)]/R(0)$.[11,13,15] They satisfy the simple relation $[\Delta G(\theta)/G_p]^{-1} - [\Delta R(\theta)/R_p]^{-1} = 1$. Using expression (8) for the tunneling conductance, we obtain the TMR for the $La_{1-x}A_xMnO_3$ tunnel junction as

$$\frac{\Delta G(\theta)}{G_p} = (1-\gamma)\sin^2\left(\frac{\theta}{2}\right), \tag{9}$$

$$\frac{\Delta R(\theta)}{R_p} = \frac{1}{\cos^2(\theta/2) + \gamma\sin^2(\theta/2)} - 1. \tag{10}$$

In what follows we shall use Eq. (10) to discuss the TMR in the manganite tunnel junctions and the CPP TMR of the layered manganite crystals, since the definition of $\Delta R(\theta)/R_p$ was used in those experimental works.[11,13,15] Figure 1 shows $\Delta R(\theta)/R_p$ as a function of θ for several γ's. The greater $\Delta R(\theta)/R_p$, the smaller γ and the greater θ in the range of $0 \leqslant \theta \leqslant \pi$. As $\theta = \pi$, corresponding to the case of both moments of two electrodes antiparallel to each other, there is a maximum TMR ratio, $(\Delta R/R_p)_{\max} = (1-\gamma)/\gamma$. According to this formula, one gets an estimate of $\gamma \simeq 0.55$ for experimental data $(\Delta R/R_p)_{\max} = 83\%$ in the $La_{0.67}Sr_{0.33}MnO_3$ tunnel junction at 4.2 K.[11] If the spin-flip tunneling was not taken into account ($\gamma = 0$), one would get an infinite $(\Delta R/R_p)_{\max}$. As a result, the spin-flip electron tunneling appears to play an important role in diminishing the amplitude of TMR. This suggests that the effective spin-diffusion length in defect-populated $SrTiO_3$ barrier is much shorter than the width of the barrier. The spin-flip tunneling may arise from the impurity states (Mn ions) inside $SrTiO_3$ barrier and the spin-flip scattering events at the interface between $SrTiO_3$ and $La_{0.67}Sr_{0.33}MnO_3$. This indicates that reducing the spin-flip effect is a potential way to obtain higher TMR ratio in the manganite tunnel junctions. At finite temperatures, increasing temperature will make the probability of spin-flip tunneling increase so as to decrease the maximum TMR ratio. In addition, the temperature effect will result in orientation fluctuations of local spins in two manganite electrodes, further decreasing the TMR ratio. The argument above may account qualitatively for the experimental result that the TMR decreases with increasing temperature.[11]

Next, we turn our attention to the interplane TMR in layered manganite crystal $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.3$, which is viewed as being composed of ferromagnetic-metallic MnO_2 bilayers with intervening nonmagnetic insulating $(La,Sr)_2O_2$ barriers. Since metallic resistances of the MnO_2 bilayers can be ignored, the CPP resistance of the system is equal to the sum of tunneling resistances of n tunnel junctions in series connection, i.e., $R = \Sigma_{i=1}^{n} R(\theta_i)$, where θ_i is the angle between the moments of two nearest-neighbor MnO_2 bilayers separated by the ith insulating barrier. In the low-temperature case, the moments of the MnO_2 bilayers are essentially parallel within domains separated by domain boundaries lying on the $(La,Sr)_2O_2$ layers.[13] Recent experiments indicate that the low-temperature phase of this system may consist of mostly antiferromagnetic and some ferromagnetic static order between the adjacent MnO_2 bilayers.[15] Suppose that there exist m domain boundaries and the angle between the moments of the nearest-neighbor domains is a constant θ.[13] The CPP resistance is given by $R = mR(\theta) + (n-m)R(0)$ with $R(\theta) = 1/G(\theta)$. At satuated fields, all moments of the MnO_2 bilayers are aligned toward the field direction so that $R_p = nR(0)$. It then follows that

$$\frac{\Delta R(\theta)}{R_p} = \eta\left(\frac{1}{\cos^2(\theta/2) + \gamma\sin^2(\theta/2)} - 1\right), \tag{11}$$

where $\eta = m/n$ is smaller than unity. If $\theta = \pi$, as suggested in Ref. 13, one obtains the maximal CPP TMR ratio as

$$\left(\frac{\Delta R}{R_p}\right)_{\max} = \frac{\eta(1-\gamma)}{\gamma}. \tag{12}$$

For layered crystals of $La_{2-2x}Sr_{1+2x}Mn_2O_7$, the spin flipping during electron tunneling may take place at domain boundaries due to electron-magnon scattering.[21] It follows that at low temperatures the spin-flip effects in the layered crystals are much weaker than those in the epitaxial $La_{1-x}A_xMnO_3$ tunnel junctions. As a result, γ in Eq. (12) should be very small, yielding an extremely high ratio of the interlayer TMR. For example, taking $\eta = 0.8$ and $\gamma = 0.02$, we have $(\Delta R/R_p)_{\max} \simeq 4000\%$, which is compatible with the experimental data of Ref. 15. We wish to point out that the incoherent tunneling model under consideration here is very appropriate to the CPP TMR in the layered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7 (x = 0.3)$ under high pressure. This is because the applied pressure can weaken the interlayer coupling and a two-dimentional-like conduction of nearly fully spin polarized carries within the magnetically interplane-decoupled MnO_2 bilayers is highly diffuse or incoherent.[15]

If the angle between the moments of the nearest-neighbor domains is assumed to distribute randomly in the range $0 \leqslant \theta \leqslant \pi$, the CPP tunneling resistance at zero field is

$$R = \frac{m}{\pi}\int_0^{\pi} R(\theta)\,d\theta + (n-m)R(0) = \left(\frac{m}{\sqrt{\gamma}} + n - m\right)R(0). \tag{13}$$

In this case, the CPP TMR ratio is given by

$$\frac{\Delta R}{R_p} = \eta\left(\frac{1}{\sqrt{\gamma}} - 1\right). \tag{14}$$

It is easily seen that for a given η, the required value of γ in Eq. (14) is much smaller than that in Eq. (12) to fit the same CPP TMR data.

In summary we have proposed an effective tunneling Hamiltonian including the spin-flip effect to account for the tunneling magnetoresistance in the manganite tunnel junctions. The effective tunneling Hamiltonian was derived by incorporating the basic physics of the manganites in tunneling theory, from which a formula for the tunneling conductance was obtained. The TMR ratio for $La_{1-x}A_xMnO_3$ tunnel junctions and the CPP TMR ratio for layered crystals $La_{2-2x}Sr_{1+2x}Mn_2O_7$ were evaluated at low temperatures. It was found that the electron spin-flip tunneling plays an important role in diminishing the amplitudes of the TMR. Using reasonable parameters, we got good agreement between theoretical and experimental results.

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