Magnetoresistance of Ferromagnetic Tunnel Junctions in the Double-Exchange Model

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We conduct a theoretical study of the temperature dependence of the spin polarization (P) and the magnetoresistance (MR) ratio using the double exchange (DE) model for ferromagnetic tunnel junctions with half-metallic systems. It is shown that the strong exchange coupling in the DE model plays an important role in the temperature dependence of both P and the MR ratio; their values can be less than the maximum values expected for half-metallic systems at low temperatures, and the MR ratio decreases more rapidly than P with increasing temperature. The calculated results, however, indicate that the MR ratio may still be large at high temperatures near the Curie temperature.

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Magnetoresistance (MR) of ferromagnetic tunnel junctions (FTJ's) [1,2] has recently attracted much interest due to potential applications such as magnetic sensors and high density magnetic memories. A simple interpretation of the MR ratio observed at low temperatures is given by the following formula [3,4]:

$$\frac{\Delta R}{R_{\mathrm{AP}}} = \frac{R_{\mathrm{AP}} - R_{\mathrm{P}}}{R_{\mathrm{AP}}} = \frac{2P^2}{1 + P^2}, \qquad (1)$$

where R_{AP} and R_{P} are the resistances with antiparallel (AP) and parallel (P) alignments of the magnetization of the electrodes, respectively, and P denotes the spin polarization of the electrodes [5]. (Here we assume symmetric FTJ's.) The largest MR ratio is then given for the so-called half-metallic ferromagnets where $P = 1$, that is, only one spin component has nonzero density of states (DOS) at the Fermi level. The *ab initio* band calculations have predicted so far that Heusler compounds [6] and several metallic oxides such as CrO_2 [7] and (La-Sr)MnO_3 [8,9] show the half-metallicity. Therefore, it is natural to utilize these materials for electrodes of FTJ's to obtain a high MR ratio. Lu *et al.* [10] and Viret *et al.* [11] fabricated FTJ's using (La-Sr)MnO_3 to obtain MR ratios as high as 0.44 and 0.8, respectively. However, the MR ratio has been found to decrease rapidly with increasing temperature (T) and to vanish far below the Curie temperature (T_{C}). The basis for the rapid decrease in the MR ratio with T may be similar to that in the case of the intergrain MR of perovskite manganites; Park *et al.* [12], on the basis of spin resolved photoemission experiments, have suggested that the rapid decrease in the intergrain MR [13] with increasing T is due to a much stronger T dependence of P at interfaces than that of the bulk.

Furthermore, the value of P itself at low temperatures is an issue to be resolved. Wei *et al.* [14] and Park *et al.* [15] have reported evidence of the half-metallicity of perovskite manganites from scanning tunneling microscopy and spin resolved photoemission experiments, respectively. In contrast, recent measurement of P through a method involving point contact between a ferromagnet and a superconductor has shown that $P \sim 0.8$ for manganites [16,17], being consistent with the MR ratio observed [11]. Therefore, in order to clarify the characteristics of the tunnel MR of FTJ's, it is of interest to calculate the T dependence of P at interfaces of FTJ's and to study their interrelation.

It is well known that the basic physics of manganites resides in the so-called double exchange (DE) model [18], though the orbital degree of freedom and the Jahn-Teller effect in manganites are responsible for the variety of magnetic states and the colossal MR near T_{C} [19,20]. The DE model, however, contains many-body interaction and its ground state properties are not fully understood yet as shown in a recent numerical study on the DE model [21]. In view of these circumstances, here we focus our attention on how Hund's rule (exchange) coupling between the itinerant electrons and localized spins in the DE model influences the T dependence of P and the MR ratio, and we treat it in an approximate way where the exchange coupling is dealt with as spin fluctuations (SF) acting on the tunneling electrons at the interfaces. In the following, we calculate the values of P and the tunnel conductance Γ in the linear response theory (Kubo formula) [22] and study the interrelation between the MR ratio and P. We will show that the exchange coupling has an intrinsic effect on the T dependence of the MR ratio and P in these FTJ's and that the MR ratio depends more strongly on temperature than P does. Nevertheless, the MR ratio may be large enough even at high temperatures.

We consider a FTJ comprised of two semi-infinite ferromagnets (FM) separated by a thin insulator (I) on a simple cubic lattice. The Hamiltonian of the double-exchange model generalized for the FTJ is given as

$$H = -t \sum_{\langle ij\rangle\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + v \sum_{i\in \mathrm{I}\sigma} c^{\dagger}_{i\sigma} c_{i\sigma} - \frac{K}{2} \sum_{i\in \mathrm{FM}} \mathbf{S}_i \cdot \boldsymbol{\sigma}_i, \qquad (2)$$

where t and v are the nearest neighbor hopping and the potential within I, respectively, and the last term denotes the Hund's rule coupling between the localized spin $\mathbf{S}_i$ and the itinerant electron spin $\boldsymbol{\sigma}_i$ with a positive value of K. The localized spins are taken to be $S = 3/2$. The other notations are standard.

In order to treat the exchange coupling, we adopt the local approximation developed by Kubo [23] and followed later by Takahashi and Mitsui [24]. The exchange interaction is rewritten as

$$\mathbf{S}_i \cdot \boldsymbol{\sigma}_i = \sum_{\sigma} \langle S_i^z \rangle c_{i\sigma}^{\dagger} c_{i\sigma} + \sum_{\sigma\sigma'} V_i^{\sigma\sigma'} c_{i\sigma}^{\dagger} c_{i\sigma'}, \quad (3)$$

with $V_i^{\sigma\sigma} = \sigma(S_i^z - \langle S_i^z \rangle)$, $V_i^{\uparrow\downarrow} = S_i^-$, and $V_i^{\downarrow\uparrow} = S_i^+$, where $\langle \cdots \rangle$ denotes the thermal average. We treat $V_i^{\sigma\sigma'}$ perturbatively in the self-consistent Born approximation where terms such as $\langle V_i^{\sigma\sigma'} V_i^{\sigma'\sigma} \rangle$ are retained as SF acting on the itinerant electrons subject to that $\langle V_i^{\sigma\sigma'} \rangle = 0$. In this approximation, the Green's function G_{ii}^{σ} of the itinerant electrons is normalized by summing up the infinite power series of $\langle V_i^{\sigma\sigma'} V_i^{\sigma'\sigma} \rangle$ to give the self-energy as $\sum_{\sigma'} \langle V_i^{\sigma\sigma'} G_{ii}^{\sigma'} V_i^{\sigma'\sigma} \rangle$. The Green's function is thus calculated self-consistently for a given T dependence of $\langle S^z \rangle$ and SF. The self-consistency, however, is not satisfied between the magnetic state of localized spin and the electronic state of the itinerant spins; that is, the Curie temperature is taken as a given parameter.

Despite the approximation to the exchange interaction, the treatment of this term still meets a difficulty due to nonexistence of the translational invariance of the FTJ's. The difficulty may be avoided by including the effect of SF at the interfacial layers only. The approximation may be justified when we note that the tunnel MR is governed by the electronic and magnetic states at the interfaces as suggested by several experimental and theoretical results; the experiments show that the MR ratio is strongly influenced by magnon excitations [25], and theories show that the tunnel conductance is correlated with the interfacial electronic state [26] and is affected by the interfacial roughness [27]. As the SF may be considered to be thermally induced disorder, its effect on the electronic state of the tunneling electrons may be the largest at the interfaces. Although we use the bulk values of $\langle S^z \rangle$ and SF, the contribution of the SF relative to that of $\langle S^z \rangle$ can be properly included, at least, qualitatively, in our treatment.

The expression Γ given by the Kubo formula is rewritten in terms of intra- and interlayer Green's functions [22], which are calculated by the adlayering method [28]. The vertex correction is taken into account in a way which is consistent with that used for the self-energy [27]. The asymmetry of the transverse SF terms, however, breaks the current conservation in the AP alignment of the magnetization of FTJ's even though the vertex function is determined self-consistently. This fact has not been noticed in previous work [23,24]. Therefore, we apply a semiclassical approximation such that $\langle S^+ S^- \rangle = \langle S^- S^+ \rangle \sim S^2 - \langle (S^z)^2 \rangle$. This approximation may not be unreasonable except for low temperatures as the spin values S are large in our case. The current conservation has been confirmed numerically as well.

In the calculations, we chose two values of Fermi level, $E_F = -5t$ and $-4t$ (see Fig. 1 inset), $K = 2t$, the barrier height $\Phi = 2t$, and barrier thickness $d = 5a$ with a lattice constant a for a simple cubic lattice. The number of itinerant electrons is thus determined to be 0.08 and 0.16 per atom for $E_F = -5t$ and $-4t$, respectively. We have neglected the T dependence of the Fermi distribution function. A small shift of the Fermi level caused by the T dependence of the exchange splitting has been adjusted by shifting the energy bands to avoid a less interesting T dependence of the MR ratio.

Figure 1 shows the T dependence of the spin polarization $P = (D_\uparrow - D_\downarrow)/(D_\uparrow + D_\downarrow)$ at the interfaces, D_σ being the interfacial DOS for the itinerant electrons. The solid and broken curves are the calculated values of P with and without SF, respectively. When $E_F = -5t$, the bulk electrodes are half-metallic up to $\sim 0.4 T_C$, and therefore the thick broken curve shows $P = 1$ below $\sim 0.4 T_C$. When the half-metallicity is lost as the exchange splitting $\pm K \langle S^z \rangle / 2$ decreases, the value of P begins to decrease. As shown by the thick solid curve, SF makes P smaller than 1 even at low temperatures and its T dependence is stronger above $\sim 0.4 T_C$ than that of P without SF. A similar tendency can be seen in results for $E_F = -4t$. Thus, both the disappearance of the half-metallicity and the existence of SF are responsible for the strong T dependence of P. A linear T dependence at low temperatures is due to the classical approximation. A quantum treatment at low temperatures will reduce the value of P further, as discussed later.

Figure 2 shows the calculated results of the tunnel MR ratio as functions of T/T_C. When $E_F = -5t$, the MR ratio without SF (thick broken curve) takes the maximum

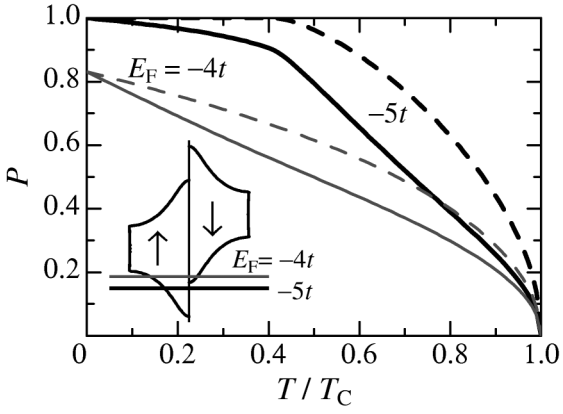

FIG. 1. Temperature dependence of P with (solid curves) and without (broken curves) spin fluctuations for $E_F = -5t$ (thick curves) and $-4t$ (thin curves). The inset shows the density of states of the bulk electrodes.

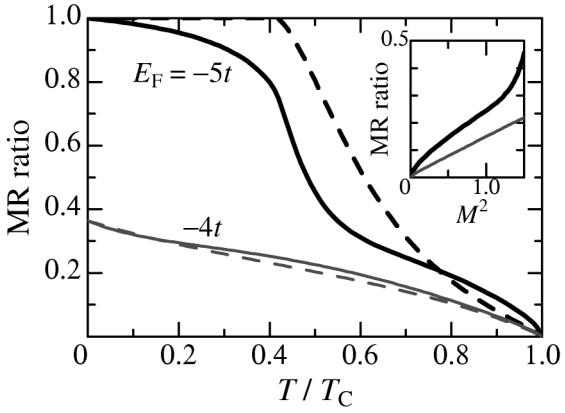

FIG. 2. Temperature dependence of the MR ratio with (solid curves) and without (broken curves) spin fluctuation for $E_F = -5t$ (thick curves) and $-4t$ (thin curves). The inset is the MR ratio as a function of the square of the magnetization M of the bulk electrodes.

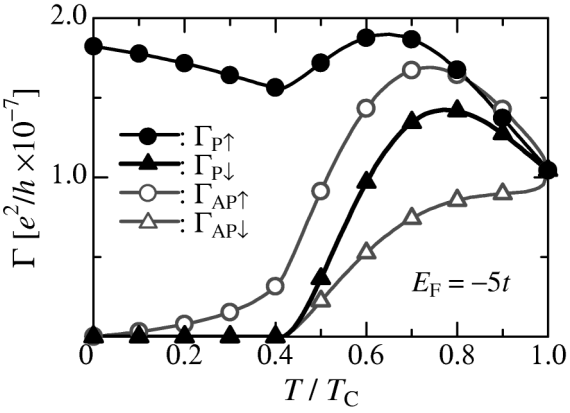

FIG. 3. Spin dependent conductances of P and AP alignments. The direction of the magnetization of the FM which electrons are incident from is chosen as the spin quantization axis.

value 1 below $\sim 0.4T_C$ and decreases rapidly above this temperature where the half-metallicity is lost. Inclusion of SF makes the MR ratio smaller than that without SF for $T \lesssim 0.7T_C$ as shown by the thick solid curve. The origin of the strong T dependence of the MR ratio for half-metallic electrodes is the same with that of the T dependence of P. In contrast, when $E_F = -4t$, the MR ratio decreases gradually with increasing temperature. A small difference between the solid and broken curves in this case may be due to a cancellation of two effects; a decrease in the MR ratio due to spin flip tunneling and an increase in the MR ratio due to diffusive tunneling [27], both of which are caused by SF.

It is worthwhile to note that the T dependence of the MR ratio for half-metallic electrodes at high temperatures is similar to that for normal ferromagnetic electrodes. This indicates that the MR ratio for FTJ's with half-metallic electrodes can be large even at high temperatures near T_C. This result is in agreement with recent experiments on FTJ's with manganites [29], where the MR ratio does not vanish even at high temperatures near T_C. The inset in Fig. 2 shows the MR ratio vs M^2 with M being the magnetization of the electrodes. We see the MR ratio $\propto M^2$ approximately. Noting that the intergrain tunnel MR in pyrochlore $Tl_2Mn_2O_7$ [13] and double perovskite $Sr_2FeMoMnO_6$ [30] shows a power law behavior, indicating that the interfacial effects on magnetism may be smaller, FTJ's with these materials might show a large MR ratio at temperatures close to T_C.

Deeper insight into the T dependence of the MR ratio can be obtained by plotting the spin dependent conductance as a function of T, as shown in Fig. 3. The increase in Γ above $T \simeq 0.4T_C$ is due to the disappearance of the half-metallicity and the decrease in Γ above $T \gtrsim 0.7T_C$ is due to SF. One should note that the SF makes $\Gamma_{P\uparrow}$ smaller with T at low temperatures, but $\Gamma_{AP\uparrow}$ increases with T in this temperature range.

The MR ratio can be estimated by using Eq. (1) with the values of P given in Fig. 1. The estimated values are the same as the MR ratio shown in Fig. 2 only qualitatively. The difference between these results originates from the treatment of Γ including vertex corrections.

The effects of spin flip tunneling due to SF on the MR ratio can be more clearly seen when we increase the transverse components of the exchange coupling K_{+-}, keeping the longitudinal component $K_z = 2t$ constant. In Fig. 4, we show the calculated results for the MR ratio where $K_{+-} = 2t$, $3t$, and $4t$ for both $E_F = -5t$ (thick curves) and $-4t$ (thin curves). We find that the decrease in the MR ratio becomes greater as K_{+-} increases, indicating that SF may reduce the power law dependence of the MR ratio on M.

In order to evaluate the MR ratio at low temperatures where the quantum effect is crucial, we first study the bulk electronic state at a low carrier density limit, adopting a variational method. We take the fully polarized ferromagnetic state of the localized spins as a vacuum $|0\rangle$. When

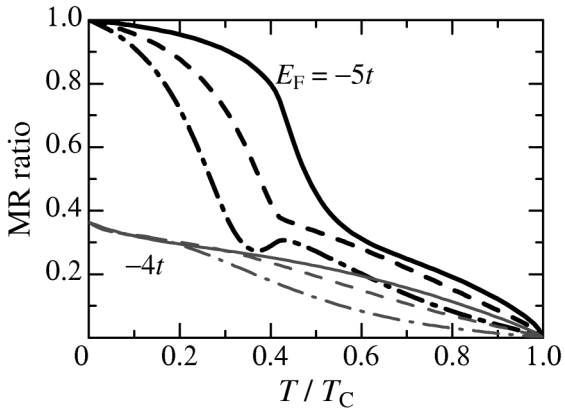

FIG. 4. MR ratio for larger values of the coupling constant in the Hund's rule coupling K_{+-}; $K_{+-} = 2t$ (solid curves), $3t$ (broken curves), and $4t$ (chained curves) for $E_F = -5t$ (thick curves) and $-4t$ (thin curves).

an up spin carrier is introduced to $|0\rangle$, the wave function with a momentum $\mathbf{k}$ is given simply as $|\mathbf{k}\uparrow\rangle = c^{\dagger}_{\mathbf{k}\uparrow}|0\rangle$. On the other hand, the wave function of a down spin electron introduced into $|0\rangle$ may be given as

$$|\mathbf{k}\downarrow\rangle \sim \left(A_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\downarrow} + \frac{1}{\sqrt{N}} \sum_{\mathbf{Q}} B_{\mathbf{k}-\mathbf{Q}} c^{\dagger}_{\mathbf{k}-\mathbf{Q}\uparrow} b^{\dagger}_{\mathbf{Q}}\right)|0\rangle , \quad (4)$$

because the down spin electron is able to couple to the magnon excitations. Here, $b^{\dagger}_{\mathbf{Q}}$ is a creation operator of a magnon with wave vector $\mathbf{Q}$. Equation (4) is the variational function for the down spin electron to minimize the energy with respect to the coefficients $A_{\mathbf{k}}$ and $B_{\mathbf{k}-\mathbf{Q}}$. By using Eq. (4), the Hamiltonian is diagonalized for 144 k points for a square lattice, for simplicity, with $K = 2t$. We find that the DOS for the down spin state hybridizes with that of the up spin state, as already pointed out by several authors [23,24].

The MR ratio in the dilute limit of the carrier density can be calculated in the following way, using the tunnel Hamiltonian method. By noting that the tunneling electron is a bare electron instead of a magnetic polaron as given in Eq. (4) and that the electron with momentum $\mathbf{k} = 0$ contributes dominantly to the tunnel conductance $\Gamma_{\mathrm{P(AP)}}$, we get $\Gamma_{\mathrm{P}} \propto 1 + |A_{\mathbf{k}=\mathbf{0}}|^2$ and $\Gamma_{\mathrm{AP}} \propto 2|A_{\mathbf{k}=\mathbf{0}}|$ leading to the MR ratio:

$$\frac{\Delta R}{R_{\mathrm{AP}}} = \frac{\Gamma_{\mathrm{P}} - \Gamma_{\mathrm{AP}}}{\Gamma_{\mathrm{P}}} = \frac{(1 - |A_{\mathbf{k}=\mathbf{0}}|)^2}{1 + |A_{\mathbf{k}=\mathbf{0}}|^2} . \quad (5)$$

The MR ratio thus calculated for $K = 2t$ is 0.83. By increasing the value of K, the MR ratio decreases, in accordance with the general trend shown in Fig. 4. One should note that no spin flip tunneling is included in this method. The decrease in the MR ratio originates from the intrinsic properties of the ground state of the double exchange model. The MR ratio thus calculated is reasonable as compared with the experimental values of P at low temperatures [11,16]. As the present treatment may be suitable in the case of pyrochlore manganites where the density of itinerant electrons is quite small, it may be interesting to measure P of these materials by the point contact method.

Summarizing, we have studied the temperature dependence of the MR ratio using the DE model for the FTJ's with half-metallic manganites in mind. It has been shown that the strong exchange coupling in the DE model plays an important role in the temperature dependence of the MR ratio as well as P; that is, both P and the MR ratio can be less than their maximum values expected for half-metallic systems at low temperatures, and they decrease rapidly around temperatures where the half-metallicity disappears. Although the T dependence of the MR ratio is similar to that of P, the MR ratio decreases more rapidly than P does with increasing temperature. The present results indicate that the MR ratio can be large enough at high temperatures near T_{C} when the exchange coupling is relatively weak. It should be noted that no spin flip tunnel matrix elements have been introduced in our framework, contrary to the work done by Lyu *et al.* [31]. Finally, we stress here the importance of the role of the exchange coupling intrinsically existing in the DE model. A self-consistency between itinerant electrons and localized spins may be a future problem. The existence of the half-metallicity at the ground state should also be examined theoretically [32].

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