Influence on tunnel magnetoresistance of spin configurations localized within insulators

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We theoretically study effects on tunnel magnetoresistance (TMR) of spin configurations localized within insulating barriers. Two cases are treated herein: interaction between a tunneling electron and an isolated classical spin being canted at an angle to the magnetization axis of the ferromagnetic leads, and interaction between a tunneling electron and many quantum spins within the insulating barrier. The characteristic features of TMR observed in δ -doped ferromagnetic tunnel junctions and in grain boundaries of metallic manganites are discussed in view of the results obtained.

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I. INTRODUCTION

The tunnel magnetoresistance (TMR) of ferromagnetic tunnel junctions (FTJ's) has attracted much interest for its potential use in technological applications.^{1,2} It is well known that TMR depends crucially on the quality of insulating barriers; magnetic impurities and/or magnetically active defects in the barrier strongly influence TMR. Recent development of fabrication techniques for samples has made it possible to control the quality of the insulating barrier. Jansen and Moodera^{3,4} have measured TMR for FTJ's where thin Fe, Co, or Ni layers are inserted within the barrier (δ doping). They have observed an enhancement of the TMR ratio for Fe doping, but decreases for Co and Ni doping. Another example of TMR being influenced by the quality of barriers may be grain boundary (GB) tunneling in manganites. GB tunneling was originally observed for polycrystalline samples of metallic manganites at low temperatures, in which TMR ratios of several 10% have been reported.^{5,6} Recent experiments have demonstrated that tunneling through a well-controlled, artificially fabricated GB brings about quite high TMR ratios.⁷ Detailed study of the voltage and temperature dependence of GB-TMR has revealed that spindependent inelastic tunneling⁸ or inelastic tunneling due to imperfection of the GB (Ref. 9) plays an important role. The magnetic-field dependence of TMR, on the other hand, suggests a second-order tunneling through GB.¹⁰

The interpretations of the characteristic behaviors of GB-TMR in manganites described above are quite reasonable, as a GB should include both structural and magnetic imperfections. In the GB of manganites, the atomic disorder prevents electrons from hopping, making the double exchange interaction^{11,12} ineffective and resulting in an insulating character of the GB. Because the localized spins of manganese ions should still exist in the GB, the tunneling electrons may interact with the localized spins. A similar phenomenon may occur in δ -doped FTJ's, while TMR of δ -doped FTJ's is at first sight different from the GB-TMR in manganites, as Fe, Co, and Ni atoms introduced into oxide barriers can be oxidized to form localized spins.

The interaction between the tunneling electrons and localized spins gives rise to spin-flip tunneling, which is known to reduce the magnitude of TMR. Spin-flip tunneling has so far been studied for systems in which the localized spins are located at or near the interfaces,^{13–17} or paramagnetic impurities.¹⁸ However, because the tunneling electron should interact with many localized spins in the GB, the extent to which the spin-flip tunneling influences the GB-TMR may still be nontrivial. As for the enhancement of TMR in δ -doped FTJ's, Jansen and Lodder¹⁹ have extended the theory of resonant tunneling^{20,21} to show that a spindependent distribution of resonant levels can afford the observed results. However, the role of a possible canting of magnetic moments of Fe, Co, or Ni within the barrier has not yet examined.

The purpose of the present work is to investigate the influence on TMR of various spin configurations of both classical and quantum spins within the insulators. We deal herein with the following two cases. The first involves an interaction of a tunneling electron with an isolated classical spin being canted at an angle in relation to the magnetization axis of the ferromagnetic leads. In view of the results obtained, a possible mechanism of enhanced TMR in Fe-doped FTJ's will be discussed. The other involves the interaction between a tunneling electron and quantum spins of finite number in the insulating barrier, which may be considered a model of GB tunneling in manganites. We will study the effects of the spin configurations and spin-flip tunneling on TMR in this case. The linear-response theory is adopted to calculate the tunnel conductance at a zero-bias limit in a one-dimensional model. The outline of the paper is as follows: The model and basic formula for tunnel conductance are given in Sec. II, TMR's for junctions with classical spins and quantum spins are treated in Secs. III and IV, respectively, and Sec. V is devoted to a summary of this paper.

II. MODEL

We adopt herein a one-dimensional tight-binding model for FTJ's consisting of two ferromagnetic metal leads separated by an insulating barrier. Localized spins exist within the barrier and interact with the tunneling electrons. A schematic figure of the system is presented in Fig. 1.

The Hamiltonian is given by

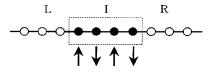


FIG. 1. Schematic figure of a one-dimensional tunnel junction treated in the present work. Arrows indicate localized spins in the barrier region I. L and R indicate the left- and right-hand side leads, respectively.

$$H = H_t + H_v + H_K + H_J. \tag{1}$$

 H_t represents electron hopping between nearest-neighbor sites and is given as

$$H_t = \sum_{ij\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma}, \qquad (2)$$

where $\sigma = \uparrow$ or \downarrow . The second term,

$$H_v = \sum_{i\sigma} v_{i\sigma} c_{i\sigma}^{\dagger} c_{i\sigma}, \qquad (3)$$

gives site- and spin-dependent energy levels $v_{i\sigma}$, which also represent the exchange splitting of the ferromagnetic leads. H_K and H_J indicate the exchange interaction between tunneling electrons and localized spins, and between localized spins within the insulating barrier, respectively. They can be written as

$$H_K = -K \sum_n \mathbf{S}_n \cdot \mathbf{s}_n \tag{4}$$

and

$$H_J = J \sum_{\langle mn \rangle} \mathbf{S}_m \cdot \mathbf{S}_n - \mu_{\rm B} \sum_n S_n^z h^{ex}.$$
 (5)

Here, **S** and **s** are the localized spin and the spin of the tunneling electron, respectively, *K* and *J* indicate the coupling between localized spins and the tunneling electron and between localized spins, respectively, and h^{ex} is the external magnetic field acting on the localized spins. The *g* factor is taken to be 2.0. The external magnetic field is introduced only to control the spin configurations of the localized spins. The Zeeman effect on the itinerant electrons is neglected. We also assume that the antiparallel (AP) alignment of the magnetization of the ferromagnetic leads can be altered to a parallel (P) alignment by an infinitesimally small magnetic field.

The tunnel conductance at the zero-bias limit can be calculated using the Kubo formula or, equivalently, a formula given by Caroli *et al.*,^{22,23}

$$\Gamma = \frac{e^2}{h^2} 2\pi^2 t^2 t'^2 \operatorname{Tr}(D_L G_I^- D_R G_I^+ + D_L G_I^+ D_R G_I^-), \quad (6)$$

where $D_{L(R)\sigma}$ is the density of states at the edge site of the L(R) lead, and $G_{I\sigma}^{+(-)}$ is a retarded (advanced) Green's function for the barrier region I. The trace includes the summation over σ , and t and t' are the hopping integrals between the *L* lead and region *I*, and between the *R* lead and region *I*, respectively.

III. TMR FOR JUNCTIONS WITH CLASSICAL SPINS

In this section, we formulate an expression of the TMR

ratio for FTJ's with a single classical spin within the barrier, which is canted at an angle θ in relation to the magnetization axis of the ferromagnetic leads. The conductance of a real FTJ may be obtained by taking an average over the distribution of θ by assuming that the real FTJ is a parallel junction formed by the present one-dimensional model.

A. Formalism of the TMR ratio

As we deal with a single spin, the H_J term in the Hamiltonian is irrelevant. We first consider a case in which $h^{ex} = 0$. The effect of the external field will be taken into consideration in the next subsection. The H_K term gives spindependent energy levels within the barrier for the tunneling electrons,

$$v_n - (+)KS/2 \equiv \epsilon_{\uparrow(\downarrow)}. \tag{7}$$

The index *n* is omitted for simplicity, as there is only one localized spin. In order to obtain an explicit expression of the TMR ratio as a function of the canting angle θ of the localized spin, we take a local spin axis parallel to the localized spin. Consequently, the expression of the conductance Γ is given by a 2 by 2 matrix equation where Green's function is expressed as

$$G_{I}^{\pm} = \begin{pmatrix} G_{\uparrow}^{\pm} \cos^{2} \frac{\theta}{2} + G_{\downarrow}^{\pm} \sin^{2} \frac{\theta}{2} & (G_{\uparrow}^{\pm} - G_{\downarrow}^{\pm}) \cos \frac{\theta}{2} \sin \frac{\theta}{2} \\ (G_{\uparrow}^{\pm} - G_{\downarrow}^{\pm}) \cos \frac{\theta}{2} \sin \frac{\theta}{2} & G_{\uparrow}^{\pm} \sin^{2} \frac{\theta}{2} + G_{\downarrow}^{\pm} \cos^{2} \frac{\theta}{2} \end{pmatrix},$$

$$(8)$$

with spin-dependent Green's functions G_{σ}^{\pm} , the expression of which will be given later, and the local density of states is given by

$$D_{\alpha} = \begin{pmatrix} D_{\alpha\uparrow} & 0\\ 0 & D_{\alpha\downarrow} \end{pmatrix}, \tag{9}$$

with $\alpha = L, R$.

By introducing spin polarization $P_{\alpha}, \alpha = L, R$, of the ferromagnetic leads as

$$D_{\alpha\pm} = (1 \pm P_{\alpha}) D_{\alpha0}, \qquad (10)$$

and taking the trace over the spin, we get

$$\Gamma = \frac{4\pi^2 e^2}{h} t^2 t'^2 D_{R0} D_{L0} [(1 + P_L \cos \theta)(1 + P_R \cos \theta)G_{\uparrow}^- G_{\uparrow}^+ + (1 - P_L \cos \theta)(1 - P_R \cos \theta)G_{\downarrow}^- G_{\downarrow}^+ + P_L P_R \sin^2 \theta (G_{\uparrow}^- G_{\downarrow}^+ + G_{\downarrow}^- G_{\uparrow}^+)], \qquad (11)$$

which is the general expression obtained for tunnel conductance.

Because we focus our attention on TMR for δ -doped FTJ's where the Fe, Co, and Ni ions are located at the middle of the barrier, the lifetime broadening of the energy level ϵ_{σ} due to intermixing with the ferromagnetic leads can be neglected. Consequently, the local Green's functions G_{σ}^{\pm} can be approximated as $G_{\sigma}^{-}G_{\sigma}^{+} \sim 1/\Delta_{\sigma}^{2}$ and $G_{\uparrow}^{-}G_{\downarrow}^{+}+G_{\downarrow}^{-}G_{\uparrow}^{+} \sim 2/\Delta_{\uparrow}\Delta_{\downarrow}$, with $\Delta_{\sigma} = \epsilon_{\sigma} - \epsilon_{\rm F}$, where the energy of Green's

function is taken as the Fermi energy $\epsilon_{\rm F}$. Here we assume $\Delta_{\sigma} > 0$ due to strong on-site Coulomb repulsion at the ionic site within the barrier.

The expression of the tunnel conductance is simplified as

$$\Gamma = \Gamma_0 \bigg[\frac{1}{\Delta_{\uparrow}^2} + \frac{1}{\Delta_{\downarrow}^2} + \frac{2}{\Delta_{\uparrow}\Delta_{\downarrow}} P_L P_R + \bigg(\frac{1}{\Delta_{\uparrow}^2} - \frac{1}{\Delta_{\downarrow}^2} \bigg) (P_L + P_R) \\ \times \langle \cos \theta \rangle + \bigg(\frac{1}{\Delta_{\uparrow}} - \frac{1}{\Delta_{\downarrow}} \bigg)^2 P_L P_R \langle \cos^2 \theta \rangle \bigg],$$
(12)

with

$$\Gamma_0 = 4 \,\pi^2 \frac{e^2}{h} t^2 t'^2 D_{L0} D_{R0}, \qquad (13)$$

where $\langle \cdots \rangle$ indicates an average over the distribution of θ .

The tunnel conductance Γ_P for P alignment of magnetization of the ferromagnetic leads is calculated by taking P_L >0 and P_R >0, and Γ_{AP} for AP alignment is calculated by choosing P_L >0 and P_R <0. The TMR ratio is defined as

MR ratio=
$$\frac{\Gamma_P - \Gamma_{AP}}{\Gamma_P}$$
. (14)

We see that when $\Delta_{\uparrow}\!=\!\Delta_{\downarrow}$, the expression of the TMR ratio is reduced to $1^{24,25}$

$$MR \text{ ratio} = \frac{2P_L P_R}{1 + P_P P_R}.$$
 (15)

In the following we study the effects of the angle distribution on the TMR ratio, because the spin-dependent distribution of the energy ϵ_{σ} has already been studied.¹⁹

B. Calculated results and discussion

First, we calculate the $\Delta_{\perp} / \Delta^{\uparrow} \equiv \delta$ dependence of TMR for a uniform distribution of θ , $-\theta_0 < \theta < \theta_0$. Calculated results of the TMR ratio as functions of δ are shown in Fig. 2 for $\theta_0 = 0.0, 0.5\pi$, and π , with $|P_L| = |P_R| \equiv P = 0.3$. We see that the TMR ratio is enhanced for $\delta > 1.0$ when the distribution of θ_0 is confined to around $\theta_0 = 0$. These results are quite reasonable, as \downarrow spin electrons are blocked from tunneling, while \uparrow spin electrons are not when $\Delta_{\perp}/\Delta_{\uparrow} > 1.0$. This is a kind of spin-filtering effect. In the case that $\Delta_{\perp}/\Delta_{\uparrow}$ > 1.0 and P > 0, the spin filtering is effective when the localized spin is nearly parallel to the magnetization of the ferromagnets, while it is ineffective when the distribution of the localized spins is isotropic. The spin-filtering effect caused by the spin-dependent energy level may be clearly understood in a schematic diagram shown in Fig. 2(b). When the up spin energy level of the impurity is closer to the Fermi energy than the down spin energy level, the spin polarization of the transmittance through the barrier becomes higher.

Second, we study the effects of local uniaxial anisotropy of the localized moments within the barrier. We assume that the direction of the localized spin is determined by the following anisotropy energy:

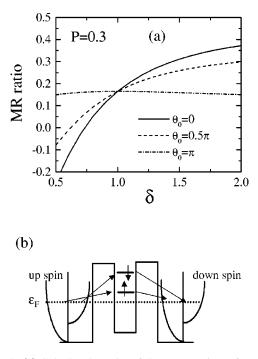


FIG. 2. (a) Calculated results of the TMR ratio as functions of $\Delta_{\downarrow}/\Delta \uparrow \equiv \delta$ with P = 0.3 and $\theta_0 = 0.0.5\pi$ and π . (b) A schematic diagram showing the spin-filtering effect due to the spin-dependent energy levels of the magnetic impurity.

$$E_a = \kappa \sin^2(\theta - \theta_a) - \mu_{\rm B} h^{ex} S \cos \theta, \qquad (16)$$

where the angle θ_a characterizes the anisotropy axis. We determine the direction of the localized spin θ by minimizing E_a for a given value of $h^{ex}S/\kappa$, assuming a uniform distribution of θ_a such that $-\theta_0 < \theta_a < \theta_0$. Figure 3 represents the calculated TMR ratios as functions of $h^{ex}S/\kappa$ for $\delta = 1.5$ and 2.0 with P = 0.3, $\theta_0 = 0.5\pi$. The MR ratio increases with increasing $h^{ex}S/\kappa$ as expected.

Last, we present calculated results of sample-to-sample fluctuations of the tunnel conductance for both P and AP alignments. The fluctuations are defined by

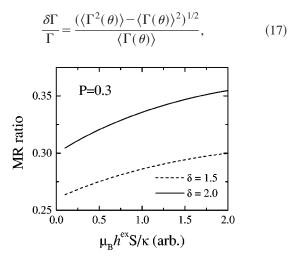


FIG. 3. Calculated results of the TMR ratio as functions of $h_{ex}S/\kappa$ with P=0.3 and $\delta=1.5$ and 2.0.

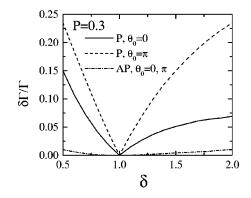


FIG. 4. Calculated results of fluctuations in conductance for parallel and antiparallel alignment of the magnetizations as functions of δ with P=0.3 and $\theta_0=0$ and π .

for both P and AP alignments. Figure 4 shows the calculated results of $\delta\Gamma/\Gamma$ as functions of δ with $\theta_0 = 0.0$ and π and P = 0.3. As can be seen from Eq. (12), there is no fluctuation at $\delta = 1.0$. Large fluctuations in the P alignment come from the linear term of $\cos\theta$. Because the linear term vanishes when $P_L = -P_R$, the fluctuations in the AP alignment are very small.

In order to compare theoretical and experimental results, one should relate Δ_{σ} with the electronic state of Fe, Co, and Ni atoms resolved into the barrier of δ -doped FTJ's in which we are interested. Because the resolved atoms may form magnetic layers with a thickness of less than one monolayer, the transition atoms could be oxidized by nearby oxygen atoms to form localized spins. If this is the case, the magnetic states of these ions depend on the number of d electrons. When this number is <5, the magnetic state may be a high-spin state, and the spin of an additional electron therefore tends to align parallel to the localized spin, which means that K is positive in the present model. In contrast, when the number of d electrons is ≥ 5 , the spin of the tunneling electron will be antiparallel to the localized spin, meaning that K is negative. Because Fe ions have the least number of delectrons among Fe, Co, and Ni, it is highly probable that Fe ions satisfy a relation of $\Delta_{\perp}/\Delta_{\uparrow} > 1.0$, while the others do not, offering a possible explanation for the enhanced TMR ratio for the δ dopant of Fe atoms in FTJ's. Jansen and Lodder¹⁹ have included a spin-dependent distribution of the energy level of impurities by extending the work done by Bratkovsky.²⁰ They showed that the spin-dependent distribution of the energy level of impurities may enhance the TMR, although the spin-independent distribution may reduce the TMR. Our work focused on the role of the spin-dependent energy level itself and the angle distribution of the localized moment. Our results may be supplemental to the works done by Jansen and Lodder.¹⁹

IV. TMR FOR JUNCTIONS WITH QUANTUM SPINS

In this section, we treat the tunneling of an electron that interacts with localized quantum spins within the barrier. Because the localized spins themselves interact with each other, one should basically treat a many-body problem to calculate the tunnel conductance. One should therefore perform nu-

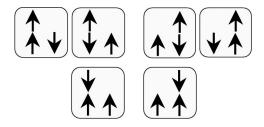


FIG. 5. An example of a basis set generated from an antiferromagntic alignment of the localized spins. Upper and lower arrows in each box indicate the spins of the tunneling electron and localized spins, respectively.

merical calculations in this case to obtain the TMR ratio instead of formulating a simple expression of the TMR ratio as in the previous section.

A. Method of calculation

We now deal with the full Hamiltonian; a uniform hopping *t* between the nearest-neighbor sites is assumed for H_t , and H_v includes uniform exchange splitting $-(+)\Delta_{ex}$ for $\uparrow(\downarrow)$ spin electrons in the ferromagnetic leads and a spin-independent potential *u* in the barrier region.

In order to treat H_K and H_J , we prepare the following many-body basis function for the tight-binding basis set,

$$\Psi_{i\sigma\mu} = c_{i\sigma}^{\dagger} |\mu\rangle, \qquad (18)$$

where i and $|\mu\rangle$ indicate the electron position and spin configurations of the localized spins in the barrier, respectively. When site *i* is on the leads, $|\mu\rangle$ represents the ground state of the localized spins determined by H_I , while there are many intermediate states when the electron is inside the barrier region because the electron creates new states via the exchange interaction. The new states can be generated by operating H to the ground state of the localized spins, keeping $s^{z} + S_{tot}^{z}$ constant where S_{tot}^{z} is the z component of the total spin angular momentum of the localized spins.²⁶ All of these new states are taken into consideration to calculate the tunnel conductance. An example of the basis set for a barrier with two localized spins is shown in Fig. 5, where there are six states generated from an antiferromagnetic alignment of two localized spins. Because the number of basis functions thus constructed increases rapidly with increases in the number of localized spins, we deal with only eight localized spins with S = 1/2 each in the present work. The tunnel conductance at the zero-bias limit is calculated by using Eq. (6) by taking the intermediate states thus constructed into consideration.

The following parameter values have been used in units of t; K=2 (ferromagnetic interaction), J=0.1 (antiferromagnetic interaction), and u=2.0 for the barrier potential. The specific choice of the sign of K and J has been made by considering the grain-boundary tunneling in manganites. The exchange field acting on the nearest-neighbor localized spins from the ferromagnetic leads is assumed to be negligibly small in the present case. The spin configuration of the localized spins in the ground state is determined by diagonalizing H_J for a given external magnetic field h^{ex} . In order to

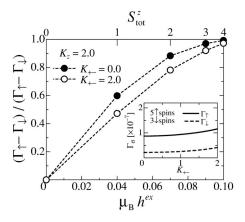


FIG. 6. Calculated results of the spin-asymmetry ratio of tunnel conductance as a function of the external magnetic field or S_{tot}^z . Inset: Tunnel conductance as a function of K_{+-} for a case where the barrier region includes five \uparrow and three \downarrow localized spins.

clarify the contribution of the spin-flip process to the tunnel conductance, we have separately treated the constant *K* for the longitudinal (K_z) and transverse (K_{+-}) components of the exchange interaction.

B. Calculated results

First, we treat systems with paramagnetic leads where the Fermi level $\epsilon_{\rm F}$ is taken to be -1.0. Figure 6 shows the calculated results of the spin-asymmetry ratio $(\Gamma_{\uparrow} - \Gamma_{\downarrow})/(\Gamma_{\uparrow})$ $+\Gamma_{\perp}$) of the tunnel conductance, where Γ_{σ} is the σ component of the tunnel conductance Γ . The inset shows the dependence of Γ_{σ} on K_{+-} for a fixed value of $K_z = 2.0$. With increases in the value of K_{+-} , the tunnel conductance increases, because the lowest energy level for the tunnel electron interacting with the localized spins decreases due to the transverse terms of the exchange interaction. The ratio (Γ_{\uparrow} $-\Gamma_{\parallel})/(\Gamma_{\uparrow}+\Gamma_{\parallel})$ plotted as a function of the external magnetic field h^{ex} (or S_{tot}^{z}) increases with increasing magnetic field. This result is quite reasonable, as the \uparrow spin electrons become more transparent due to the ferromagnetic exchange interaction with increasing alignment of the localized spins. We see that the spin-flip tunneling, i.e., the contribution from the K_{+-} term, decreases the spin-asymmetry ratio by \sim 10%. The contribution is large for intermediate values for S_{tot}^z of the localized spins.

Next we deal with systems with ferromagnetic leads. The exchange potential $\pm \Delta_{ex}$ of the ferromagnetic leads is taken to be ± 1.0 . With this choice for the exchange splitting, the bottom of the minority spin density of states is -1.0. When $h^{ex}=0$, the localized spins align antiferromagnetically so that $S_{tot}^z=0$. By changing the magnetization direction of the ferromagnetic leads, keeping the antiferromagnetic spin alignment of the localized spins unchanged, the TMR ratio defined by $(\Gamma_P - \Gamma_{AP})/\Gamma_P$ can be calculated. With further increases in h^{ex} , S_{tot}^z increases.

The TMR ratio at $h^{ex}=0$ is dependent on the position of the Fermi level $\epsilon_{\rm F}$. Figure 7 shows the dependence of the TMR ratio on $\epsilon_{\rm F}$ for $S^z_{\rm tot}=0.0$ and 1.0 with $K_{+-}=0.0$. The solid curve for $S^z_{\rm tot}=0.0$ corresponds to a tunneling through

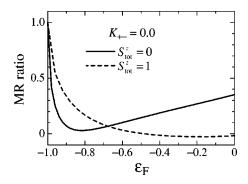


FIG. 7. Calculated results of the TMR ratio as a function of the Fermi energy $\epsilon_{\rm F}$ for $S_{\rm tot}^z = 0$ and 1 with $K_{+-} = 0.0$.

the usual potential barrier because no spin-filtering effect exists in this case, and the result is quite similar to that obtained previously in the free-electron model.²⁷ For $\epsilon_F \approx -0.8$, $\Gamma_{P\uparrow} > \Gamma_{P\downarrow}$ is satisfied. Especially $\epsilon_F \sim -1.0$, $\Gamma_{P\uparrow}$ is much higher than $\Gamma_{P\downarrow}$, resulting in a quite high TMR ratio. As ϵ_F is raised, $\Gamma_{P\downarrow}$ becomes higher and the TMR ratio becomes zero around $\epsilon_F \sim -0.8$. For $\epsilon_F > -0.8$, we get $\Gamma_{P\uparrow} < \Gamma_{P\downarrow}$. When $S_{tot}^z = 1.0$, the \uparrow spin electrons becomes more transparent, and $\Gamma_{P\uparrow}$ therefore increases. Therefore the TMR ratio at $S_{tot}^z = 1$ is large for $\epsilon_F \approx -0.7$, while it is small for $\epsilon_F \approx -0.7$ as compared with the TMR ratio at $S_{tot}^z = 0$.

Figure 8 shows the TMR ratio with and without spin-flip tunneling as a function of h^{ex} (or S_{tot}^z) for $\epsilon_F = -0.99$ and -0.5. The TMR ratio increases monotonically with increasing h^{ex} when $\epsilon_F = -0.99$, while it becomes zero once around $S_{tot}^z = 1$ for $\epsilon_F = -0.5$, as the value $\Gamma_{P\uparrow}$ being smaller than $\Gamma_{P\downarrow}$ is compensated for due to the exchange interaction, as mentioned above. We see that the spin-flip tunneling decreases the ratio by only $\sim 10\%$.

Because the ferromagnetic manganites are nearly half metallic with a spin polarization of ~ 0.7 ,²⁸ the position of the Fermi level should be close to the band bottom. Therefore the high TMR ratio observed by Philipp *et al.*⁷ may be reasonably understood. Furthermore, the approximately linear increase in the TMR ratio with increasing magnetic field is in qualitative agreement with experimental increases.^{5,10} Lee

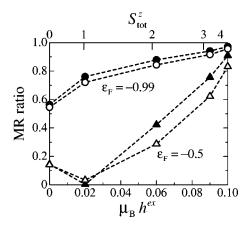


FIG. 8. Calculated results of the TMR ratio as a function of the external field or S_{tot}^z for $\epsilon_{\text{F}} = -0.99$ (circles) and -0.5 (triangles) with $K_{+-} = 0$ (closed symbols) and $K^{+-} = 2.0$ (open symbols).

*et al.*¹⁰ have attributed the dependence of GB-TMR on h^{ex} at high magnetic fields to a second-order process, pointing out that a $\langle \cos^2 \theta \rangle$ term appears in the expression of the tunnel conductance. It is interesting to note that the same factor appears in Eq. (12) obtained in Sec. III. Although the size of the magnetic domains considered by Lee *et al.* is much larger than the system treated in Sec. III, a similar mechanism might be responsible in GB-TMR.

Finally, we would like to remark on the method we have adopted. As the exchange interaction in the Hamiltonian is of a many-body interaction, our method can be applied only for the low-carrier (tunneling electron) density limit for a finite number of localized spins. Because we have dealt with a zero-bias limit, we assumed that the final states are the same with the initial ones in the calculation of the trace in Eq. (6). Despite these limitations, we believe that the present calculation may capture some essential features of GB-TMR. It should also be noted that our method is different from that used for quantum dots showing Kondo resonance because we are dealing with ferromagnetic Kondo coupling in this section. Thus the situation in our case is different from those considered by Appelbaum²⁹ and Anderson³⁰ where the leads are paramagnetic.

V. SUMMARY

We have studied TMR in FTJ's in which the tunneling electrons interact with localized spin in the barrier. In the case where the localized spins are classical at angles in relation to the magnetization axis of the ferromagnetic leads, we have shown, by giving an explicit expression of the TMR ratio, that the enhancement of the TMR ratio in δ -doped FTJ's can be understood in terms of the spin dependence of the energy levels within the barrier region, and that the TMR ratio increases with increasing external magnetic field when the localized spin has a uniaxial anisotropy. We have further demonstrated that the sample-to-sample fluctuations of the tunnel conductance are weaker in the AP alignment of the magnetization of the leads. In the case where the tunneling electron interacts with quantum spins, we have extended the treatment of tunnel conductance to take into account the many-body effects of the exchange interaction. We found that the TMR ratio is decreased by approximately 10% due to the spin-flip process caused by the exchange interaction, and that it increases with an increasing total magnetic moment of GB, in agreement with experimental results. A unified theory to treat the exchange interaction between the tunneling electrons and localized spins in larger regions remains to be studied in future. Our theory has been restricted to zero-bias limit and therefore any relevance to tunnel spectroscopy for spin excitations, etc., may be given in a future work. Quantitative comparison between the theoretical and experimental results may require adequate information on the distribution of the spin-dependent energy levels as well as the angle distribution of localized spins.

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- ¹T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).
- ²J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).
- ³R. Jansen and J. S. Moodera, J. Appl. Phys. 83, 6682 (1998).
- ⁴R. Jansen and J. S. Moodera, Appl. Phys. Lett. **75**, 400 (1999).
- ⁵H. Y. Hwang, S-W. Cheong, N. P. Ong, and B. Batlogg, Phys. Rev. Lett. **77**, 2041 (1996).
- ⁶A. Gupta, G. Q. Gong, G. Xiao, P. R. Duncombe, P. Lecoeur, P. Trouilloud, Y. Y. Wang, V. P. Dravid, and J. Z. Sun, Phys. Rev. B 54, R15 629 (1996).
- ⁷J. B. Philipp, C. Höfener, S. Thienhaus, J. Klein, L. Alff, and R. Gross, Phys. Rev. B **62**, R9248 (2000).
- ⁸M. Ziese, Phys. Rev. B **60**, R738 (1999).
- ⁹C. Höfener, J. B. Philipp, J. Klein, L. Alff, A. Marx, B. Büchner, and R. Gross, Europhys. Lett. **50**, 681 (2000).
- ¹⁰S. Lee, H. Y. Hwang, B. I. Shraiman, W. D. Ratcliff II, and S-W. Cheong, Phys. Rev. Lett. **82**, 4508 (1999).
- ¹¹P.-G. de Gennes, Phys. Rev. **118**, 141 (1960).
- ¹²J. Inoue and S. Maekawa, Phys. Rev. Lett. **74**, 3407 (1995).
- ¹³F. Guinea, Phys. Rev. B 58, 9212 (1998).
- ¹⁴J. Inoue and S. Maekawa, J. Magn. Magn. Mater. **198-199**, 158 (1999).

- ¹⁵P. Lyu, D. Y. Xing, and J. Dong, Phys. Rev. B 58, 54 (1998).
- ¹⁶H. Itoh, T. Ohsawa, and J. Inoue, Phys. Rev. Lett. 84, 2501 (2000).
- ¹⁷R. Jansen and J. S. Moodera, Phys. Rev. B **61**, 9047 (2000).
- ¹⁸A. Vedyayev, D. Bagrets, A. Bagrets, and B. Dieny, Phys. Rev. B 63, 064429 (2001).
- ¹⁹R. Jansen and J. C. Lodder, Phys. Rev. B **61**, 5860 (2000).
- ²⁰A. M. Bratkovsky, Phys. Rev. B 56, 2344 (1997).
- ²¹A. I. Larkin and K. A. Matveev, Zh. Eksp. Teor. Fiz **93**, 1030 (1987) [Sov. Phys. JETP **66**, 580 (1987)].
- ²²C. Caroli, R. Combescot, P. Nozieres, and D. Saint-James, J. Phys. C 4, 916 (1971).
- ²³H. Itoh, A. Shibata, T. Kumazaki, J. Inoue, and S. Maekawa, J. Phys. Soc. Jpn. **69**, 1632 (1999).
- ²⁴M. Julliere, Phys. Lett. **54A**, 225 (1975).
- ²⁵S. Maekawa and U. Gafvert, IEEE Trans. Magn. 18, 707 (1982).
- ²⁶S. Akazawa, J. Inoue, and S. Maekawa, J. Phys. Soc. Jpn. 66, 2758 (1997).
- ²⁷J. C. Slonczewski, Phys. Rev. B **39**, 6995 (1989).
- ²⁸D. C. Worledge and T. H. Geballe, Appl. Phys. Lett. **76**, 900 (2000).
- ²⁹J. Appelbaum, Phys. Rev. Lett. 17, 91 (1966).
- ³⁰ P. W. Anderson, Phys. Rev. Lett. **71**, 95 (1966).