Effect of Coulomb Blockade on Magnetoresistance in Ferromagnetic Tunnel Junctions

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We study spin-dependent electron tunneling in ferromagnetic junctions containing small metallic islands. The tunneling matrix elements depend on the relative direction of magnetization of the island and electrodes. The dependence of the matrix elements amplifies the cotunneling in the Coulomb blockade regime. We show that in single-electron ferromagnetic transistors the magnetoresistance is strongly enhanced by the Coulomb blockade. The results provide a theoretical basis for recent experiments on ferromagnetic single-electron transistors, ferromagnetic double tunnel junctions, and ferromagnetic granular materials. [S0031-9007(97)05222-8]

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Magnetoresistance phenomena in magnetic nanostructures have received considerable attention since the discovery of the giant magnetoresistance (GMR) in magnetic multilayers [1]. In addition, there is a growing interest in the study of magnetoresistive devices based on magnetic nanostructures; this has led to renewed studies of ferromagnetic-metal/insulator/ferromagnetic-metal tunnel junctions [2–5]. In these junctions, the tunnel resistance decreases when the magnetic moments of the electrodes are parallel in an applied magnetic field. The magnetoresistance in the tunnel junctions is called tunnel magnetoresistance (TMR).

Large magnetoresistance is also observed in highly resistive magnetic granular systems such as Co grains embedded in Al_2O_3 [6]. In granular systems, the conductance (G) shows a characteristic temperature (T) dependence as well as a non-Ohmic behavior; this indicates that electron transport between Co grains is through tunneling across the insulating Al_2O_3 barrier. For this transport the charging energy in the grains will be quite important [7,8].

Let an electron jump onto a grain. For a very small grain, the electrostatic energy increases by $e^2/2C$, where e is the electronic charge and C is the capacitance of the grain. Therefore, unless the charging energy is overcome by bias voltage (V) or thermal energy (k_BT), an electron is not able to propagate between the grains. This is called the Coulomb blockade.

Effects of the Coulomb blockade in metallic single-electron transistors have been extensively studied [9–18]. For small islands, where the Coulomb blockade is strong, sequential tunneling in the transistors is superseded by coherent tunneling via a virtual state of the island; this is called cotunneling [12].

In this Letter, we study cotunneling in ferromagnetic double-tunnel junctions. Since cotunneling is a fourth order process in the tunneling matrix elements, it is sensitive to the relative orientation of magnetization between electrodes and island. As a result, we find that TMR is strongly enhanced by the Coulomb blockade.

Single-electron transistors consisting of ferromagnetic metals such as Ni/NiO/Co/NiO/Ni [18] and double ferromagnetic tunnel junctions with small ferromagnetic islands [19] have recently been fabricated. In both systems, the enhanced TMR was observed in the Coulomb blockade regime. A similar effect was also observed in Co/Al_2O_3 granular systems [20]. Our results provide a theoretical basis for these experiments.

Let us consider a single-electron transistor (SET) with two junctions and a capacitively coupled gate as shown in Fig. 1. The right and left electrodes and the central island are ferromagnetic metals such as Fe, Ni, and Co. The tunnel resistance R_T of each junction in Fig. 1 depends on the relative orientation of the magnetization between the electrodes, i.e., parallel or antiparallel. When the electrodes are made of the same ferromagnetic metal, the tunnel resistance in the ferromagnetic alignment is given by $1/R_T^{(F)} \propto (\mathcal{D}_M^2 + \mathcal{D}_m^2)$, while in the antiferromagnetic alignment $1/R_T^{(A)} \propto 2\mathcal{D}_M\mathcal{D}_m$, where $\mathcal{D}_M$ and $\mathcal{D}_m$ are the densities of states for the majority and minority spin bands at the Fermi level. Their resistance ratio is given by $R_T^{(A)}/R_T^{(F)} = (1 + P^2)/(1 - P^2) > 1$, where $P = (\mathcal{D}_M - \mathcal{D}_m)/(\mathcal{D}_M + \mathcal{D}_m)$ is the spin polarization of the electrodes. In the following, we treat the tunnel resistances $R_T^{(A)}$ and $R_T^{(F)}$ as junction resistances that characterize the effect of ferromagnetism on single-electron tunneling.

When the island in a SET (Fig. 1) is small enough and the temperature low enough, the electrostatic energy

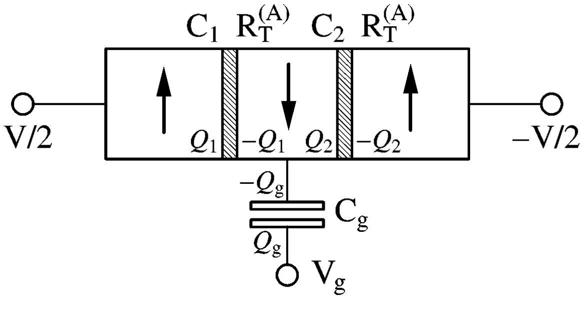

FIG. 1. Schematic of a ferromagnetic single-electron transistor with double tunnel junctions and gate. The arrows indicate the magnetization in island and electrodes.

of excess electrons on the island has significant effect on charge transport in the SET. The increase of the electrostatic energy due to the excess electrons is derived as follows. The average charge on the capacitors in electrostatic equilibrium for given bias and gate voltages, V and V_g, respectively, and given excess island charge, $ne = -Q_1 + Q_2 - Q_g$, is determined by the free energy

$$F(n) = \frac{Q_1^2}{2C_1} + \frac{Q_2^2}{2C_2} + \frac{Q_g^2}{2C_g} - \frac{1}{2} Q_1 V - \frac{1}{2} Q_2 V - Q_g V_g \,, \tag{1}$$

where the C's are the capacitances of junctions 1 and 2 and the gate; we subsequently set $C_1 = C_2 = C$. The variation of $F(n)$ with respect to Q_1 and Q_2 yields Kirchhoff's law. The energy change associated with forward tunneling of an electron through the first junction $(n \rightarrow n + 1)$ is given by $E_1^+(n) = F(n + 1) - F(n) - eV/2 = (1 + 2n)E_c + (C_g/C_\Sigma)eV_g - \frac{1}{2} eV$, and that through the second junction $(n \rightarrow n - 1)$ is given by $E_2^-(n) = F(n - 1) - F(n) - eV/2 = (1 - 2n)E_c - (C_g/C_\Sigma)eV_g - \frac{1}{2} eV$, where $E_c = e^2/2C_\Sigma$ and $C_\Sigma = 2C + C_g$. The energy change for the backward tunneling, $E_1^-(n)$ and $E_2^+(n)$, is given by $E_1^-(n) = E_2^-(n) + eV$ and $E_2^+(n) = E_1^+(n) + eV$, respectively.

Let us first discuss tunneling in the SET based on the sequential tunneling model, in which tunneling events in junctions 1 and 2 occur independently. For small bias and gate voltages, all $E_j^\pm$ $(j = 1, 2)$ are positive, so that tunneling in either of the two junctions increases the electrostatic energy. Therefore, sequential tunneling of electrons through the junctions is suppressed for $T \ll E_c$ (Coulomb blockade).

It has been pointed out that, in the Coulomb blockade regime, there is a higher order process of tunneling through both junctions via a virtual intermediate state with increased electrostatic energy. This *cotunneling* process is energetically favorable since the electrostatic energy of the final state is negative, i.e., $E_1^+(n) + E_2^-(n + 1) = -eV < 0$. Following Averin and Nazarov [12], we calculate the current through the double junctions

$$I_\sigma = e \sum_{n=-\infty}^{\infty} p_n[\overrightarrow{\Gamma}(n) - \overleftarrow{\Gamma}(n)] \,, \tag{2}$$

where $\overrightarrow{\Gamma}(n)$ and $\overleftarrow{\Gamma}(n)$, respectively, are the forward and backward tunneling rates

$$\overleftrightarrow{\Gamma}(n) = \frac{\hbar}{8\pi e^4} \left[\frac{1}{R_T^{(\sigma)}} \right]^2 \int_{-\infty}^{\infty} d\epsilon \, \frac{\epsilon(\epsilon \mp eV) \exp[\pm eV/2T]}{\sinh[\epsilon/2T] \sinh[(\epsilon \mp eV)/2T]} \times \left| \frac{1}{\epsilon + E_1^\pm(n) + i\gamma_\sigma^\pm} + \frac{1}{-\epsilon + E_2^\mp(n) \pm eV + i\gamma_\sigma^\mp} \right|^2 . \tag{3}$$

Here σ is either F (ferromagnetic) or A (antiferromagnetic) alignment of the moments, and $\gamma_\sigma^\pm$ is given by

$$\gamma_\sigma^\pm = (g_\sigma/2) \sum_{j=1,2} E_j^\pm(n) \coth[E_j^\pm(n)/2T] \,, \tag{4}$$

which represents the decay rate of the charge state $n \pm 1$ [21]; here $g_\sigma = (\hbar/e^2)/R_T^{(\sigma)}$. In Eq. (2), p_n is the probability of charge state n and is obtained from the condition for detailed balancing: $p_n[\Gamma_1^+(n) + \Gamma_2^+(n)] = p_{n+1}[\Gamma_1^-(n + 1) + \Gamma_2^-(n + 1)]$, where $\Gamma_j^\pm(n)$ are the tunneling rates of $n \rightarrow n \pm 1$ in the jth junction $\Gamma_j^\pm(n) = -(1/e^2 R_T^{(\sigma)}) E_j^\pm(n)/(1 - \exp[E_j^\pm(n)/T])$. From Eq. (2) we can calculate the current I_σ.

To extract an analytical behavior, we calculate the resistance $R_\sigma = dV/dI_\sigma$ at zero applied bias and gate voltages ($V = V_g = 0$). The result is

$$R_\sigma^{-1} = \frac{1}{2\pi R_T^{(\sigma)}} \int_{-\infty}^{\infty} d\epsilon \left[\frac{\epsilon/T}{\sinh(\epsilon/T)} \right] \times \frac{\gamma_\sigma(\epsilon)}{(\epsilon - E_c)^2 + \gamma_\sigma^2(E_c)} \,, \tag{5}$$

with $\gamma_\sigma(\epsilon) = g_\sigma \epsilon \coth(\epsilon/2T)$. At high temperatures ($T \gg E_c$), Eq. (5) is replaced with the result of the thermally assisted sequential tunneling,

$$R_\sigma \approx 2R_T^{(\sigma)}(1 + E_c/3T) \,, \tag{6}$$

whereas at low temperatures in the Coulomb blockade regime ($T \ll E_c$), we obtain

$$R_\sigma \approx \frac{3e^2}{4\pi\hbar} [R_T^{(\sigma)}]^2 (E_c/T)^2 \,, \tag{7}$$

which expresses the inelastic cotunneling. We emphasize that the resistance R_σ for sequential tunneling is proportional to the *sum* of the resistances $2R_T^{(\sigma)}$ of the two junctions, while R_σ in the cotunneling case is proportional to the *product* of the resitances $[R_T^{(\sigma)}]^2$ of the two junctions. The resistance ratio R_A/R_F is given by

$$R_A/R_F = R_T^{(A)}/R_T^{(F)}, \qquad (T \gg E_c) \tag{8}$$

in the sequential-tunneling regime, and

$$R_A/R_F = [R_T^{(A)}/R_T^{(F)}]^2, \qquad (T \ll E_c) \tag{9}$$

in the cotunneling regime. Therefore, the TMR for $T \ll E_c$ is enhanced by $[R_T^{(A)}/R_T^{(F)}]^2$ compared with $R_T^{(A)}/R_T^{(F)}$ in the absence of the Coulomb blockade. For example, if we use Fe for the electrodes and $P = 0.4$ for the spin polarization, we expect an enhancement of $(R_A - R_F)/R_F$ from 38% to 91% by the Coulomb blockade.

Figure 2(a) shows the temperature dependence of the resistance R_σ at $V = V_g = 0$ for three values of tunnel resistance $R_T^{(\sigma)}/R_Q$ with $R_Q = \pi\hbar/e^2$. The curves clearly show the crossover near $T/E_c = 0.1$, below which the cotunneling gives the dominant contribution since the sequential tunneling is exponentially suppressed and the resistance shows the T^{-2} dependence, and above which sequential tunneling is recovered. Note that, in the limit of $R_T^{(\sigma)}/R_Q \to \infty$, cotunneling disappears and R_σ follows the classical expression $2R_T^{(\sigma)} \sinh(E_c/T)/(E_c/T)$ in the whole temperature range below $\sim 0.4E_c$ [cf. Eq. (6) is valid for $T/E_c \gtrsim 1$]. The thin dashed line indicates the resistance at the gate voltage of $eV_g/E_c = C_\Sigma/C_g$, where the charging energy is canceled out by this gate voltage and the Coulomb blockade is neutralized. Figure 2(b) shows the ratio R_A/R_F as a function of temperature for different values of $R_T^{(A)}$ and $R_T^{(F)}$, keeping their ratio $R_T^{(A)}/R_T^{(F)} = 2$. As the temperature is lowered, the MR is enhanced from $(R_A/R_F) = 2$, where the sequential tunneling is dominant, to $(R_A/R_F) = 4$, where the cotunneling is dominant. In the crossover region both tunneling processes contribute to the tunnel current.

Figure 3(a) shows the resistance R_σ as a function of gate voltage at $V = 0$. At about $eV_g/E_c = 0$ and ± 10, the resistance R_σ increases rapidly with decreasing temperature. On the other hand, the values of the minima in R_σ at $eV_g = (C_\Sigma/C_g)E_c = \pm 5E_c$ (mode $10E_c$) are almost unchanged. The minima in the resistance correspond to well-known conductance resonance peaks, where the neighboring charge states, $n = 0$ and $n \pm 1$, have the same electrostatic energy. Figure 3(b) shows R_A/R_F as a function of gate voltage. We see that the behavior of the MR curves is well correlated with that of $\log(R_\sigma)$. In the Coulomb blockade regime ($T \lesssim 0.2E_c$) the MR near $V_g = 0$ progressively increases from $(R_A/R_F) = 2$ to $(R_A/R_F) = 4$ as temperature is lowered, while the MR at about $eV_g/E_c = \pm 5$ is unchanged.

Figure 4(a) shows resistance R_σ normalized to the zero bias resistance $R_\sigma(0)$ as a function of bias voltage at $V_g = 0$. The values of $R_\sigma(0)$ for $T/E_c = 0.04$, 0.08, and 0.16 are $4.5 \times 10^4 R_Q$, $9.7 \times 10^3 R_Q$, and $6 \times 10^2 R_Q$, respectively. The resistance steeply decreases, as the bias voltage increases. R_σ at low temperatures shows a power law dependence, $R_\sigma \propto 1/V^2$, as expected for a cotunneling process. Figure 4(b) shows the MR (R_A/R_F) as a function of bias voltage. As temperature is decreased, the MR is enhanced from $(R_A/R_F) = 2$ to $(R_A/R_F) = 4$. At $T/E_c = 0.04$, the enhanced MR is constant up to $eV/E_c \sim 1.2$, whereas the corresponding resistance is reduced by several orders of magnitude.

Ono *et al.* [18] measured the MR in the Ni/NiO/Co double junctions and observed Coulomb oscillations in the R_σ vs V_g curves. Off resonance (i.e., at the peaks of R_σ), they found that the MR ratio, $(R_A - R_F)/R_F$, is enhanced to 40%, which is larger than the value of 17.5% expected from $P_{\mathrm{Co}} = 0.35$ and $P_{\mathrm{Ni}} = 0.23$ in the absence of the Coulomb blockade effect. The present theory explains this enhancement since $(R_A - R_F)/R_F = [R_T^{(A)}/R_T^{(F)}]^2 - 1 = 0.38$, i.e., the MR is 38%. However, they found an MR of $\sim 4\%$ at resonance, which is considerably smaller than the expected MR of 17.5%. A reduction in MR at resonance may occur in case of strong tunneling $R_T^{(\sigma)} \lesssim R_Q$. Recent experiments and theories for the SET have

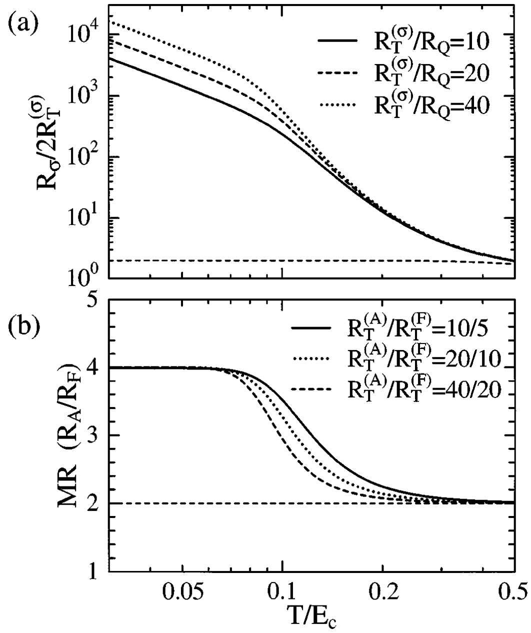

FIG. 2. (a) Resistance R_σ and (b) magnetoresistance ratio R_A/R_F as functions of temperature at $V = 0$ and $V_g = 0$, where σ is F (ferromagnetic) or A (antiferromagnetic), depending on whether the magnetizations in the electrodes are parallel or antiparallel.

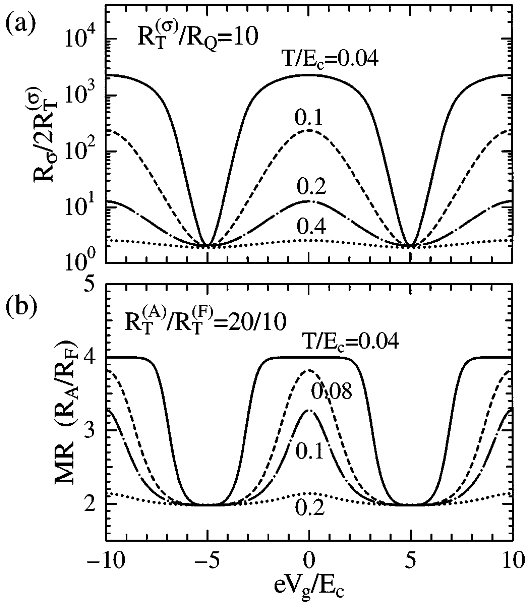

FIG. 3. (a) Resistance R_σ and (b) magnetoresistance ratio R_A/R_F as functions of gate voltage V_g at zero bias voltage $V = 0$, where σ is F (ferromagnetic) or A (antiferromagnetic), depending on whether the magnetizations in the electrodes are parallel or antiparallel.

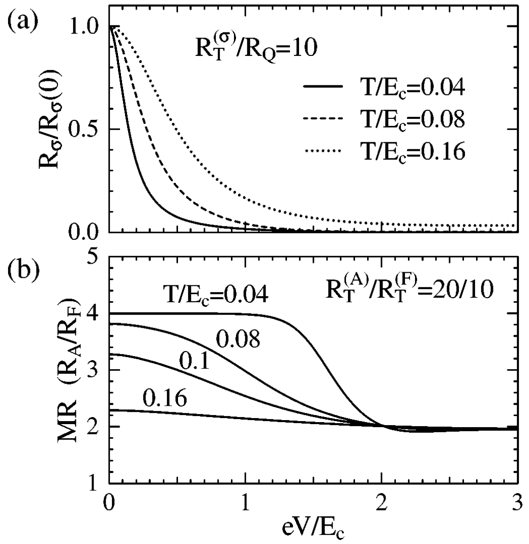

FIG. 4. (a) Resistance R_σ and (b) magnetoresistance ratio R_A/R_F as functions of bias voltage V at zero gate voltage $V_g = 0$, where σ is F (ferromagnetic) or A (antiferromagnetic), depending on whether the magnetizations in the electrodes are parallel or antiparallel.

shown that the conductance in the strong tunneling case significantly deviates from that in the weak tunneling case $R_T^{(\sigma)} \gg R_Q$. Using the theory of König *et al.* [16], we obtain the MR at the resonance

$$\frac{R_A}{R_F} - 1 = \frac{(R_T^{(A)}/R_T^{(F)}) - 1}{1 + (2/\pi^2)(R_Q/R_T^{(F)})[\gamma_E + \ln(E_c/\pi T)]}, \tag{10}$$

where γ_E is Euler's constant. For $R_T^{(F)}/R_Q \lesssim (2/\pi^2)[\gamma_E + \ln(E_c/\pi T)]$, the MR in Eq. (10) is considerably reduced from $(R_T^{(A)}/R_T^{(F)}) - 1$. If this is the case for the Ni/NiO/Co double junctions, we can explain the small value of MR mentioned above. In contrast, the TMR off resonance remains unaltered [22].

Spin-dependent tunneling with the Coulomb blockade has also been found in sputtered Co/Al$_2$O$_3$ granular thin films by Fujimori *et al.* [6] and Mitani *et al.* [20], and in fabricated Co/Al$_2$O$_3$/Co tunnel junctions with small Co clusters in the Al$_2$O$_3$ barrier by Schelp *et al.* [19]. In granular systems, the MR is enhanced by the Coulomb blockade effect at low temperatures; this is consistent with the present theory. Moreover, the bias voltage dependence of the resistance as well as the MR is quite similar to those at $T/E_c = 0.04$ in Fig. 4. Recently, they have observed a sudden decrease in MR above a certain applied voltage [20], in good accord with the curve of $T/E_c = 0.04$ in Fig. 4(b). The similarities between our results obtained for the SET and those in the granular system strongly suggest that the mechanism we presented here may work in the granular system, even though the two systems are apparently different. On the other hand, the monotonic decrease in the MR vs V curve by Schelp *et al.* suggests that their junctions lie in the crossover regime between sequential tunneling and cotunneling. If different kinds of ferromagnetic metals are used between clusters and electrodes in the junctions, the oscillation of the MR, as well as the resistance, may be observed in the magnetic field. This is because the shift of the chemical potentials is different between them in the field, and thus the field plays a role similar to the gate voltage [18].

In conclusion, we have studied spin-dependent electron tunneling in ferromagnetic single-electron transistors, and have shown that the dependence of the tunneling matrix elements on the relative orientation of the electrode magnetizations amplifies cotunneling in the Coulomb blockade regime; this results in an enhanced magnetoresistance. The spin-dependent tunneling together with the Coulomb blockade provides the basis for understanding magnetic transport in magnetic nanostructures.

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