Spin-dependent Coulomb blockade in ferromagnet/normal-metal/ferromagnet double tunnel junctions

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We study theoretically the spin-dependent transport in ferromagnet/normal-metal/ferromagnet double tunnel junctions by special attention to cotunneling in the Coulomb blockade region. The spin accumulation caused by cotunneling squeezes the Coulomb blockade region when the magnetizations in the ferromagnetic electrodes are antiparallel. Outside the squeezed Coulomb blockade region, we propose an anomalous region where the sequential tunneling in one of the spin bands is suppressed by the Coulomb blockade and that in the other is not. In this region, the tunnel magnetoresistance oscillates as a function of bias voltage. The temperature dependences of the tunnel magnetoresistance and the magnitude of the spin accumulation are calculated. [S0163-1829(99)08209-0]

The spin-dependent transport in magnetic materials, in particular, the tunnel magnetoresistance (TMR) in ferromagnetic junctions, has attracted much interest.[1–3] Recent advances in nanolithography and thin-film processing make it possible to fabricate very small double tunnel junctions called single electron transistor (SET), where the electrostatic energy of excess electrons in the island has a significant effect on charge transport, i.e., the Coulomb blockade (CB).[4] In the CB region, sequential tunneling, where tunneling events in each junction occur independently, is blocked at $T=0$ by the electrostatic energy, and superceded by a more complex tunneling process called ''cotunneling.''[5,6] In ferromagnetic SET's,[7] it was recently pointed out that TMR oscillates in sequential tunneling regime[8] and TMR is enhanced in the CB region by cotunneling.[9]

In this paper, we study theoretically the spin-dependent single electron tunneling in ferromagnet/normal-metal/ferromagnet (FNF) double tunnel junctions shown in Figs. 1(a) and 1(c), and find an anomalous region intrinsic to the spin accumulated system. For the antiferromagnetic (A) alignment, where the magnetizations of the right and left electrodes are antiparallel as shown in Fig. 1(b), electrons with up (down) spin are easy (difficult) to tunnel into the normal metal, and difficult (easy) to tunnel out of it, because the densities of states of the left and right electrodes are different between up and down spin bands. This imbalance among the tunneling currents causes the spin accumulation,[10–12] when the spin-relaxation time is sufficiently long in the normal metal. In the CB region, where sequential tunneling is blocked at $T=0$, cotunneling causes the spin accumulation.

As shown in Fig. 1(b), the spin accumulation increases (decreases) the chemical potential for electrons with up (down) spin. This shift of the chemical potential decreases the energy for adding (extracting) an electron with down (up) spin to (from) the normal-metal island. Therefore the critical voltage, above which electrons with down (up) spin can tunnel into (out of) the normal-metal island, is lowered and the CB region is squeezed as shown in Fig. 2. On the other hand, the critical voltage above which electrons with up (down) spin can tunnel into (out of) the normal-metal island is raised by spin accumulation. Consequently, an anomalous region appears between the lowered and raised critical voltages. We call this region the half CB region, since the sequential tunneling in one of the spin bands is suppressed and that in the other is not. Cotunneling is essential for appearance of the half CB region. If cotunneling is not taken into account, the spin accumulation does not occur in the CB region and the boundary of the CB region does not change, since there is no tunneling current. In the half CB region, we find an anomalous oscillation in TMR as a function of bias voltage. The crossover between cotunneling and sequential tunneling[9] is also found in the temperature dependence of the TMR and the spin accumulation.

The TMR in the ferromagnet/ferromagnet (FF) single junctions has been extensively studied,[1–3,13,14] where the tun-

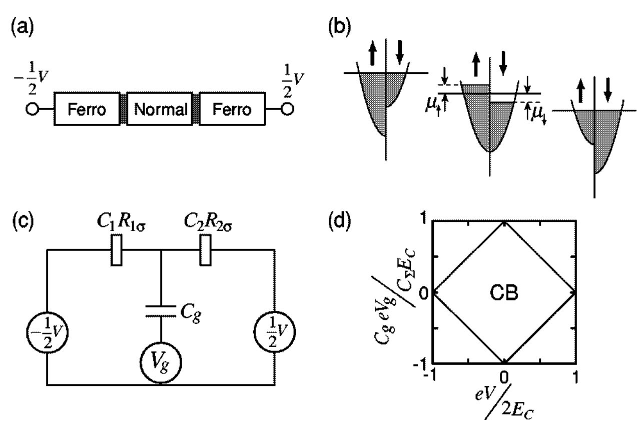

FIG. 1. (a) FNF double junction. Insulating barriers are shown by the shaded rectangles. (b) The densities of states for the antiferromagnetic alignment are schematically shown. $\mu_\uparrow$ and $\mu_\downarrow$ denote the shifts of chemical potential for up and down spins, respectively. (c) FNF single electron transistor with a gate. (d) The Coulomb blockade region for the ferromagnetic alignment with $n=0$.

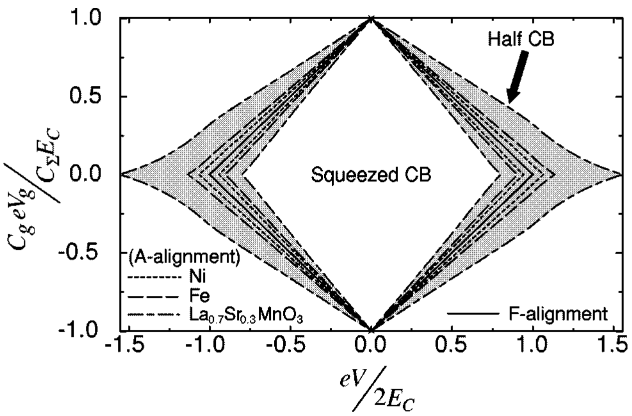

FIG. 2. The boundary of the CB region for the ferromagnetic alignment is indicated by the solid line. Boundaries of the half CB region for the antiferromagnetic alignment with Ni, Fe, and $La_{0.7}Sr_{0.3}MnO_3$ electrodes are plotted by dotted, dashed, and dot-dashed lines, respectively. The half CB region for $La_{0.7}Sr_{0.3}MnO_3$ electrodes is indicated by the shade. The tunnel resistance for the majority spin band is taken to be $R_M=2R_K$ for all systems.

nel resistance depends on the relative orientation of the magnetizations between the electrodes, i.e., parallel or antiparallel. When the electrodes are made of the same ferromagnetic metal, the tunnel resistance for the ferromagnetic (F) alignment, where the magnetizations of the left and right electrodes are parallel, is given by $1/R_F\propto(\mathcal{D}_M^2+\mathcal{D}_m^2)$, while for the A alignment $1/R_A\propto 2\mathcal{D}_M\mathcal{D}_m$. Here $\mathcal{D}_M$ and $\mathcal{D}_m$ are the densities of states for the majority and minority spin bands at the Fermi level, respectively. The tunnel magnetoresistance is written as

$$\frac{R_A-R_F}{R_F}=\frac{2P^2}{1-P^2}, \tag{1}$$

where $P=(\mathcal{D}_M-\mathcal{D}_m)/(\mathcal{D}_M+\mathcal{D}_m)$ is the spin polarization of the electrodes. The key point is that the difference between the products of the densities of states for each spin band causes the TMR in FF junctions.

We now turn to FNF double tunnel junctions shown in Fig. 1(a). It should be noted that the TMR does not exist in FN single junctions, because there is no difference between the densities of states for up and down spin bands in the normal metal. For FNF junctions, however, the TMR exists, if the spin flip process in the normal-metal island can be neglected. The origin of the TMR in FNF junctions is quite different from that for FF junctions. We assume that the right and left electrodes are made of the same ferromagnetic metal such as Ni, Fe, and mixed-valent manganites like $La_{0.7}Sr_{0.3}MnO_3$ and the central electrode is made of a normal metal such as Al with sufficiently long spin relaxation time.

We first consider what happens if we neglect the Coulomb charging energy. For the A alignment, introducing the chemical potential shift for up (down) spin electrons, $\mu_\uparrow$ ($\mu_\downarrow$), the spin-resolved tunneling currents through each junction at $T=0$ are given by $I_{1\uparrow}\propto\mathcal{D}_N\mathcal{D}_M(\frac{1}{2}\ eV-\mu_\uparrow)$, $I_{1\downarrow}\propto\mathcal{D}_N\mathcal{D}_m(\frac{1}{2}\ eV-\mu_\downarrow)$, $I_{2\uparrow}\propto\mathcal{D}_N\mathcal{D}_m(\frac{1}{2}\ eV+\mu_\uparrow)$, and $I_{2\downarrow}\propto\mathcal{D}_N\mathcal{D}_M(\frac{1}{2}\ eV+\mu_\downarrow)$. Here, $\mathcal{D}_N$ is the density of states in the normal metal for each spin band, and subscripts 1 and 2 represent the left and right junctions, respectively.

The shifts $\mu_\uparrow$ and $\mu_\downarrow$ are determined by the stability condition, i.e., $I_{1\uparrow}=I_{2\uparrow}$ and $I_{1\downarrow}=I_{2\downarrow}$. Then, we have $\mu_\uparrow=-\mu_\downarrow=\frac{1}{2}P\ eV$. The tunnel resistance for the A alignment is written as $R_A\propto(1/\mathcal{D}_M+1/\mathcal{D}_m)/4\mathcal{D}_N$. For the F alignment, the spin-resolved currents are balanced without spin accumulation. Therefore, the chemical potential for each spin electron does not shift and the tunnel resistance is given by the usual manner as $R_F\propto 1/\mathcal{D}_N(\mathcal{D}_M+\mathcal{D}_m)$. The TMR for FNF junctions is obtained as

$$\frac{R_A-R_F}{R_F}=\frac{(\mathcal{D}_M-\mathcal{D}_m)^2}{4\mathcal{D}_M\mathcal{D}_m}=\frac{P^2}{1-P^2}, \tag{2}$$

which is half of that for FF junctions in Eq. (1). Equation (2) has also been obtained by Brataas *et al.*[15] The polarizability P for Ni, Fe, and $La_{0.7}Sr_{0.3}MnO_3$ are, respectively, 0.23, 0.40,[16] and 0.83.[17,18] Introducing these P values into Eq. (2), we estimate the TMR for FNF double junctions made of Ni, Fe, and $La_{0.7}Sr_{0.3}MnO_3$ electrodes to be 5.6, 19, and 220 %, respectively.

Let us next examine the single electron transistor with a capacitively coupled gate as shown in Fig. 1(c). The transistor is made of the same materials as the FNF double junctions discussed above, and its size is small enough to observe the CB. For simplicity, we assume that the insulating barriers for junctions 1 (left) and 2 (right) are the same; we subsequently set $C_1=C_2\equiv C$.

The energy changes due to the forward tunneling of an electron with spin σ through the junction 1 ($n\to n+1$) and the junction 2 ($n\to n-1$) are given by $E_{1\sigma}^{(+)}(n)=E_C(1+2n)-(C_g/C_\Sigma)\ eV_g-\frac{1}{2}\ eV+\mu_\sigma$ and $E_{2\sigma}^{(-)}(n)=E_C(1-2n)+(C_g/C_\Sigma)\ eV_g-\frac{1}{2}\ eV-\mu_\sigma$, respectively, where n is the number of excess electrons in the normal-metal island, V_g the gate voltage, $E_C=e^2/2C_\Sigma$, and $C_\Sigma=2C+C_g$. The energy changes due to the backward tunneling, $E_{1\sigma}^{(-)}(n)$ and $E_{2\sigma}^{(+)}(n)$, are given by $E_{1\sigma}^{(-)}(n)=E_{2\sigma}^{(-)}(n)+eV$ and $E_{2\sigma}^{(+)}(n)=E_{1\sigma}^{(+)}(n)+eV$, respectively.

The total current through the double junctions[9] is expressed as

$$I=\sum_{n=-\infty}^{\infty}\sum_{\sigma,\sigma'=\uparrow\downarrow}p_n[I_{1\sigma 2\sigma'}(n)-I_{2\sigma 1\sigma'}(n)], \tag{3}$$

where p_n is the probability of charge state n and $I_{j\sigma k\sigma'}(n)$ represents the current, where electrons with spin σ tunnel into the central island through the jth junction and electrons with spin σ' tunnel out of it through the kth junction. The probability p_n is determined by 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 $\Gamma_j^{(\pm)}(n)=\Sigma_\sigma(1/e^2R_{j\sigma})E_{j\sigma}^{(\pm)}(n)/\{\exp[E_{j\sigma}^{(\pm)}(n)/T]-1\}$ are the tunneling rates of $n\to n\pm 1$ in the jth junction. $I_{j\sigma k\sigma'}(n)$ is obtained by the ''golden rule,''[5] as

$$I_{j\sigma k\sigma'}(n)=I_0\frac{1}{R_{j\sigma}R_{k\sigma'}}\int d\epsilon_1 d\epsilon_2 d\epsilon_3 d\epsilon_4 f(\epsilon_1)[1-f(\epsilon_2)]$$
$$\times f(\epsilon_3)[1-f(\epsilon_4)]\left|\frac{1}{\epsilon_2-\epsilon_1+E_{j\sigma}^{(+)}(n)+i\gamma^{(+)}}\right.$$
$$\left.+\frac{1}{\epsilon_3-\epsilon_4+E_{k\sigma'}^{(-)}(n)+i\gamma^{(-)}}\right|^2$$
$$\times\delta(\epsilon_1-\epsilon_2+\epsilon_3-\epsilon_4+\Delta E), \tag{4}$$

where $I_0\equiv(E_C/eR_K)$, $R_K=h/e^2$ is the resistance quantum, $R_{j\sigma}$ is the tunnel resistance of the jth junction for electrons with spin σ, ΔE is the energy difference between initial and final states, and $\gamma^{(\pm)}=\frac{1}{2}\Sigma_\sigma\Sigma_j g_{j\sigma}E_{j\sigma}^{(\pm)}(n)\coth[E_{j\sigma}^{(\pm)}(n)/2T]$ represents the decay rate of the charge state $n\pm1$ with $g_{j\sigma}=(R_K/2\pi R_{j\sigma})$.[19,20] Note that in the limit of large tunnel resistance ($g_{j\sigma}\rightarrow 0$) where the cotunneling current is negligible, Eq. (3) reduces to the sequential tunneling current in the orthodox theory.[4]

For the F alignment, the tunnel resistances $R_{j\sigma}$ are given by $R_{1\uparrow}=R_{2\uparrow}=R_M$ and $R_{1\downarrow}=R_{2\downarrow}=R_m$, where R_M ($\propto 1/\mathcal{D}_M\mathcal{D}_N$) and R_m ($\propto 1/\mathcal{D}_m\mathcal{D}_N$) are the tunnel resistances for electrons in the majority and minority spin bands of the ferromagnetic electrodes, respectively. The spin does not accumulate, since $I_{i\uparrow j\downarrow}(n)=I_{i\downarrow j\uparrow}(n)$ for $\mu_\uparrow=\mu_\downarrow=0$. Therefore the CB region is the same as that for the usual metallic single electron transistors as shown in Fig. 1(d).

For the A alignment, the tunnel resistances are given by $R_{1\uparrow}=R_{2\downarrow}=R_M$ and $R_{1\downarrow}=R_{2\uparrow}=R_m$. If $\mu_\uparrow=\mu_\downarrow=0$, then $I_{i\uparrow j\downarrow}(n)\neq I_{i\downarrow j\uparrow}(n)$, which gives rise to the spin accumulation and nonzero shifts μ_σ. The shifts of the chemical potential satisfy the condition that $\mu_\uparrow=-\mu_\downarrow$, because we assumed that the left and right ferromagnetic electrodes are the same and the density of states in the normal metal is constant. We introduce the symbol $\delta\equiv\mu_\uparrow=-\mu_\downarrow$, which is determined by the stability condition $\Sigma_n\Sigma_{i,j}p_n[I_{i\uparrow j\downarrow}(n)-I_{i\downarrow j\uparrow}(n)]=0$. We carry out the numerical integration of Eq. (4) and obtain δ. Here we take $R_M=2R_K$ $[R_m=\{(1-P)/(1+P)\}R_M]$.

At $T=0$ and for low V where sequential tunneling is blocked by the CB, cotunneling causes the spin accumulation in the normal-metal island, leading to the increase (decrease) of the chemical potential of up-spin (down-spin) electrons by δ as shown in Fig. 1(b). Therefore the energy changes due to the single electron tunneling are given by $E_{i\uparrow}^{(\pm)}(n)=E_i^{(\pm)}(n)\pm\delta$ and $E_{i\downarrow}^{(\pm)}(n)=E_i^{(\pm)}(n)\mp\delta$, where $E_i^{(\pm)}(n)$ are the energy changes for $\delta=0$. The CB region for the A alignment is now determined by $E_1^{(\pm)}(n),E_2^{(\pm)}(n)>\delta$ and squeezed as shown in Fig. 2. Note that for the F alignment, the CB region is given by $E_1^{(\pm)}(n)$, $E_2^{(\pm)}(n)$ >0.

Outside the squeezed CB region, electrons with down (up) spin can tunnel into (out of) the normal-metal island. However, those with up (down) spin cannot as far as $E_1^{(\pm)}(n)$, $E_2^{(\pm)}(n)$ $>-\delta$. As a consequence, we have a new spin-dependent CB region, where the sequential tunneling in one spin bands is suppressed and the other is not. This "half CB" region is given by the condition that $-\delta<$ $E_1^{(\pm)}(n)$, $E_2^{(\pm)}(n)$ $<\delta$. In Fig. 2, the boundaries of this "half CB" region for the A alignment with Ni, Fe, and

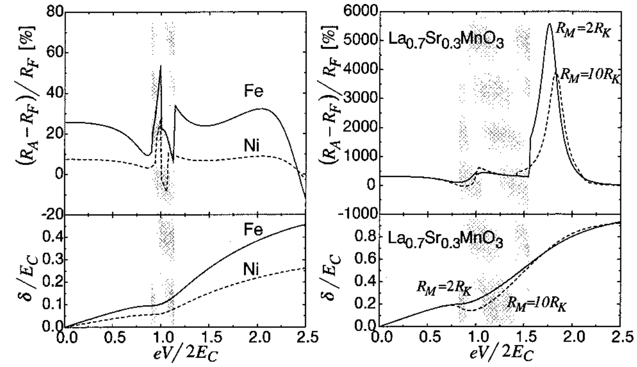

FIG. 3. Left: The tunnel magnetoresistance for Ni and Fe electrodes are plotted against the bias voltage V in the upper panel. The chemical potential shift δ is plotted in the lower panel. The shaded area represents the half CB region for Fe electrodes. The tunnel resistance for the majority spin electrons is $R_M=2R_K$, the gate voltage is $V_g=0$ and $T=0$. Right: The same plot for $La_{0.7}Sr_{0.3}MnO_3$ electrodes. The solid (dashed) lines indicate the results for $R_M=2R_K$ ($10R_K$). The half Coulomb blockade region for $R_M=2R_K$ is indicated by the shade. The gate voltage is $V_g=0$ and $T=0$.

$La_{0.7}Sr_{0.3}MnO_3$ electrodes are plotted for $n=0$. The solid line indicates the CB boundary for the F alignment and the shade represents the half CB region for the A alignment with $La_{0.7}Sr_{0.3}MnO_3$ electrodes in Fig. 2. One can see that the half CB region increases and the squeezed CB region, which is surrounded by the half CB region, decreases as P increases, because δ increases with P.

The tunnel magnetoresistance, $(R_A-R_F)/R_F$, and the shift of the chemical potential δ at $T=0$ and $V_g=0$ are plotted in Fig. 3. At $V\simeq 0$, the TMR for Ni, Fe, and $La_{0.7}Sr_{0.3}MnO_3$ electrodes are about 7.5, 26, and 310 %, respectively. The values are 35–40% larger than those in Eq. (2).

As the bias voltage V increases from 0 toward the boundary of the squeezed CB region, the tunneling current for the A alignment increases more rapidly than that for the F alignment and the TMR decreases. At the boundaries of the half

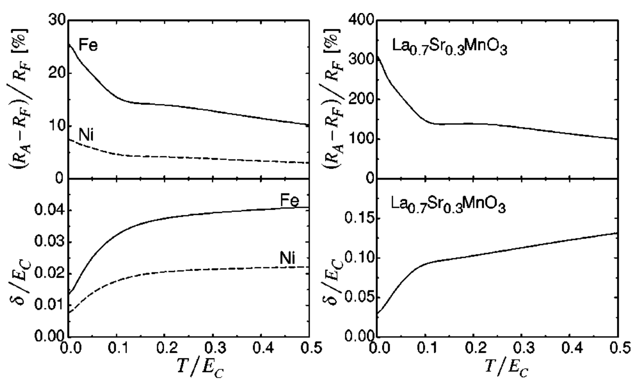

FIG. 4. Left: The temperature dependence of the tunnel magnetoresistance at $V_g=0$ and $eV/2E_c=0.1$ for Ni and Fe electrodes is shown in the upper panel. The temperature dependence of the chemical potential shift δ at $V_g=0$ and $eV/2E_c=0.1$ for Ni and Fe electrodes is plotted in the lower panel. Right: The same plot for $La_{0.7}Sr_{0.3}MnO_3$ electrodes. In both figures the tunnel resistance for the majority spin electrons is $R_M=2R_K$.

CB region, R_A jumps because the current due to cotunneling decreases rapidly and that due to sequential tunneling starts to flow. The same jump in R_F appears at $eV/2E_c=1$, which is the boundary of the CB region for the F alignment. Therefore the TMR oscillates in the half CB region as shown in Fig. 3. We also find that cotunneling enhances the TMR around $eV/2E_c \simeq 2.0$, because it suppresses the V dependence of the total current for the A alignment.

We have also studied the system with large tunnel resistance, $R_M=10R_K$, in order to see what happens when cotunneling is suppressed. In Fig. 3, TMR and δ for the electrodes of $La_{0.7}Sr_{0.3}MnO_3$ and junctions with $R_M=10R_K$ are plotted by the dashed lines. The size of the half CB region is 79% of that for $R_M=2R_K$.

The temperature dependences of TMR and δ have been calculated at $V_g=0$ and $eV/2E_c=0.1$ in the squeezed CB region. As temperature T increases, the sequential tunneling, which is exponentially suppressed at low T, is recovered. The results are shown in Fig. 4. One can see clearly the crossover between cotunneling and sequential tunneling near $T/E_c=0.1$.[9]

In conclusion, we have studied the spin-dependent transport in FNF double junctions. In the antiferromagnetically aligned SET, cotunneling brings about the spin accumulation at low V, where sequential tunneling is blocked at $T=0$. The spin accumulation causes the squeezing of the CB region. Outside the squeezed CB region, we have found a new anomalous region, where the sequential tunneling in one of the spin bands is suppressed and that in the other is not. In this ‘‘half CB’’ region, the TMR oscillates as a function of V. The crossover between cotunneling and sequential tunneling is found in the temperature dependences of TMR and δ.

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