Spin-Imbalance and Magnetoresistance in Ferromagnet/Superconductor/Ferromagnet Double Tunnel Junctions

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We theoretically study the spin-dependent transport in a ferromagnet/superconductor/ferromagnet double tunnel junction. The tunneling current in the antiferromagnetic alignment of the magnetizations gives rise to a spin imbalance in the superconductor. The resulting nonequilibrium spin density strongly suppresses the superconductivity with increase of bias voltage and destroys it at a critical voltage $V_c$. The results provide a new method not only for measuring the spin polarization of ferromagnets but also for controlling superconductivity and tunnel magnetoresistance (TMR) by applying the bias voltage.

Since the early experiments demonstrated the spin-polarized tunneling of electrons from ferromagnetic metals (FM) into superconductors (SC) in FM/SC junctions\textsuperscript{1}, the concept of the spin-polarized transport has been of vital importance in magnetic junctions and multilayers. Firstly the tunneling currents strongly depends on the relative orientation of magnetizations in FM/FM tunnel junctions; the tunnel resistance decreases when the magnetizations are aligned in a magnetic field, causing tunnel magnetoresistance (TMR)\textsuperscript{2,3}. Secondly the spin-polarized current driven from a FM into a normal metal (N) or superconductor (SC) gives rise to a nonequilibrium spin density in N or SC\textsuperscript{4,5}. In a FM/N/FM double junction the TMR effect is brought about by accumulation of spin-polarized electrons in N\textsuperscript{6,7}. In a FM/SC/FM double junction\textsuperscript{8}, on the other hand, we expect the strong competition between superconductivity and magnetism induced by the spin polarization in SC. Of particular interest in the FM/SC/FM structure is not only to find novel magneto resistive effects due to the competition but also application to magnetoelectronics.

In this Letter we show that a FM/SC/FM double tunnel junction is a new magneto resistive device to control superconductivity by applying the bias voltage (or current). In the antiferromagnetic (A) alignment of magnetizations (see Fig. 1), a nonequilibrium spin density is induced in SC due to the imbalance in the tunneling currents carried by the spin-up and spin-down electrons, so that the superconducting gap $\Delta$ is reduced with increasing bias voltage and vanishes at a critical voltage $V_c$. In the ferromagnetic (F) alignment, however, there is no spin-density in SC. Consequently, TMR has a strong voltage dependence around $V_c$; TMR is enhanced compared with that in the normal state above $V_c$, while it changes sign to show an inverse TMR effect for some voltage range below $V_c$. It is shown that $V_c$ is inversely proportional to the spin polarization $P$ of FM ($V_c \propto 1/P$), which provides a new method for determining $P$ of FM.

We consider a FM/SC/FM double tunnel junction as shown in Fig. 1. The left and right electrodes are made of the same FM and the central one is a thin film SC. The magnetization of the left FM is chosen to point up and that of the right FM is either up or down. The voltages $-V_1$ and $V_2$ ($=V-V_1$) are applied to the left and right electrodes, respectively. We assume that the energy relaxation time $\tau_\gamma$ of quasiparticles in SC is shorter than the time $\tau_t$ between two successive tunneling events, whereas the spin relaxation time $\tau_s$ is longer than $\tau_t$. Consequently, the electrons tunneling into SC relax to the Fermi distribution before leaving SC, keeping their spin direction during the stay in SC.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Double tunnel junction consisting of two ferromagnets (FM) and a superconductor (SC) separated by insulating barriers in the case of identical barriers. Schematic densities of states of FMs (left and light) and SC (middle) in the antiparallel alignment of magnetizations in FMs are shown when SC is in the normal state (a) and in the superconducting state (b). $\delta\mu$ denotes the shift of the chemical potentials in SC.}
\end{figure}
We calculate the tunneling current using a phenomenological tunneling Hamiltonian. If SC is in the superconducting state, it is convenient to rewrite the electron operators $a_{k\sigma}$ in SC in terms of quasiparticle operators $\gamma_{k\sigma}$ appropriate to the superconducting states, using the Bogoliubov transformation, $a_{k\uparrow} = a_{k\uparrow} \gamma_{k\uparrow} + u_{k\uparrow}^\dagger \gamma_{-k\downarrow}$ and $a_{k\downarrow} = u_{-k\uparrow} \gamma_{k\downarrow} - v_{-k\downarrow}^\dagger \gamma_{-k\uparrow}$, where $|u_{k\uparrow}|^2 = \frac{1}{2} (1 + \xi_k/E_k)$, $|v_{-k\downarrow}|^2 = \frac{1}{2} (1 - \xi_k/E_k)$, and $E_k = |k^2 + \Delta^2|^{1/2}$ is the quasiparticle dispersion of SC, $\xi_k$ being the one-electron energy relative to the chemical potential and $\Delta$ being the gap parameter. Then, using the golden rule formula, we calculate the spin-dependent currents $I_{j\sigma}$ across the $j$th junction. The results are

\begin{align}
I_{1\uparrow} &= (G_{1\uparrow}/e)[N_1 - S - Q^*/2], \\
I_{1\downarrow} &= (G_{1\downarrow}/e)[N_1 + S - Q^*/2], \\
I_{2\uparrow} &= (G_{2\uparrow}/e)[N_2 + S + Q^*/2], \\
I_{2\downarrow} &= (G_{2\downarrow}/e)[N_2 - S + Q^*/2],
\end{align}

where $G_{j\sigma}$ is the tunnel conductance of the $j$th junction for electrons with spin $\sigma$ if SC is in the normal state. The quantity $N_j$ is given by the usual expression [10]

\begin{equation}
N_j = \int_\Delta \mathcal{D}_S(E_k) \left[ f_0(E_k - eV_j) - f_0(E_k + eV_j) \right] dE_k,
\end{equation}

where $\mathcal{D}_S(E_k) = E_k/\sqrt{E_k^2 - \Delta^2}$ is the normalized BCS density of states and $f_0(E_k \pm eV_j)$ is the Fermi distribution function of thermal equilibrium in FM. The quantity $S$ represents the spin density normalized to the density of states of SC in the normal state $D_N$ and is given by

\begin{equation}
S = \int_\Delta \mathcal{D}_S(E_k) \left( f_{k\uparrow} - f_{k\downarrow} \right) dE_k,
\end{equation}

where $f_{k\sigma} = \langle \gamma_{k\sigma}^\dagger \gamma_{k\sigma} \rangle$ is the distribution function of quasiparticles with energy $E_k$ and spin $\sigma$ in SC. The quantity $Q^*$ is the charge density normalized to $eD_N$ due to the imbalance in populations of electronlike and hole-like quasiparticles [11]

\begin{equation}
Q^* = 2 \sum_\sigma \int_\Delta (f_{k\sigma}^{\uparrow} - f_{k\sigma}^{\downarrow}) dE_k,
\end{equation}

where $f_{k\sigma}^{\uparrow}$ and $f_{k\sigma}^{\downarrow}$ represent $f_{k\sigma}$ in the electronlike ($k > k_F$) and holelike ($k < k_F$) branches of $E_k$, respectively.

In the limit of vanishing spin-flip scattering, the spin-up and spin-down currents are treated as independent channels. The conservation of the currents at junctions 1 and 2, $I_{1\sigma} = I_{2\sigma}$, yields the relations

\begin{align}
S &= \left[ (G_{1\uparrow} G_{2\downarrow} - G_{1\downarrow} G_{2\uparrow})/(\tilde{G}_{1\uparrow} \tilde{G}_{2\downarrow}) \right] (N_1 + N_2)/2, \\
Q^* &= \sum_\sigma \left[ (G_{1\sigma}/\tilde{G}_\sigma) N_1 - (G_{2\sigma}/\tilde{G}_\sigma) N_2 \right],
\end{align}

where $\tilde{G}_\sigma = G_{1\sigma} + G_{2\sigma}$. If the tunnel barriers are symmetric [see Fig. 1], $V_1 = V_2 = V/2$ and $N_1 = N_2 = N$, so that $Q^* = 0$ for both alignments. This is because the charge transport is symmetric at the two junctions where the injected and extracted charges are balanced. In the following we assume the identical tunnel barriers and neglect the effect of the charge imbalance.

The spin density $S$ depends strongly on whether the magnetizations in FMs are parallel or antiparallel. The spin density $S_A$ in the A-alignment ($G_{1\sigma} = G_{2-\sigma}$) satisfies the relation

\begin{equation}
S_A = PN,
\end{equation}

where $P = |G_{j\uparrow} - G_{j\downarrow}|/(G_{j\uparrow} + G_{j\downarrow})$ represents the spin polarization of FM [3]. The relation (7) implies that a finite $S_A$ is induced by applying the bias voltage. The $S_A \neq 0$ is a consequence of symmetry breaking of the spin transport in the A-alignment, and is realized when the distribution of the spin-up and spin-down quasiparticles is disequilibrium, i.e., $f_{k\uparrow} \neq f_{k\downarrow}$, as seen from Eq. (8). However, the spin density $S_F$ in the F-alignment ($G_{1\sigma} = G_{2\sigma}$) has no net spin-density in SC ($S_F = 0$) due to the symmetric spin transport at the junctions.

The distribution function $f_{k\sigma}$ is determined as follows. When the thickness of SC is much smaller than the spin diffusion length [4], the distribution of spin-up and spin-down quasiparticles is spatially uniform in SC. For $r_E < r_t < r_s$, the distribution is described by the Fermi function $f_0$, but the chemical potentials of the spin-up and spin-down quasiparticles are shifted oppositely by $\delta \mu$ from the equilibrium one (see Fig. 1) to produce the nonequilibrium spin density. Thus we write $f_{k\sigma}$ as

\begin{equation}
f_{k\uparrow} = f_0(E_k - \delta \mu), \quad f_{k\downarrow} = f_0(E_k + \delta \mu),
\end{equation}

In the normal state ($\Delta = 0$), from Eqs. (8), (6), and (4), we have $\delta \mu_A = S_A = 1/2eV$ in the A-alignment [6], whereas $\delta \mu_F = S_F = 0$ in the F-alignment.

We first discuss how the superconductivity is affected by the nonequilibrium spin imbalance in SC. The gap $\Delta$ in the nonequilibrium situation is determined by $f_{k\sigma}$ through the BCS gap equation [4]

\begin{equation}
1 / D_{N_{BSCS}} = \int_0^{k_F} d \xi_k 1 / E_k - f_{k\uparrow} - f_{k\downarrow},
\end{equation}

where $f_{k\sigma}$ is given by Eq. (8). We note that Eq. (8) with Eq. (9) is the same as that of SC in the paramagnetic limit if $\delta \mu$ is taken to be the Zeeman energy $\mu_B H$ [2]. The chemical potential difference $2\delta \mu$ plays the role of pair breaking energy. Therefore the superconductivity is destroyed in the A-alignment when $\delta \mu$ exceeds a certain critical value by increasing the voltage. To show this, we solve self-consistently Eqs. (6) and (9) with respect to $\Delta$ and $\delta \mu$, and obtain $\Delta$ and $S$ as functions of $V$.

Figure 2(a) shows the gap parameter $\Delta_A$ in the A-alignment as a function of bias voltage $V$ for $P = 0.4$, the spin polarization of Fe [3]. The quantity $\Delta_0$ denotes
the value of $\Delta_A$ for $P = 0$ at $T = 0$. The gap parameter $\Delta_A$ decreases with increasing $V$ and vanishes at the critical voltage $V_c$. At very low temperatures $\Delta_A$ becomes multi-valued in a certain range of $eV$ just below $2\Delta_0$: At $T = 0$ it has three solutions, $\Delta_A = \Delta_0$ and $\Delta_A = \Delta_0(1 - 2P^2 + 2P\sqrt{(eV/2\Delta_0)^2 + P^2 - 1})^{1/2}$, in the range $0.92 < eV/2\Delta_0 < 1$. When $V$ is increased (or decreased) at $T \sim 0$, an instability into a spatially inhomogeneous state with different $\Delta_A$ takes place at a certain voltage within the range. At $V_c$ where $\Delta_A = 0$ and $\delta \mu_A = \frac{1}{2}PeV_c$, Eq. (3) reduces to an implicit equation for $PeV_c$ and $T$, which gives a universal relation between $PeV_c/\Delta_0$ and $T/T_c$, as shown in the inset of Fig. 2(a). In particular, we have $PeV_c = \Delta_0/P$ at $T = 0$. Therefore, we can determine the spin polarization of FM by measuring $V_c$. Since the paramagnetic effect caused by spin accumulation becomes stronger with decreasing $T$, $V_c$ is not a monotonic function of $T$, but has a maximum at $T/T_c = 0.5$. Figure 2(b) shows the voltage dependence of the spin density $S_A$ in the A-alignment. The dotted line indicates the values of $S_A = \frac{1}{2}PeV_c$ in the normal state. As $T$ is lowered below $T_c$, $S_A$ is suppressed below $V_c$ by the opening of the energy gap. At and near $T = 0$, $S_A$ shows the S-shaped anomaly around $eV_c \sim 2\Delta_0$, which stems from the multiplicity of $\Delta_A$ shown in Fig. 2(a). The detailed behavior of the anomaly is shown in the inset of Fig. 2(b). In the F-alignment, $\Delta_F$ has no $V$ dependence and has the same value as $\Delta_A(V = 0)$.

We now calculate the tunneling current as a function of bias voltage $V$. From Eqs. (2a)-(2d), the total currents $I_F$ and $I_A$ in the F and A alignments are given by

\begin{align}
I_F(V) &= (G_{FN}/e)N(V, \Delta_F), \\
I_A(V) &= (G_{FN}/e)(1 - P^2)N(V, \Delta_A),
\end{align}

where $N(V, \Delta)$ is given in Eq. (2) and $G_{FN} = G_{F\uparrow} + G_{F\downarrow}$. It follows from Eqs. (7) and (11) that $I_A \propto S_A$, so that Fig. 2(b) also represent the $V$ dependence of $I_A$.

Figure 3(a) shows the voltage dependence of the differential conductance $G_F$ and $G_A$ for the F and A alignments at $T/T_c = 0.4$. The $G_F$ shows the ordinary dependence on $V$ expected for the constant gap $\Delta_F$. In contrast, because of the decrease in $\Delta_A$ with increasing voltage, $G_A$ increases with voltage more rapidly than $G_F$, forming a higher peak than $G_F$, and then decreases steeply. At $V_c$, $G_A$ jumps to the conductance $G_A^N$ in the normal state. The tunnel magnetoresistance (TMR) is calculated by the formula: TMR = $(G_F/G_A) - 1$. Using the values of Fig. 3(a), we obtain the $V$ dependence of TMR shown in Fig. 3(b). At $V = 0$ where $\Delta_A = \Delta_F$, TMR takes the same value as in the normal state. A deep negative dip appears at $eV/2\Delta_0 \sim 1$ where $\Delta_A$ steeply decreases, exhibiting an inverse TMR effect ($G_A > G_F$), and is followed by the discontinuous jump at $V_c$ above which TMR is highly enhanced compared to that in the normal state.

The relation $I_A \propto S_A$ in the A-alignment directly indicates that the superconductivity of SC is strongly suppressed with increase of injection current $I_A$. Using the relation and the result of $\Delta_A$ and $S_A$ in Fig. 2, we obtain $\Delta_A$ as a function of injection current $I_A$. Figure 4 shows

![FIG. 2. (a) Gap parameter $\Delta_A$ in SC as a function of bias voltage $V$ for different temperatures below $T_c$ in the antiferromagnetic alignment. The inset shows the critical voltage $V_c$ vs temperature $T$. (b) Spin density $S_A$ in SC as a function of $V$. The inset shows an enlarged view of $S_A$ around $eV/2\Delta_0 = 1$ at $T/T_c = 0$, 0.05, and 0.1.](image)

![FIG. 3. (a) Tunnel conductance as a function of bias voltage. The dashed and solid curves indicate the conductances $G_F$ and $G_A$ for the ferromagnetic and antiferromagnetic alignments, respectively. (b) Tunnel magnetoresistance (TMR) as a function of bias voltage. The dotted line indicates TMR = $P^2/(1 - P^2)$ in the normal state.](image)
the square of $\Delta_A$ as a function of $I_A$. Since $\Delta_A^2$ is proportional to the superfluid density and thus represents the critical current of SC.

The critical current suppression by spin injection has been observed in FM/SC heterostructures made of a high-$T_C$ SC and a ferromagnetic manganite with $P \sim 100\%$ [14,15]. The experimental result of $I_c$ is surprisingly similar to that shown in Fig. 4. This strongly suggests that the injection currents from FM build up the spin density in SC of the heterostructures. Since the spin density is accumulated more efficiently in the double-junction geometry, the FM/SC/FM double junctions using high-$T_C$ SCs and ferromagnetic manganites is quite promising to test our predictions.

Another candidate for SC in the FM/SC/FM junction is a thin film of clean SC with sufficiently long spin-relaxation time such as Al. The depression of the TMR effect is caused predominantly by spin relaxation due to spin-orbit scattering in SC. Here, we derive a condition for observing the TMR effect when SC is in the normal state. By balancing the population rate $(I_{a\uparrow} - I_{a\downarrow})/e$ with the relaxation rate $AdN\delta s_A/\tau_\delta$, where $\delta$ is the thickness of SC and $A$ the junction area, we obtain $s_A = \frac{1}{2}P_{eff} eV$ with the effective spin polarization $P_{eff} = P/\left((1 + \tau_\delta/\tau_\delta)ight)$ where $\tau_\delta = AdN h (R_{FN}/R_K)$ [13] with $R_K = h/e^2 \approx 26$ k$\Omega$ and $R_{FN} = 1/G_{FN}$. To retain a substantial value of $P_{eff}$, we are required to satisfy $\tau_\delta \approx \tau_\delta$. For the case of clean Al with the values of $\tau_\delta \sim 10^{-8}$ $s$ [14] and $D_N \sim 10^{22}/(eV\cdot cm^3)$, we have the condition $R_{FN}A \lesssim (63/m) \times 10^{-5}$ $\Omega$ $cm^2$ for the specific junction resistance $R_{FN}A$ and $d$ ($A$).

In the above calculations we have not considered the effect of Andreev reflection (AR) on the tunnel conductance, since the resistance of a tunnel junction with a thin insulating layer is much higher than that of a metallic contact, i.e., $R_{FN}A \gg \pi R_K/2k_F^2 \approx 10^{-11}$ $\Omega$ $cm^2$, where $k_F$ is the Fermi momentum. In metallic contacts with resistance comparable to $\pi R_K/2k_F^2$, on the other hand, the conductance is dominated by AR for $eV < 2\Delta$ [14]. Recently, the suppression of AR in FM/SC nanocontacts has been used to measure $P$ in ferromagnets [17].

In summary, we have studied the spin-dependent tunneling in a FM/SC/FM double tunnel junction. The spin imbalance in the tunneling currents gives rise to the nonequilibrium spin density in SC. The superconductivity is strongly suppressed with increase of bias voltage and destroyed at the critical voltage $V_c (\propto 1/P)$. The tunnel magnetoresistance exhibits an unusual voltage dependence around $V_c$ below $T_c$. The results predicted in this Letter provide a method for measuring the spin polarization of FM’s as well as for controlling superconductivity and TMR by application of bias voltage.

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1. R. Meservey and P. M. Tedrow, Phys. Rep. 238, 173 (1994); Phys. Rev. B 7, 318 (1973).
2. M. Julliere, Phys. Lett. 8, 225 (1975).
3. S. Maekawa and U. Gfver, IEEE Trans. Magn. 18, 707 (1982).
4. J.S. Moodera et al., Phys. Rev. Lett. 74, 3273 (1995).
5. T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. 139, L231 (1995).
6. M. Johnson and R. H. Silsbee, Phys. Rev. Lett. 55, 1790 (1985); Phys. Rev. B 37, 5326 (1988).
7. A.G. Aronov, Sov. Phys. JETP, 44, 193 (1976).
8. M. Johnson, Appl. Phys. Lett. 65, 1460 (1994).
9. A. Brataas et al., Phys. Rev. B 59, 93 (1999); J. Barna’s and A. Fert, Europhys. Lett. 44, 85 (1998); A.N. Korotkov and V.I. Safarov, Phys. Rev. B 59, 89 (1999); H. Imamura et al., Phys. Rev. B 59, 6017 (1999).
10. M. Tinkham, Phys. Rev. B 6, 1747 (1972); Introduction to Superconductivity (McGraw-Hill, New York, 1996).
11. The spin splitting in the normal state is described by the term $- \sum_{k\sigma} \sigma \delta \mu_{k\sigma} \delta g_{k\sigma}$, which is transformed into $- \sum_{k\sigma} \sigma \delta \mu_{k\sigma} \delta g_{k\sigma}$ by the Bogoliubov transformation. Thus the quasiparticle excitation energy is given by $E_k - \sigma \delta \mu$ in the superconducting state.
12. G. Sarma, J. Phys. Chem. Solids 24, 1029 (1963).
13. The tunneling time $\tau_\delta$ is identical to the inverse of the tunneling rate $\Gamma$ introduced by D.R. Heslinga and T.M. Klapwijk, Phys. Rev. B 47, 5157 (1993).
14. V.A. Vas’ko et al., Phys. Rev. Lett. 78, 1134 (1997).
15. Z.W. Dong et al., Appl. Phys. Lett. 71, 1718 (1997).
16. G.E. Blonder et al., Phys. Rev. B 25, 4515 (1982).
17. R.J. Soulen et al., Science 282, 85 (1998); S.K. Upadhyay et al., Phys. Rev. Lett. 81, 3247 (1998); M.J.M. de Jong and C.W.J. Beenakker, Phys. Rev. Lett. 78, 1137 (1997).