Josephson Effect due to Odd-frequency Pairs in Diffusive Half Metals

Yasuhiro Asano\textsuperscript{1}, Yukio Tanaka\textsuperscript{2,3} and Alexander A. Golubov\textsuperscript{4}

\textsuperscript{1}Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan
\textsuperscript{2}Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan
\textsuperscript{3}CREST, Japan Science and Technology Corporation (JST) Nagoya, 464-8603, Japan
\textsuperscript{4}Faculty of Science and Technology, University of Twente, 7500 AE, Enschede, The Netherlands

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The Josephson effect in superconductor / diffusive ferromagnet / superconductor (SFS) junctions is studied using the recursive Green function method in the regime of large exchange energy in a ferromagnet. Motivated by recent experiment [R. S. Keizer, et. al., Nature 439, 825 (2006)] we also address the case of superconductor / diffusive half metal / superconductor junctions. The pairing function in spin-singlet and triplet channels, the Josephson current and their mesoscopic fluctuations are calculated. We show that the spin-flip scattering at the junction interfaces opens the Josephson channel of the odd-frequency spin-triplet Cooper pairs. As a consequence, the local density of states in half metals has a large peak at the Fermi energy. Therefore odd-frequency pairs can be detected experimentally by using the scanning tunneling microscopy.

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Ferromagnetism and spin-singlet superconductivity are competing orders against each other because the exchange field breaks down the spin-singlet pairs. The Cooper pairs, however, do not always disappear under exchange fields. The Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state and the proximity effect in ferromagnets\textsuperscript{3,4,5,6,7,8,9,10} are typical examples. Under exchange fields, the pairing function oscillates and changes its sign in the real space. As a consequence, superconductor / ferromagnet / superconductor (SFS) junctions undergo the $0\rightarrow\pi$ transition with varying length of a ferromagnet or temperature. It was also predicted that the odd-frequency spin-triplet pairs appear in weakly polarized ferromagnet or temperature. It was also predicted that the odd-frequency spin-triplet pairs induced by the spin-flip scatterings are introduced at the interface can carry the Josephson current. In real samples, the pairing function oscillates and changes its sign in the real space. As a consequence, the Josephson current interfaces opens the Josephson channel of the odd-frequency spin-triplet Cooper pairs. As a consequence, the local density of states in half metals has a large peak at the Fermi energy. Therefore odd-frequency pairs can be detected experimentally by using the scanning tunneling microscopy.

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Half metal is an extreme case of completely spin polarized material because its electronic structure is insulating for one spin direction and metallic for the other. At a simple thought, the spin-singlet Cooper pairs would not be able to penetrate into half metals. However recent experiment\textsuperscript{11} showed the existence of the Josephson coupling in superconductor / half metal / superconductor (S/HM/S) junctions, where NbTiN was used as a s-wave superconducting electrode and CrO$_2$ as a half metal. Thus one has to seek a new state of Cooper pairs in half metals attached to spin-singlet superconductors. Prior to the experiment, Eschrig et. al\textsuperscript{12} have addressed this challenging issue. In the clean limit, they showed that the p-wave spin-triplet pairs induced by the spin-flip scattering at the interface can carry the Josephson current. In real S/HM/S junctions, however, half metals are in the diffusive transport regime; the elastic mean free path of an electron is much smaller than the size of a half metal. In addition, real S/HM/S junctions are close to the dirty limit because the coherence length in a ferromagnet may become comparable to the mean free path. In such diffusive half metals, the p-wave symmetry of Cooper pairs is not possible because the pair wave function is isotropic in the momentum space due to impurity scattering\textsuperscript{13}. In this paper, we provide a general theory for the Josephson effect in diffusive SFS junctions with arbitrary magnitude of the exchange field $V_{ex}$. When $V_{ex}$ is much larger than the superconducting pair potential at zero temperature $\Delta_0$, mesoscopic fluctuations of the Josephson current are much larger than the ensemble averaged value. In addition, we focus on interesting case of diffusive S/HM/S junctions. We show that the odd-frequency spin-triplet s-wave pairing state is realized in half metals and propose an experimental method to detect this property.

Let us consider the two-dimensional tight-binding model for a SFS junction as shown in Fig. 1(a). The vector $r = j\mathbf{x} + m\mathbf{y}$ points to a lattice site, where $\mathbf{x}$ and $\mathbf{y}$ are unit vectors in the $x$ and $y$ directions, respectively. In the $y$ direction, we apply the periodic boundary condition for the number of lattice sites being $W$. Electronic states in superconducting junctions are described by the mean-field Hamiltonian

$$H_{BCS} = \frac{1}{2} \sum_{\mathbf{r}, \mathbf{r}'} \left[ c_{\mathbf{r}}^\dagger \hat{h}_{\mathbf{r}, \mathbf{r}'} c_{\mathbf{r}'} - \overline{c}_{\mathbf{r}} \hat{h}_{\mathbf{r}, \mathbf{r}'} \overline{c}^\dagger_{\mathbf{r}'} \right] + \frac{1}{2} \sum_{\mathbf{r} \in S} \left[ c_{\mathbf{r}}^\dagger \Delta \overline{c}_{\mathbf{r}} - \overline{c}_{\mathbf{r}} \Delta^* \overline{c}_{\mathbf{r}} \right],$$

with $\overline{c}_{\mathbf{r}} = (c_{\mathbf{r}, \uparrow}, c_{\mathbf{r}, \downarrow})$, where $c_{\mathbf{r}, \sigma}$ is the creation (annihilation) operator of an electron at $\mathbf{r}$ with spin $\sigma = (\uparrow$ or $\downarrow)$. $\Delta$ means the transpose of $\overline{c}$, $\delta_0$ for $l = 1 - 3$ are the Pauli’s matrices, and $\delta_0$ is $2 \times 2$ unit matrix. The hopping integral $t$ is considered among nearest neighbor sites in both superconductors and ferromagnets. In a ferromagnet, the on-site scattering potentials are given randomly in the range of $-V_1/2 \leq \epsilon_r \leq V_1/2$ and the uniform exchange potential is given by $V(r) = V_{ex} \theta_3$, where $\theta_l$ for $l = 1 - 3$ is unit vector in a spin space. The Fermi energy $\mu$ is set to be $2t$ in a normal metal with $V_{ex} = 0$, while a ferromagnet and a half metal are respectively described by $V_{ex}/t = 1$ and $2.5$ in Fig. 1(b). The spin-flip scatterings are introduced at $j = 1, 2, L_N - 1$, and...
Throughout this paper we fix the following parameters: $L_N = 74$, $W = 25$, $\mu = 2t$, $V_f = 2t$ and $\Delta_0 = 0.005t$. This parameter choice corresponds to the diffusive transport regime in the N, F and HM layers. The results presented below are not sensitive to variations of these parameters.

We first discuss the Josephson current in SFS junctions as shown in Fig. 2(a) for $T = 0.1T_c$ where $T_c$ is the transition temperature. We assume that the spin-flip scattering at the interfaces is absent (i.e., $V_S = 0$) and fix the phase difference across the junctions $\varphi$ equal to $\pi/2$. The results are normal-
\[ \delta J \] approximately corresponds to the typical amplitude of the Josephson current expected in a single sample. The relation \( \langle J \rangle = 0 \) has different meaning for SFS and S/HM/S cases. In SFS junctions, \( \langle J \rangle = 0 \) at the transition points is the result of the ensemble averaging and the Josephson current remains finite in a single sample. The characteristic temperature and length of a ferromagnet at the \( 0 - \pi \) transitions vary from one sample to another. In S/HM/S junctions at \( V_{ex} = 2.5 t \), however, \( \langle J \rangle = 0 \) means vanishing Josephson current even in a single sample because \( \langle J \rangle = \delta J = 0 \).

The origin of large fluctuations of the Josephson current can be understood by considering the behavior of the pairing function in a ferromagnet. The pairing function in Eq. (3) can be decomposed into four components,

\[
\frac{1}{W} \sum_{m=1}^{W} \hat{f}_{\omega_n}(r, r') = i \sum_{\nu=0}^{3} f_{\nu}(j) \hat{\sigma}_\nu \hat{\sigma}_2,
\]

where \( f_{\nu}(f_3) \) is the pairing function of the spin-singlet (spin-triplet) pairs with the spin structure of \(|\uparrow\downarrow\rangle - (+) |\downarrow\uparrow\rangle\)/\(\sqrt{2} \), respectively, and the pairing function of \(|\uparrow\uparrow\rangle \) (\(|\downarrow\downarrow\rangle \)) is given by \( f_{1\uparrow} = i f_2 - f_1 \) (\( f_{1\downarrow} = i f_2 + f_1 \)). In Figs. 2(b) and (c), we show \( \langle f_0 \rangle \) and \( \delta f_0 \) as a function of position \( j \) in a diffusive ferromagnet, where \( f_0 \) is the pairing function in bulk superconductor, \( \omega_n \) is fixed at 0.02\( \Delta_0 \), \( V_S = 0 \), and \( \varphi = 0 \).

The junction interface and the center of a ferromagnet correspond to \( j = 1 \) and \( j = 37 \), respectively. In SNS junctions (Fig. 2(b)), \( \langle f_0 \rangle \) is larger than \( \delta f_0 \) and very weakly decays with \( j \). The spin-singlet Cooper pairs exist everywhere in a normal metal. Near the interface, \( \delta f_0 \) is slightly suppressed due to the tight contact to the superconductor. On the other hand, in SFS junctions in (Fig. 2(b)), the average \( \langle f_0 \rangle \) decreases exponentially with \( j \) according to \( \exp(-j/\xi_h) \) as indicated by a broken line. The fact that \( \delta f_0 \) remains finite at the center of a ferromagnet means that the spin-singlet pair penetrates far beyond \( \xi_h \) even though \( \langle f_0 \rangle \sim 0 \) there.

In Fig. 2(d), we show the pairing function for three different realization of disorder in SFS junctions. The phase difference is in the presence of disorder even for \( j \gg \xi_h \), whereas they are out of phase far from the interface. We obtain the relation \( \delta f_0 \propto e^{-j/\xi_h} \) with \( \xi_T = \sqrt{D/2\omega_n} \), in agreement with Ref. [2]. Thus we conclude that spin-singlet Cooper pairs do exist in a single sample of ferromagnet even for \( j \gg \xi_h \) and the mesoscopic fluctuations of the pairing function provide the origin of the large fluctuations in the Josephson current. In S/HM/S junctions for \( V_{ex} = 2.5 t \) as shown in Fig. 3(b), both \( \langle f_0 \rangle \) and \( \delta f_0 \) vanish for \( j \gg 1 \), which indicates the absence of spin-singlet Cooper pairs in a half metal.

The relation \( \langle J \rangle \ll \delta J \) is the characteristic feature of the Josephson current in diffusive SFS junctions. This feature, however, is drastically changed by the spin-flip scattering at the interfaces. In Figs. 3(a) and (b) we show \( \langle J \rangle \) and \( \delta J \) vs \( V_S \) for \( V_{ex}/t = 1 \) and 2.5, respectively. In both cases (a) and (b), we find that \( \langle J \rangle \geq \delta J \) for \( V_S \geq 0.3 t \). The Josephson current becomes self-averaging in the presence of the spin-flip scattering. The reason can be explained by calculating the pairing functions of equal-spin pairs shown in Figs. 3(c) and (d), where \( f_{\nu} \) is plotted as a function of position \( j \). Here we show \( \delta f_0 \) and \( \delta f_3 \) instead of \( \langle f_0 \rangle \) and \( \langle f_3 \rangle \) because the ensemble averages are much smaller than their fluctuations. The fast decay of \( \delta f_0 \) and \( \delta f_3 \) is determined by strong spin polarization. In both cases (c) and (d), \( \langle f_{1\uparrow} \rangle \) becomes larger than \( \delta f_0 \) and \( \delta f_3 \) because the pairing function \( f_{1\uparrow} \) does not change sign for various impurity configurations. Thus the averaged quantities become larger than their fluctuations. Thus the Josephson current becomes self-averaging as shown in Figs. 3(a) and (b).

Finally we address an unusual symmetry property of the Josephson current in S/HM/S junctions. In Fig. 4(a), we show \( \langle f_{1\uparrow} \rangle \) as a function of \( \omega_n \), where \( j = 37 \), \( V_S = 0.2 t \), \( \varphi = 0 \), and \( V_{ex} = 2.5 t \). For comparison, we also show \( \langle f_0 \rangle \) on the normal side of a SNS junction. The pairing function \( \langle f_0 \rangle \) in a normal metal is the even function of \( \omega_n \), whereas \( \langle f_{1\uparrow} \rangle \) in a half metal is the odd function of \( \omega_n \) Ref. [4]. The pairing function obeys the Pauli’s rule

\[
\hat{f}_{\omega_n}(r, r') = -\hat{f}_{-\omega_n}(r', r),
\]

where \( \hat{f} \) denotes the transpose of \( f \) meaning the interchange of spins. It is well known that ordinary even-frequency pairs are classified into two symmetry classes: the spin-singlet even-parity and the spin-triplet odd-parity one. In the former case, the negative sign arises due to the interchange of spins, while in the latter case due to \( r \leftrightarrow r' \). In the present calculation, all components on the right hand side of Eq. (5) have
the $s$-wave symmetry. The pairing functions are isotropic in both the real and momentum spaces due to diffusive impurity scattering. As a result, $f_{1\uparrow\uparrow}$ must be the odd function of $\omega_n$ to obey the Pauli’s rule. Both even- and odd-frequency pairs are mixed in ferromagnets as shown in Fig. 3(c). The fraction of odd-frequency pairs depends on parameters such as the exchange potential and the spin-flip scattering. On the other hand, in a diffusive half metal all Cooper pairs have the odd-frequency character, which causes drastic change in the quasiparticle density of states.

The density of states is given by

$$N(E, j) = \frac{1}{\pi W} \sum_{m=1}^{W} \text{ImTr} G_{E+i\gamma}(r, r),$$

where $\gamma$ is a small imaginary part chosen to be $0.05\Delta_0$ in the following. In Fig. 4(b), the local density of states (LDOS) at $j = 37$ is shown, where $\varphi = 0$ and $N_0$ is the density of states in the normal state at $V_{ex} = 0$. For comparison, we show LDOS on the normal side of SNS junction with $V_S = 0$ which has a minigap at $E < E_{Th} \sim 0.3\Delta_0$, where $E_{Th}$ is the Thouless energy. In contrast to that, LDOS in a half metal has a peak at the Fermi energy, it’s width is characterized by $E_{Th}$. This peak is generated at the spin active interface by the mechanism discussed in Ref. [24] and is transferred into a half metal due to long range property of odd-frequency spin-triplet even-parity pairing function. The peak is much stronger than the enhancement of the LDOS found in weak ferromagnets [10,21]. In addition, in a half metal the peak shape is almost independent of position, while in the SF junctions the LDOS has an oscillatory peak/dip structure at $E = 0$ which rapidly decays with the distance from the SF interface. Therefore the large peak at $E = 0$ in LDOS is a robust and direct evidence of the odd-frequency pairing in half metals. To test the existence of such peculiar pairing state, the scanning tunneling spectroscopy could be used.

In conclusion, we have studied Josephson effect in superconductor /diffusive ferromagnet /superconductor junctions by using the recursive Green function method. The Josephson current in these junctions basically is not self-averaging because the spin-singlet Cooper pairs penetrating into ferromagnets far beyond $\xi_h$ cause the large fluctuations of the pairing function. In the presence of the spin-flip scattering at the interfaces, the equal-spin odd-frequency pairs drastically suppress the fluctuations. When ferromagnets are half-metallic, all Cooper pairs have the odd-frequency property. As a result, the low energy peak in the quasiparticle density of states in a half metal exists at the distances far beyond $\xi_h$ from the interface and could be probed by scanning tunneling spectroscopy.

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