Long-range proximity effect for opposite-spin pairs in S/F heterostructures under non-equilibrium quasiparticle distribution

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By now it is known that in a singlet superconductor/ferromagnet (S/F) structure the proximity effect is negligible at distances exceeding the magnetic coherence length \( \xi_F = \sqrt{D/h} \) (See, for example, [1] and references therein). Here \( D \) is the diffusion constant and \( h \) is the exchange energy of the ferromagnet. For the most part of the ferromagnets, which are used for S/F heterostructures (including weak ferromagnetic alloys like CuNi [2] and PdNi [3]) this length is much shorter than the normal metal coherence length \( \xi_N = \sqrt{D/2\pi T} \). This suppression of the proximity effect can be understood as follows. If the magnetization direction is homogeneous in the considered system, then the Cooper pairs, penetrating into the nonsuperconducting region due to the spin rotation of one of the paired electrons. The latter component penetrates into the ferromagnet over a large distance, which can be of the order of \( \xi_N \) in some cases. The reason is that it corresponds to the correlations of the type \( |\uparrow\uparrow\rangle \) with parallel spins and is not as sensitive to the exchange field as the opposite-spin correlations. Various superconducting hybrid structures, where this type of long-range proximity effect (LRPE) can arise, were considered in the literature (See Refs. [5], [6], [1] and references therein). In addition, the LRPE was theoretically predicted in structures containing domain walls [5,6], spin-active interfaces [3,10], spiral ferromagnets [11-12] and multilayered SFS systems [14,15]. There are several experimental works, where the long-range Josephson effect [16-18] and the conductance of a spiral ferromagnet attached to two superconductors [19] were measured.

In the present paper we show that LRPE at an S/F interface can be generated not only by equal-spin pairs with \( S_z = \pm 1 \). It is also created by opposite-spin pairs with \( S_z = 0 \) under the condition that the appropriate non-equilibrium and spin-dependent quasiparticle distribution is produced and maintained in the ferromagnet. At first we concentrate on the physical essence of the effect and after that turn to the exact calculation of the Josephson current through a S/F/S junction under the corresponding conditions.

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As it was discussed above, the source of the rapid decay of an opposite-spin Cooper pair in the ferromagnet is the impurity averaging of the rapidly oscillating pair wave function. In turn, the reason of these rapid oscillations is the non-zero pair momentum \( Q \). It is inevitably acquired by the pair of electrons, which have the same energy (in the particular case \( \varepsilon = 0 \)) and opposite spins upon entering the F region. Now let us assume that the spin-dependent quasiparticle distribution \( f_{\uparrow,\downarrow}(\varepsilon) = 1/[1 + \exp[(\varepsilon \pm eV)/T]] \) is created in the ferromagnet. The energy is counted from the chemical potential of the superconductor. Then the electrons forming a pair, which is located at the Fermi level \( \varepsilon = 0 \) in the superconductor, can only enter the F region with different energies \( \varepsilon_{\uparrow,\downarrow} = \mp eV \), thus conserving the total energy of the pair. As a result, the difference between the spin-up and spin-down electron momenta is modified and in this case \( Q \propto (h - eV)/\sqrt{\varepsilon} \). Therefore, the creation of appropriate spin-dependent quasiparticle distribution with \( eV = h \) in the ferromagnet makes the electrons enter the F region with different energies, but with equal (in absolute value) momenta. Thus, the additional rapid decay of an opposite-spin Cooper pair in the ferromagnet is absent and the decay length can be close to \( \xi_N \).

It is worth noting here that this physics is similar to some extent to the effect discussed recently in Ref. [21], where it was found that for a thin superconducting film the destructive effect of the exchange field can be fully compensated by the creation of spin-dependent quasiparticle distribution in it. The effect reported in [21] and the LRPE discussed here are two aspects of the same problem: coexistence of singlet superconductivity and ferromagnetism under nonequilibrium spin-dependent distribution.

The discussed LRPE has a profound impact on the Josephson current through an S/F/S junction under the condition of the appropriate quasiparticle distribution in the F layer. Now we turn to quantitative analysis of this effect. We consider a plane diffusive junction of two s-wave superconductors with the F interlayer, which is in the parameter range \( |\Delta| \ll h \ll \varepsilon_F \), where \( \varepsilon_F \) is the Fermi energy of the ferromagnet. As we consider a non-equilibrium system, we make use of Keldysh framework of the quasiclassical theory, where the fundamental quantity is the momentum average of the quasiclassical Green’s function \( \bar{g}(x, \varepsilon) = \langle \bar{g}(p_f, x, \varepsilon) \rangle_{p_f} \). Here \( x \) is the coordinate normal to the S/F interface and \( x = 0 \) is the middle of the F layer. In the interlayer \( g(x, \varepsilon) \) obeys the Usadel equation [21]

\[
\frac{D}{\pi} \partial_x (\bar{g} \partial_x \bar{g}) + \left[ \varepsilon \tau_0 \sigma_0 \rho_0 + h \hat{\sigma} \rho_0, \bar{g} \right] = 0 ,
\]

where \( \tau_i, \sigma_i \), and \( \rho_i \) are Pauli matrices in particle-hole, spin and Keldysh spaces, respectively. \( \hat{\sigma} = \sigma(\tau_0 + \tau_3)/2 + \sigma(\tau_0 - \tau_3)/2 \) is the spin operator for a quasiparticle. Eq. (1) should be supplied with the normalization condition \( \bar{g}^2 = -\pi^2 \tau_0 \sigma_0 \rho_0 \). The Usadel equation in the interlayer should be also supplemented by Kupriyanov-Lukichev boundary conditions at SF interfaces [22]: \( \partial_x \bar{g} = -\alpha(R_F/R_0 d_F) \bar{g}, \partial_x \bar{y}, \bar{y} \). Here \( R_0 \) and \( R_F \) stand for the resistances of the S/F interface and the F interlayer, \( \alpha = +1 (-1) \) at the left (right) interface, \( \bar{y}_S \) is the value of the Green’s function at the superconducting side of the corresponding boundary.

It is convenient to express Keldysh part of the full Green’s function via the retarded and advanced components and the distribution function: \( \bar{g} = \bar{g}_R \bar{g} - \bar{g}_A \bar{g}^* \). The distribution function is diagonal in particle-hole space: \( \bar{g} = \bar{g}(\tau_0 + \tau_3)/2 + \bar{g}_2 \bar{g}_2(\tau_0 - \tau_3)/2 \). The hole component \( \bar{g}_2 \bar{g} \) of the distribution function is connected to \( \bar{g} \) by general symmetry relation [23] \( \bar{g} = -\sigma_2 \bar{g}(-\varepsilon) \sigma_2 \). In the particle-hole space the retarded and advanced Green’s functions take the form \( \bar{g}_R = \bar{g}_R(\tau_0 + \tau_3)/2 + \bar{f}_R \bar{f}_R(\tau_0 + \tau_3)/2 + \bar{g}_R(\tau_0 - \tau_3)/2 \). For the junction under consideration the electric current through it can be written as follows

\[
j = -\frac{d}{eR_F} \int_{-\infty}^{+\infty} d\varepsilon \left\{ \left[ \int \partial_x j_R^{\bar{f}}(\bar{x}) R - (\partial_x j_R^{\bar{f}}) R \right] \bar{g}^* \right. \\
- \left. \left[ \int \partial_x j_A^{\bar{f}}(\bar{x}) A + (\partial_x j_A^{\bar{f}}) A \right] \bar{g} \right\} .
\]

We assume that the direction of the exchange field \( \bm{h} \) is spatially homogeneous and choose the quantization axis along the field. In this case equal-spin pairs do not occur in the interlayer. The distribution function and the normal part \( \bar{g}_R \bar{A} \) of the Green’s function are diagonal matrices in spin space. The anomalous Green’s functions only contain singlet and \( S_z = 0 \) triplet components and can be represented as \( \bar{f}_R^{\bar{A}} = \bar{f}_R^{\bar{A}} i \sigma_2 \) and \( \bar{f}_A^{\bar{A}} = -i \sigma_2 \bar{f}_A^{\bar{A}} \), where \( \bar{f}_R^{\bar{A}} \) and \( \bar{f}_A^{\bar{A}} \) are diagonal in spin space. Further, we assume that \( d_F \gg \xi_F \). This is the most reasonable regime to demonstrate the LRPE.

The anomalous Green’s function can be easily found analytically in the middle part of the interlayer up to the first order in the parameter \( \exp[-d_F/2 \xi_F] \ll 1 \). It takes the form (\( \sigma = \pm 1 \))

\[
(f_d^{R,A})_\sigma = \sum_{\alpha=\pm 1} 4\pi \kappa e^{-i\alpha\chi/2} K^{R,A}_\sigma e^{-\lambda^{R,A}_\sigma(\alpha x + d_F/2)}
\]

where \( \chi \) is the superconducting order parameter phase difference between the leads and \( \lambda^{R,A}_\sigma = \sqrt{-2i\kappa (\varepsilon + \sigma \hbar)} /D \) with \( \kappa = +1 (-1) \) for the retarded (advanced) functions. \( K^{R,A}_\sigma \) should be found from the boundary conditions for a given S/F interface without taking into account the influence of the other S/F interface. It is determined by the equation

\[
\lambda^{R,A}_\sigma K^{R,A}_\sigma (1 - K^{R,A}_\sigma) = \frac{1}{4\gamma_0} \left[ \sin \theta^{R,A}_\sigma (1 + 6K^{R,A}_\sigma^2) + K^{R,A}_\sigma + K^{R,A}_\sigma^4 \right] - \cosh \theta^{R,A}_\sigma 4K^{R,A}_\sigma (1 + K^{R,A}_\sigma^2) ,
\]
where $\gamma_0 = R_0 d_F / R_F$, $\cosh \Theta_S^{R,A}$ and $\sinh \Theta_S^{R,A}$ originate from the normal and anomalous Green’s functions at the superconducting side of S/F interfaces. We assume that the parameter $(R_F \xi_S / R_0 d_F) / (\sigma_F / \sigma_s) \ll 1$, where $\xi_S = \sqrt{D/\Delta}$ is the superconducting coherence length in the leads, $\sigma_F$ and $\sigma_S$ stand for conductivities of ferromagnetic and superconducting materials, respectively. It allows us to neglect the suppression of the superconducting order parameter in the S leads near the interface and take the Green’s functions at the superconducting side of the boundaries to be equal to their bulk values: $\cosh \Theta_S^{R,A} = -\kappa \varepsilon / \sqrt{|\Delta|^2 - (\varepsilon + \kappa i 0)^2}$, $\sinh \Theta_S^{R,A} = -\kappa |\Delta| / \sqrt{|\Delta|^2 - (\varepsilon + \kappa i 0)^2}$.

However, approximation (3)-(4) is only valid for $|\exp(-\lambda^{R,A}_b d_F/2)| / \exp[-d_F/2|a_S|] \ll 1$. That is, it is not valid if $|\varepsilon + \sigma_i \hbar| \lesssim \Delta$. If one studies equilibrium problems, this high energy region practically does not contribute to the Josephson current and can be neglected. At the same time, for the problem we consider it is the most important energy region for the regime $eV \approx \hbar$, as it is shown below. It appears that in this resonant energy region the solution can also be easily found analytically taking into account that for high energies $|\varepsilon| \sim \hbar \gg \Delta$ the anomalous Green’s function in the superconductor is small: $\sinh \Theta_S^{R,A} \sim (\Delta / \varepsilon) \ll 1$. Therefore, the solution for $f^{R,A}$ in the interlayer region can be found up to the first order in this parameter. It can be also expressed by Eq. (3), but $K^{R,A}_{\sigma}$ takes the form

$$K^{R,A}_{\sigma} = \frac{\sinh \Theta^{R,A}_{\sigma}}{4\gamma_b} \times \frac{\lambda^{R,A}_b + \gamma_b^{-1}}{(\lambda^{R,A}_b + \gamma_b^{-1})^2 - e^{-2\xi_{S,N}^{-1}} d_F (\lambda^{R,A}_b - \gamma_b^{-1})^2}. \tag{5}$$

Now let us turn to the discussion of the distribution function. In order to create the spin-dependent quasiparticle distribution in the interlayer one can attach two additional half metal (HM) electrodes to the F region (see Fig. 1(a)) and apply a voltage bias $2V$ between them. The magnetization of one of the HM’s is directed along with the exchange field of the interlayer and the magnetization of the other one is opposite. We neglect energy relaxation in the interlayer, that is assume that the time $\tau_{exc}$, which an electron spends in the F region is much less than the energy relaxation time $\tau_e$. Spin relaxation processes are also not taken into account. We discuss their influence below. Then it can be calculated that the distribution function in the film takes the form

$$\varphi_\sigma = \tanh \frac{\varepsilon + \sigma eV}{2T}. \tag{6}$$

In this case $\varphi_{\sigma} = \varphi_\sigma$. Eq. (6) has a simple physical interpretation. For spin-up subband the main voltage drop occurs at one of HM, while for spin-down subband - at the other. As a result, the distribution functions for spin-up and spin-down electrons in the interlayer are to be close to the equilibrium form with different electrochemical potentials. For our special case of HM/F/HM structure the resulting chemical potential of the F region is equal to the chemical potential of the superconducting leads. Indeed, the sum of the distribution functions in the two spin subbands is symmetric with respect to zero energy.

It is worth noting here that the distribution function has such a one-step shape (in each of the spin subbands) due to the fact that the additional electrodes are HM: the electrons from spin-up (spin-down) subband can flow only to/from the top (bottom) electrode. In this case the LRPE effect is maximal. However, the nonequilibrium LRPE can be also observed if one takes strong ferromagnets or even normal metals instead of HMs. We discuss these cases below.

![FIG. 1. (a) Scheme of the system under consideration. (b) Critical Josephson current as a function of $eV$. $h = 8\Delta$ (solid line), 6$\Delta$ (dashed line). $d_F = 3\xi_S$. (c) Critical Josephson current as a function of $d_F$. $h = 8\Delta$ (solid line), 6$\Delta$ (dashed line). (d) SC DOS $N_\uparrow$ for the spin-up subband as a function of $\varepsilon$ at $h = 6\Delta$ and $d_F = 2.5\xi_S$ (black line), 1.5$\xi_S$ (gray line). $N_\uparrow(\varepsilon) = -N_\downarrow(-\varepsilon)$. $T = 0.01\Delta$ and $\gamma_0 = 10$ for all the plots.](image-url)
reducing factor \((\Delta/h)^2\). This is originated from the fact that the superconducting correlations in the leads are suppressed by the factor \(\Delta/\varepsilon\) for large enough energies \(\varepsilon \sim h\), which are important for the LRPE.

It is worth noting here that, in addition to the sharp increase of the current at \(eV = h\), the current manifests a number of 0–\(\pi\) transitions as a function of \(eV\). The region of small voltages \(|eV| < \Delta\) has been studied in detail in Ref. \(^{22}\).

The dependence of the critical currents \(j_\parallel\) and \(j_\perp\) on the junction length \(d_F\) is plotted in Fig. 1(c) in the logarithmic scale. As it is well-known \(^{19}\), \(j_\parallel\) exhibits oscillations with a period \(2\pi\xi_F\) and simultaneously decays exponentially on the length scale of \(\xi_F\). At the same time \(j_\perp\) does not oscillate. It decays exponentially on the length scale of \(\xi_N\). In order to study in more detail this LRPE we plot in Fig. 1(d) the supercurrent-carrying density of states \(N_s(\varepsilon)\) (SCDOS). This quantity represents the density of states weighted by a factor proportional to the current that each state carries in a certain direction \(^{23,28}\).

The full current can be represented as the integrated over energy (and summed up over spin subbands) product of the SCDOS and the distribution function. It is seen from Fig. 1(d) that the amplitude of the SCDOS low-energy part (corresponding to \(|\varepsilon| \lesssim \Delta\), which determines the Josephson current under equilibrium conditions, diminishes very strongly as a function of the junction length due to the suppression by the factor \(\exp[-d_F/\xi_F]\). At the same time the SCDOS have sharp peaks at energies \(\varepsilon = \pm h\), which correspond to the paired states with zero total momentum and, therefore, are not suppressed by the factor \(\exp[-d_F/\xi_F]\). Under equilibrium conditions these parts of the SCDOS multiplied by the corresponding distribution function \(\tanh[\varepsilon/2T]\) give very small contribution into the current. On the contrary, shifting the argument \(\varepsilon\) of the distribution function by \(\pm eV\) for spin-up and spin-down spin-subbands one makes the peaks to give the maximal contribution to the current.

Now we discuss briefly the influence of spin relaxation, which can take place in the interlayer, on the Josephson current. It influences directly the distribution function: (i) reduces the height of the main step of the distribution function \(\varphi_{\uparrow,\downarrow}\) at \(\varepsilon_{\uparrow,\downarrow}^{\text{main}} = \mp eV\) and (ii) gives rise to an additional step of the distribution function \(\varphi_{\uparrow,\downarrow}^{\text{add}}\) at \(\varepsilon_{\uparrow,\downarrow}^{\text{add}} = \pm eV\). The correction to the distribution function can be roughly estimated as \(\delta\varphi_{\uparrow,\downarrow} = \mp[(\tau_{\text{esc}}/\tau_{sf})(\varphi_{\uparrow,\downarrow} - \varphi_{\downarrow,\uparrow})]/(1 + 2\tau_{\text{esc}}/\tau_{sf})\). Here \(\varphi_{\uparrow,\downarrow} - \varphi_{\downarrow,\uparrow}\) is defined by Eq. \(^{19}\) and \(\tau_{sf}\) is the characteristic spin relaxation time. By looking at Fig. 1(d) it is easy to see that such modification of the distribution function does not qualitatively modify the result for \(j_\parallel\), but only reduces its magnitude by the factor \((\tau_{sf} + \tau_{\text{esc}})/(\tau_{sf} + 2\tau_{\text{esc}})\). The additional current peak of small height \(j_\parallel\tau_{\text{esc}}/(\tau_{sf} + 2\tau_{\text{esc}})\) can also appear at \(eV = -h\). This is the essential difference between the LRPE discussed here and the superconductivity recovered by the nonequilibrium distribution, discussed in \(^{20}\). While in the later case spin relaxation processes lead to the effective reduction of the coupling constant \(\lambda \to \lambda_{eff} = \lambda(1 + \tau_{\text{esc}}/\tau_{sf})^{-1}\) and, therefore, can destroy the effect quite rapidly, the Josephson current discussed here is much more stable against their influence.

Analogous modification of the Josephson current can be observed if one uses strong ferromagnets instead of HM’s for generation of the spin-dependent quasiparticle distribution in the interlayer. In this case the nonequilibrium distribution function inside the interlayer is represented by a sum of the distribution functions coming from the top and bottom electrodes, weighted by factors depending on the interface transparencies (this is a double-step structure). In general, if inelastic energy relaxation can be neglected in the interlayer, the distribution function at low temperatures manifests \(n\) steps of different height at different energies \(\varepsilon_n\). In this case instead of one peak of maximal height at \(eV = h\) the LRPE generated critical current (as a function of \(V\)) would exhibit \(n\) peaks of the corresponding height. Therefore, these peaks of the critical current can provide information about the particular distribution function, created in the interlayer. For example, if normal metals are used for additional electrodes instead of HM’s, the resulting distribution function manifests a double-step spin-independent structure \(\varphi(\varepsilon) = (1/2)\{\tanh[(\varepsilon - eV)/2T] + \tanh[(\varepsilon + eV)/2T]\}\), measured in \(^{24}\). Under the nonequilibrium distribution of such type the LRPE generated critical current would manifests two peaks of the same height \(j_\parallel/2\) at \(eV = \pm h\) instead of one peak \(j_\parallel\) at \(eV = h\), as it should be for the one-step spin-dependent distribution. It is worth noting here that the Josephson current under the above-mentioned spin-independent distribution has been already studied at \(h \ll \Delta\) in \(^{30}\). However, in this limit one cannot speak about the LRPE generated by nonequilibrium distribution because there is no rapid extra decay for such extremely small exchange fields.

In summary, we have predicted a new type of LRPE, which can take place in S/F heterostructures under nonequilibrium conditions. The condensate wave function in the F region is generated by opposite-spin Cooper pairs and equal-spin pairs are not involved. The possibility for an opposite-spin pair to penetrate into the ferromagnet over a large distance \(\sim \xi_N\) is provided by creation of the proper non-equilibrium quasiparticle distribution there. The LRPE can be observed as a sharp increase (up to a few orders of magnitude) of the critical Josephson current through a S/F/S junction under the condition that the voltage controlling the nonequilibrium distribution in the F interlayer is adjusted appropriately.

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