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Note on possibility of proximity induced spontaneous currents in superconductor/normal metal heterostructures[†]

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We analyse the possibility of the appearance of spontaneous currents in proximated superconducting/normal metal (S/N) heterostructure when Cooper pairs penetrate into the normal metal from the superconductor. In particular, we calculate the free energy of the S/N structure. We show that whereas the free energy of the N film F_N in the presence of the proximity effect increases compared to the normal state, the total free energy, which includes the boundary term F_B, decreases. The condensate current decreases F_N, but increases the total free energy making the current-carrying state of the S/N system energetically unfavorable.

1. Introduction

Penetration of Cooper pairs into the normal metal (N) in superconductor/normal metal (S/N) heterostructures, provided the interface transparency is not too small, is a well-known effect [1–4]. This so-called proximity effect (PE) is related to the Andreev reflections of electrons at the interface of the S/N bilayer [5]. In particular, the depth of Cooper pairs penetration into the N in the diffusive case is equal to $\xi_N \cong \sqrt{D_N/2\pi T}$ where $D_N = vl/3$ is the diffusion coefficient, and becomes smaller if the condensate moves. The proximity effect is utilized in various S/N/S Josephson junctions [6–8] and other superconducting devices [9–12] as it leads to a number of interesting physical phenomena. The most famous of these is the advantageous Josephson coupling in S/N/S Josephson junctions with the N layer being significantly thicker (up to a few microns) than the insulating (I) barrier in tunnel Josephson junctions [6–8]. Furthermore, in contrast to the conventional tunnel S/I/S junction, the properties of the Josephson S/N/S junctions can be modified by varying the characteristics of the normal metal layer. For example, if there is an exchange field in the N metal, i. e. a ferromagnetic metallic layer F is used, then the critical current I_c may even change sign [12–17], yielding the so-called π-junctions. Note, the change of sign in I_c may also be achieved in conventional S/N/S multi-terminal Josephson junctions if the electric potential of the normal metal N is shifted with respect to the S counterparts [9,18–21]. More recently, spectrum of Andreev bound states in S/N-multiterminal structures with potentially non-trivial topological bands with Weyl points was also investigated [22, 23].

Despite of these continuous research efforts in simple S/N systems and their derivatives, outlined above, the origin of certain effects remains mysterious. For example, an interesting paramagnetic re-entrant effect (sometime called Mota effect) caused by spontaneous currents in S/N bilayer was observed in Refs. [24–27]. The authors of Ref. [28] proposed an explanation in terms of a repulsive interaction with a negative small coupling constant λ_N i.e. assuming the normal metal may acquires a gap, Δ_N, which sign is opposite to that in a superconductor, Δ. However, the predicted paramagnetic response caused by spontaneous currents turned out to

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[†]This paper is dedicated to Professor Boris I. Kochelaev on the occasion of his 90th birthday.

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be too small because of the smallness of the superconducting order parameter in N $\Delta_N \sim \lambda_N$, and thus the origin of the Mota effect remains unclear [27]. Note, the paramagnetism and spontaneous currents may occur in S/F system [29–32] or S/F/N structures [33, 34]. However, there the origin of the paramagnetic effect should be quite different from that in S/N structures as in the former it is related to internal exchange fields existing in the ferromagnet F and to the triplet Cooper pairs induced in the film F by the PE [12–17]. In S/N/S Josephson junctions in a non-equilibrium [35] spontaneous currents arise when the Josephson current in S/N/S junctions changes sign [9, 18–21] but this situation then resembles the case of S/F/S junctions with a negative Josephson current [12–17]. Therefore, the situation of the S/N bilayer in equilibrium requires a separate study.

In this paper, we consider a simple S/N bilayer heterostructure with a superconducting coupling constant in the N layer equal to zero, i. e., $\lambda_N = 0$ and $\Delta_N = 0$. We calculate the total free energy $F_{S/N}$ of the system that consists of bulk terms F_S and F_N as well as the boundary term F_B. Below the critical temperature T_c, the energy F_S (F_N) decreases (increases), respectively. On the contrary to F_N, the boundary term F_B decreases the total free energy in such a way that the contribution of the terms $F_N + F_B$ is negative. The contribution $F_S + F_B$ remains negative as it is in the absence of the PE. The condensate current gives a positive contribution to both terms $F_N + F_B$ and $F_S + F_B$ making the current-carrying state unfavorable.

2. Theory

Frequently the analysis of the free energy (F) is performed using the Ginzburg-Landau free energy expansion, assuming the smallness of the order parameter Δ. This approach is not applicable to the considered heterostructure because the superconducting order parameter Δ_N in the N film is assumed to be zero. On the other hand, a part of electrons in N condense due to the PE and therefore the free energy F_N changes also in the superconducting state. Thus, in order to calculate the variation δF, we need to find first the quasiclassic matrix Green's functions $\hat{g}$ in the S and N regions using the boundary conditions and to express the free energy in terms of the functions $\hat{g}$. We consider a simple case of diffusive S/N structure when the function $\hat{g}$ obeys the Usadel equation [36]. In particular, the system under consideration is a bilayer which consists of S and N films with thicknesses $d_{S,N}$, respectively as shown in Fig.1. The current is assumed to flow along the interface in the y-direction. We integrate out the phase $\chi(y)$ by making the transformation $\hat{g}_n = \hat{S}^\dagger \cdot \hat{g} \cdot \hat{S}$, where $\hat{S} = \exp[(iQy/2)\hat{\tau}_3]$. This means that the phase χ and the functions $\hat{g}_n$ after the transformation depend only on the x coordinate and we drop the subscript "n" in what follows. We represent the matrix $\hat{g}$ in a standard form $\hat{g} = \hat{\tau}_3 \cos\theta + \hat{\tau}_1 \sin\theta$, which is typically used in studying S/N structures [37–42]

so that the normalization condition $\hat{g} \cdot \hat{g} = \hat{1}$ is automatically fulfilled. The function θ depends on x and obeys the Usadel equations in the S and N regions

$$-D_S\partial_{xx}^2\theta_S + 2\omega\sin\theta_S - 2\Delta\cos\theta_S + (D_S P_S^2/2)\sin(2\theta_S) = 0, \quad \text{S film} \tag{1}$$

$$-D_N\partial_{xx}^2\theta_N + 2\omega\sin\theta_N + (D_N P_N^2/2)\sin(2\theta_N) = 0, \quad \text{N film} \tag{2}$$

where $D_{S;N}$ are the diffusion coefficients in the S(N) films, ω is the Matsubara frequency, $\mathbf{P} = \nabla\chi - 2\pi\mathbf{A}/\Phi_0$ is the gauge-invariant condensate momentum and $\Phi_0 = hc/2e$ is the magnetic flux quantum. The Usadel equations are complemented by the standard Kurpiyanov-Lukichev boundary conditions for $\theta_{S,N}$ [43] at the interface

$$\partial_x\theta_{S(N)} = -\kappa_{B,S(N)}\sin(\theta_S - \theta_N)|_{x=0} \tag{3}$$

Figure 1. (Color online.) A schematic representation of the S/N bilayer structure.

where $\kappa_{B,S(N)} = 2/R_B\sigma_{S(N)}$, R_B is the S/N interface resistance per unit area and $\sigma_{S,N}$ are the conductivities in the S and N films in the normal state. The order parameter Δ, which is non-zero in the S film, is determined by the self-consistency equation

$$\Delta = \lambda(2\pi T)\sum_{\omega \geqslant 0}\sin\theta_S(\omega) \tag{4}$$

Note, Eq.(4) and Eqs.(1-2) are obtained by the variation of the total free energy F_S and F_N with respect to Δ and θ

$$F_S = 2\nu_S\int_{-d_S}^{0}dx\Bigg\{\frac{\Delta^2}{2\lambda} + 2\pi T\sum_{\omega \geqslant 0}\Bigg[\frac{D_S}{4}(\partial_x\theta_S)^2 + \omega(1 - \cos\theta_S) - \Delta\sin\theta$$
$$+ \frac{D_S P^2}{8}\left(1 - \cos(2\theta_S)\right)\Bigg]\Bigg\}, \tag{5}$$

$$F_N = 2\nu_N\int_{0}^{d_N}dx\Bigg\{2\pi T\sum_{\omega \geqslant 0}\Bigg[\frac{D_N}{4}(\partial_x\theta_N)^2 + \omega(1 - \cos\theta_N) + \frac{D_N P^2}{8}\left(1 - \cos(2\theta_N)\right)\Bigg]\Bigg\}, \tag{6}$$

where ν, P, D are the density of states, momentum, and diffusion coefficient in either S or N film, respectively. We set Δ_N equal to zero since we assume that $\lambda_N = 0$. The energy F is counted from its value in the normal state, *i.e.* $\theta = 0$. This expression for F_N can be also derived from a more general expression for the free energy of a superconductor in the presence of an exchange field [44, 45]. We also note by passing that taking the variation of the sum of the F and the magnetic energy $(\nabla \times \mathbf{A})^2/8\pi$ one obtains the London equation $\nabla^2\mathbf{A} = (4\pi/c)\mathbf{j}$, where $\mathbf{j} = -(c/4\pi)\Lambda_L^{-2}\mathbf{P}$. Here, $\Lambda_L^{-2} = [2\sigma/(c^2\hbar)](2\pi T)\sum_{\omega \geqslant 0}\sin^2\theta(\omega))$ is the inverse squared London penetration depth. In order to take into account the boundary conditions (3), we need to add the boundary term F_B [47, 48] to $F_S + F_N$ so that the total functional F is given by

$$F = F_S + F_N + F_B \tag{7}$$

In the following we solve Eqs.(1-2) for the functions $\theta_{S,N}$ together with the self-consistency equation (4) and find a minimum of the free energy F as a function of the condensate velocity $\mathbf{V} = \mathbf{P}/m$. In a general case, this can be done only numerically. Here we restrict the analysis with the simplest case of a weak proximity effect when the Usadel equation for θ_N2 can be linearized and the function θ_S is weakly perturbed by the PE. The latter assumption is valid if

the condition $\delta\theta_S \lesssim \xi_S/(R_B\sigma_S) \ll 1$ is fulfilled, where $\xi_S \cong \sqrt{D_S/2\Delta}$ is a coherence length in S. Yet we do take into account a suppression of the order parameter Δ by the condensate flow. In the case of small suppression of Δ, we find $\delta\Delta \cong -(D_S P_S^2/2\Delta)\sum_{\omega\geqslant 0}(\omega^2/\zeta_\omega^4)/\sum_{\omega\geqslant 0}\zeta_\omega^{-3}$, where $\zeta_\omega = \sqrt{\omega^2 + \Delta_0^2}$. At low temperatures ($T \ll \Delta$) the gap variation is $\delta\Delta \cong -D_S P_S^2/2$. Note that a strong suppression of Δ by the condensate flow was studied in Refs. [44, 46]. In the absence of the PE and the condensate flow, one has $\sin\theta_{S0} \equiv f_S = \Delta_0/\zeta_\omega$ and $\cos\theta_{S0} = \omega/\zeta_\omega$ with $\zeta_\omega = \sqrt{\omega^2 + \Delta_0^2}$. The direct calculation of F_{S0} gives a well known result $F_{S0} = -\nu_S\Delta^2 d_S/2$ [2]. The correction δF_{S0} caused by the condensate flow is $\delta F_{S0} = \nu_S D_S P_S^2(2\pi T)\Delta\sum_{\omega\geqslant 0}\zeta_\omega^{-2}$. Thus, the energy F_S of the S film with a spontaneous current is

$$F_S = -\nu_S d_S \frac{\Delta^2}{2}\left[1 - D_S P_S^2(2\pi T)\sum_{\omega\geqslant 0}\zeta_\omega^{-2}\right] \tag{8}$$

and as expected the condensate flow reduces the condensation energy.

Next we evaluate the contribution to the free energy of the N film, F_N. Linearized Eq.(2) has the form

$$-\partial_{xx}^2\theta_N + \kappa_q^2\theta_N = 0, \tag{9}$$

with a solution

$$\theta_N(x) = \frac{\kappa_B}{\kappa_q}f_S\frac{\cosh(\kappa_q(x - d_N))}{\sinh(\kappa_q d_N)}, \tag{10}$$

where $\kappa_q = \sqrt{2\tilde{\omega} + q^2}/\xi_N$, $\xi_N = \sqrt{D_N/2\Delta}$, $q = Q\xi_N$ and $f_S = \Delta/\zeta_\omega$. The solution describes correctly the condensate Green's function in N provided the condition $R_B > \rho_N\xi_N$ is fulfilled.

In the limit of a weak PE the energy $F_N + F_B$ can be written in the form

$$F_N + F_B = \nu_N(2\pi T)D_N\sum_{\omega\geqslant 0}\left\{\int_0^{d_N}dx\frac{1}{2}[(\partial_x\theta_N)^2 + \kappa_q^2\theta_N^2] + \kappa_B[1 - \cos(\theta_S - \theta_N)]\right\}, \tag{11}$$

where the last term is the boundary free energy [47,48]. Substituting the solution (10) into (11), we come to the formula for $F_N + F_B$ and θ_S one can easily calculate the

$$F_N + F_B = \nu_N(2\pi T)(D_N\kappa_B^2)\sum\left[\frac{f_S^2}{2\kappa_q\tanh(\kappa_q d_N)} + \frac{1}{\kappa_B}(1 - \cos\theta_S) - \frac{f_S^2}{\kappa_q\tanh(\kappa_q d_N)}\right]. \tag{12}$$

The first term in the figure brackets is the contribution of the bulk N region whereas the last term stems from the boundary contribution to the free energy. The second term is a reduction of the free energy due to the PE. One can see that the first term gives a positive contribution to the F and decreases with increasing the condensate velocity $V_S \sim q$. However the boundary term (the last one) is twice larger than the first one and therefore the total contribution of the terms due to condensate current, Eq.(8,12), is positive. This means that the condensate current reduces the free energy.

3. Conclusions

To conclude, we analyzed the free energy for S/N bilayer in the presence of the condensate current. We have shown that the bulk of the N film gives a positive contribution $F_N(q)$ to the free energy which decreases with increasing condensate velocity $V \sim q$. However the contribution of boundary term F_B to F is twice larger in magnitude than $F_N(q)$ and is also negative as the contribution F_S of the superconductor S. Therefore the total free energy F increases when condensate moves; this makes the current-carrying state unfavorable.

Acknowledgments

4. Supplementary Information

4.1. General Case

Here we present the evaluation of the free energy F_N in the N film. We integrate once Eq.(7) in the main text

$$-\frac{D_N}{2}(\partial_x\theta_N)^2 + 2\omega(1-\cos\theta_N) + \frac{D_N P_N^2}{4}[1-\cos(2\theta_N)] = 0 \tag{S1}$$

and assumed that $d_N \gg \xi_{N,\Delta} \equiv \sqrt{D_N/2\Delta}$ so that $\theta_N = 0$ at $x = d_N$. Taking into account Eq.(S1), the energy F_N can be written as follows

$$F_N = \nu_N(2\pi T)D_N \sum_{\omega\geqslant 0}\int_0^\infty dx(\partial_x\theta_N)^2 \equiv \nu_N(2\pi T)D_N\sum_{\omega\geqslant 0}\tilde{F}_{N,\omega}, \tag{S2}$$

where $\tilde{\omega} = \omega/\Delta$ and the function $\tilde{F}_{N,\omega}$ is defined as

$$\begin{aligned}
\tilde{F}_{N,\omega} &= \int_0^\infty dx(\partial_x\theta_N)^2 = -\int_0^{\theta_{N0}}d\theta_N(\partial_x\theta_N) = -\int_0^{\theta_{N0}}d\bar{\theta}_N\sin\bar{\theta}_N\sqrt{\kappa_\omega^2 + P^2\cos^2\bar{\theta}_N} \\
&= \frac{P}{2}\left[\sqrt{1+a_\omega^2} - t_0\sqrt{t_0^2+a_\omega^2} + a_\omega^2\ln\frac{1+\sqrt{1+a_\omega^2}}{t_0+\sqrt{t_0^2+a_\omega^2}}\right],
\end{aligned} \tag{S3}$$

where $t_0 \equiv \cos\bar{\theta}_N \equiv \cos(\theta_N/2)|_{x=0}$ and $a_\omega^2 = \kappa_\omega^2/P^2$. The parameter t_0 is found from the boundary condition

$$\sqrt{1-t_0^2}\sqrt{1+(q/\kappa_\omega\xi_N)^2 t_0} = \frac{\kappa_{BN}}{2\kappa_\omega}\left[\Delta(2t^2-1) - 2\omega t_0\sqrt{1-t_0^2}\right], \tag{S4}$$

4.2. Weak PE

Consider now a weak PE when the function θ_N is small. In this case one can obtain a formula for F_N for arbitrary thickness d_N. At $\theta_N \ll 1$, Eq.(2) in the main text can be linearised

$$-\partial_{xx}^2\theta_N + \kappa_N^2\theta_N = 0, \tag{S5}$$

where $\kappa_N^2 = \kappa_{N\omega}^2 + P^2$, $\kappa_{N\omega}^2 = 2\omega/D_N$. The boundary conditions, Eq.(3), have the form

$$\begin{aligned}
\partial_x\theta_N &= -\kappa_{B,N}[\sin\theta_S - \theta_N\cos\theta_S]|_{x=0}, \\
\partial_x\theta_N &= 0|_{x=d_N},
\end{aligned} \tag{S6}$$

where $\kappa_{BN} = 2/R_B\sigma_N$. The solution for Eq.(S5) obeying the condition (S6) is

$$\theta_N(x) = \frac{\kappa_{BN}}{\kappa_N}\frac{\cosh(\kappa_N(x-d_N))}{\cosh\vartheta_N D_N}\sin\theta_S, \tag{S7}$$

where $D_N = \tanh \alpha_N + (\kappa_{BN}/\kappa_N)\tilde{\omega}/\sqrt{\tilde{\omega}^2 + 1}$, $\alpha_N = \kappa_N d_N$, $\sin\theta_S = 1/\sqrt{\tilde{\omega}^2 + 1}$. The energy of the N film, F_N, is

$$
\begin{aligned}
F_N &= 2\nu_N(2\pi T)\sum_{\omega \geqslant 0}\int_0^{d_N} dx \left[\frac{D_N}{4}(\partial_x\theta_N)^2 + \frac{1}{4}(2\omega + D_N P^2)\theta_N^2\right] \\
&= 2\nu_N\frac{D_N\kappa_{BN}^2}{4}\xi_{N,\Delta}(2\pi T)\sum_{\omega \geqslant 0}\frac{\tanh\alpha_N}{D_N^2}\frac{1}{\sqrt{\tilde{\omega} + q^2}}\frac{1}{\tilde{\omega}^2 + 1}.
\end{aligned}
\tag{S8}
$$

In the limit of a thick N film $(d_N \gg \xi_{N,\Delta})$ Eq.(S6) acquires the form

$$
F_N = 2\nu_N\frac{D_N\kappa_{BN}^2}{4}\xi_{N,\Delta}(2\pi T)\sum_{\omega \geqslant 0}\frac{1}{\sqrt{\tilde{\omega} + q^2}}\frac{1}{\tilde{\omega}^2 + 1}.
\tag{S9}
$$

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