Recovering the superconducting state via spin accumulation above the pair-breaking magnetic field of superconductor/ferromagnet multilayers

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We study theoretically the simultaneous influence of the spin accumulation potential $eV_\uparrow$-$eV_\downarrow$ and the Zeeman exchange field on singlet superconductivity. It is shown that the pair-breaking effect of the Zeeman field can be fully compensated by creation of the appropriate spin accumulation potential in the superconductor. Moreover, superconductivity can be recovered for exchange fields well exceeding the Pauli limiting field. It is proposed that the effect can be experimentally realized on the basis of a voltage-biased junction consisting of a thin superconducting film sandwiched between two half metals.

One of the mechanisms destroying singlet superconductivity is the Zeeman interaction of electron spins with magnetic field. The behavior of a magnetic superconductor with an exchange field h was studied long ago.[1–4] It was found that the homogeneous superconducting state becomes energetically unfavorable above the paramagnetic (Pauli) limit $h = \Delta/\sqrt{2}$. As it was predicted,[1,2] in a narrow region of exchange fields exceeding this value, the superconductivity can appear as an inhomogeneous state with a spatially modulated Cooper-pair wave function (LOFF state). Thus, the exchange field is destructive to singlet superconductivity. An exception, proposed in the literature, is a special type of a clean superconducting multilayered system, where the paramagnetic limit can be enhanced and a nonuniform superconducting state can be induced under an in-plane magnetic field.[5]

Superconductor/ferromagnet (S/F) hybrid structures also can behave analogous to magnetic superconductors. In particular, it was shown[6] that a thin S/F bilayer with thicknesses obeying the conditions $d_F \ll \xi_F$ and $d_S \ll \xi_S$ is equivalent to a magnetic superconductor with an effective exchange field $h_{\mathrm{eff}} = hN_F d_F/(N_F d_F + N_S d_S)$ and an effective superconducting order parameter $\Delta_{\mathrm{eff}} = \Delta N_S d_S/(N_F d_F + N_S d_S)$. Here $\xi_F = \sqrt{D_F/h}$ and $\xi_S = \sqrt{D_S/|\Delta|}$ are the magnetic and superconducting coherence lengths, $|\Delta|$ is the superconducting order parameter in the bulk material, $N_{F,S}$ denote the densities of states at the Fermi level for the ferromagnet and superconductor, and $D_{F,S}$ are the corresponding diffusion constants. Another way to "apply" an exchange field on a thin superconducting film is to contact it to a ferromagnetic insulator.[7–11] It was observed experimentally[10] and justified theoretically[11] that the effective exchange field induced in the film scales with d_S^{-1}.

The simultaneous applying of the exchange field and creation of spin-dependent quasiparticle distribution in S/F heterostructures can lead to qualitatively unique phenomena. In particular, it was shown recently[12,13] that the creation of spin-dependent quasiparticle distribution in the interlayer of a S/F/S junction (or a S/N/S junction with magnetic S/N interfaces) leads to the appearance of an additional contribution to the Josephson current through the junction, which under certain conditions can enhance it considerably. In the present Rapid Communication we demonstrate 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. This effect takes place even if the exchange field exceeds the paramagnetic limit considerably, that is, under the condition that the superconductivity of the equilibrium film is fully suppressed.

As an example of a system where the uniform exchange field and the spin-dependent quasiparticle distribution can be realized simultaneously, we propose here a voltage-biased half metal/superconductor/half metal (HM/S/HM) heterostructure. A thin film ($d_S \ll \xi_S$) made of a dirty s-wave superconducting material is sandwiched between two half-metallic layers with opposite directions of magnetization. Half-metallic behavior has been reported in CrO_2 (Refs. 14 and 15) and in certain manganites.[16] The in-plane effective uniform exchange field h_{eff} in the film is supposed to be created by spin-active interfaces with half metals. The spin-dependent quasiparticle distribution in the film can be generated by applying a voltage bias between the two half metals. In this case for a spin-up subband the main voltage drop occurs at one of the HM/S interfaces, while for the spin-down subband it occurs at the other interface. As a result, the distribution functions for spin-up and spin-down electrons in the superconducting film are to be close to the equilibrium form with different electrochemical potentials.

As we consider a nonequilibrium system, we make use of a Keldysh framework of the quasiclassical theory, where the fundamental quantity is the momentum average of the quasiclassical Green's function $\check{g}(x,\varepsilon) = \langle \check{g}(\boldsymbol{p}_f,x,\varepsilon)\rangle_{\boldsymbol{p}_f}$. Here x is the coordinate normal to the S/HM interfaces and ε is a quasiparticle energy. The Green's function is an 8×8 matrix form in the product space of Keldysh, particle-hole, and spin variables. In the superconducting film it obeys the Usadel equation

$$\frac{D}{\pi}\partial_x(\check{g}\partial_x\check{g}) + [\varepsilon\tau_3\sigma_0\rho_0 - \Delta\tau_1 i\sigma_2\rho_0,\check{g}] = 0, \quad (1)$$

where τ_i, σ_i, and ρ_i are Pauli matrices in particle-hole, spin, and Keldysh spaces, respectively. τ_0, σ_0, and ρ_0 stand for the corresponding identity matrices. Equation (1) should be supplied with the normalization condition $\check{g}^2 = -\pi^2\tau_0\sigma_0\rho_0$. It is convenient to express the Keldysh part of the full Green's function via the retarded and advanced components and the distribution function $\check{g}^K = \check{g}^R\check{\varphi} - \check{\varphi}\check{g}^A$. The distribution

function is diagonal in the particle-hole space $\check{\varphi} = \hat{\varphi}(\tau_0 + \tau_3)/2 + \sigma_2\hat{\tilde{\varphi}}\sigma_2(\tau_0 - \tau_3)/2$. The hole component $\hat{\tilde{\varphi}}$ of the distribution function is connected to $\hat{\varphi}$ by a general symmetry relation[17] $\hat{\tilde{\varphi}}(\varepsilon) = -\sigma_2\hat{\varphi}(-\varepsilon)\sigma_2$.

We consider the case when there is the only magnetization axis in the system (the magnetization directions of the HM's are antiparallel). Then there are no equal-spin triplet superconducting correlations in the system and all the matrices in spin space can be represented as sums of two spin subband contributions ($\sigma = \uparrow, \downarrow$). For later use we define here the anomalous Green's function $f^{R,A} = [f_\uparrow^{R,A}(\sigma_0 + \sigma_3)/2 + f_\downarrow^{R,A}(\sigma_0 - \sigma_3)/2]i\sigma_2$ and the distribution function $\hat{\varphi} = \varphi_\uparrow(\sigma_0 + \sigma_3)/2 + \varphi_\downarrow(\sigma_0 - \sigma_3)/2$.

The Usadel equation is subject to appropriate boundary conditions at S/HM interfaces, which for the diffusive limit can be written in the form[11]

$$\check{g}\partial_x\check{g} = -\alpha\frac{G_T^{l,r}}{2\sigma_S}[\check{g}, \check{g}_{HM}^{l,r}] - \alpha\frac{G_{MR}^{l,r}}{2\sigma_S}[\check{g}, \{\check{m}^{l,r}, \check{g}_{HM}^{l,r}\}]$$
$$+ \alpha\frac{G_\phi^{l,r}\pi}{2\sigma_S}[\check{m}^{l,r}, \check{g}], \quad (2)$$

where $\check{g}$ is the Green's function value at the superconducting side of the appropriate S/HM interface (at $x = \mp d_S/2$), $\alpha = +1(-1)$ at the left (right) S/HM interface and σ_S stands for the conductivity of the film. $\check{m}^{l,r} = \boldsymbol{m}^{l,r}\boldsymbol{\sigma}\rho_0(1 + \tau_3)/2 + \boldsymbol{m}^{l,r}\boldsymbol{\sigma}^*\rho_0(1 - \tau_3)/2$, where $\boldsymbol{m}^{l,r}$ is the unit vector aligned with the magnetization direction of the left or right half metal. We assume that the half metals have opposite magnetization directions, that is, $\boldsymbol{m}^r = -\boldsymbol{m}^l$. The second term accounts for the different conductances of different spin directions and $G_{MR} \sim G_{T,\uparrow} - G_{T,\downarrow}$. The third term $\sim G_\phi$ gives rise to spin-dependent phase shifts of quasiparticles being reflected at the interface. Microscopically,[11] $G_\phi^{l,r} = 2(G_q/S)\sum_n(T_n^{l,r} - 1)d\phi_n^{l,r}$, where S is the junction area and $G_q = e^2/h$ is the quantum conductance. Summation over n means summation over transmission channels. $T_n^{l,r}$ is the transmission probability for the nth channel and $d\phi_n^{l(r)}$ is the phase difference between wave functions of spin-up and spin-down electrons, acquired upon reflection from the S/HM interface (spin-dependent phase shift). Boundary conditions (2) are only valid for small (with respect to unity) values of transparency T_n and spin-dependent phase shift $d\phi_n$ in one transmission channel. The value of $d\phi_n^{l(r)}$ can be roughly estimated, for example, by modeling the barrier at the S/HM interface by $U_\sigma(x) = U_\sigma\delta(x)$. Then in the tunnel limit $T_n \ll 1$ one obtains $d\phi_n \approx v_F(U_\downarrow - U_\uparrow)/U_\uparrow U_\downarrow$. In general, the boundary conditions can contain another term proportional to G_χ accounting for spin-dependent phase shifts of quasiparticles upon transmission.[11] However, we are mostly interested in the tunnel limit, where this term can be disregarded with respect to G_ϕ.

$\check{g}_{HM}^{l,r}$ stands for the Green's functions at the half-metallic side of the interface. Since in half metals a Fermi surface only exists for one of the spin orientations, the standard quasiclassical description is inapplicable. However, half metals still allow for a straightforward quasiclassical treatment in the separate-band picture: Quasiparticle trajectories simply exist only for one of the spin orientations.[18] If one chooses the

quantization axis along the left HM magnetization, $\check{g}_{HM}^{l,r}$ takes the form

$$\check{g}_{HM}^{R,A} = -i\pi\kappa\tau_3(\sigma_0 + \alpha\sigma_3)/2. \quad (3)$$

Here labels (l,r) are omitted for brevity and $\kappa = +1(-1)$ for the retarded (advanced) Green's functions. The distribution functions in the half metals are assumed to have the equilibrium form shifted by the applied voltages $V_{l,r}$. We suppose that $V_r = -V_l = V$. In this case

$$\check{g}_{HM}^K = -2i\pi\left[\tanh\frac{\varepsilon + \alpha eV}{2T}\frac{(\tau_0 + \tau_3)}{2}\right.$$
$$\left. - \tanh\frac{\varepsilon - \alpha eV}{2T}\frac{(\tau_0 - \tau_3)}{2}\right]\frac{\sigma_0 + \alpha\sigma_3}{2}. \quad (4)$$

The self-consistent order parameter in the film is expressed via the Keldysh part of the anomalous Green's function. We assume that the pairing constant is nonzero only for the singlet pairing channel. Then the corresponding self-consistency equation takes the form

$$\Delta = \frac{\lambda}{2}\int_{-\omega_D}^{\omega_D}\frac{d\varepsilon}{4\pi i}\sum_\sigma\left[f_\sigma^R(\varepsilon)\tilde{\varphi}_\sigma(\varepsilon) + f_\sigma^A(\varepsilon)\varphi_\sigma(\varepsilon)\right], \quad (5)$$

where λ is the dimensionless coupling constant.

Equations (1)–(5) constitute a full system of equations for solving the problem. For the case of a thin superconducting film $d_S \ll \xi_S$ they can be solved analytically just analogously to the case of a thin S/F bilayer,[6] mentioned above. Averaging the retarded and advanced parts of Eq. (1) over the thickness of the film and taking into account the boundary conditions, one can reduce the Usadel equation to an equation describing a uniform magnetic superconductor with an effective exchange energy h_{eff} and a decoherence factor Γ. This equation can be easily solved. The corresponding anomalous components of retarded and advanced Green's function, entering Eq. (5), take the form

$$f_\sigma^{R,A} = \frac{\pi\Delta}{\sqrt{\Delta^2 - [\varepsilon + \sigma h_{eff} + i\kappa\Gamma]^2}}, \quad (6)$$

where the decoherence factor $\Gamma = (G_T^r + 2G_{MR}^r + G_T^l + 2G_{MR}^l)D/4\sigma_S d_S$ physically describes the leakage of superconducting correlations from the film into the HM regions. This term is quite standard (including S/N systems) and is responsible for the destruction of superconductivity in thin films by the proximity effect.

The effective exchange energy $h_{eff} = (G_\phi^r - G_\phi^l)D/2\sigma_S d_S$ is generated by S/HM interfaces. It is inversely proportional to the film width d_S. As it was mentioned above, the boundary conditions (2) and, correspondingly, Eq. (6) are valid for $d\phi_n \ll 1$. Beyond this limit the effect of magnetic boundaries cannot be reduced to the effective exchange in the film,[11] but there appear additional terms that are to some extent analogous to the magnetic impurities. We assume that the condition $d\phi_n \ll 1$ is fulfilled. However, this does not mean that the resulting exchange fields are small. In order to observe the recovering of superconductivity, suppressed by the exchange field, one needs $h_{eff} \gtrsim \Delta$. For the film with $d_S \lesssim \xi_S$ this condition is accomplished if (i) the left and right interfaces are not identical, that is, $G_\phi^r \neq G_\phi^l$ and (ii) the dimensionless parameter $h_{eff}/\Delta \sim G_\phi\xi_S^2/\sigma_S d_S \gtrsim 1$. This parameter can be

estimated as $-(2N\xi_S^2 G_q/S\sigma_S d_S)d\phi \sim -(\xi_S^2/ld_S)d\phi$, where N is the number of transmission channels and l is the mean free path. From this estimate it is seen that for dirty superconductors G_ϕ can generate large h_{eff} even for $d\phi \ll 1$.

Now let us turn to the discussion of the distribution function. We neglect energy relaxation in the film, that is, we assume that the time $\tau_{\mathrm{esc}} = \Gamma^{-1}$, which an electron spends in the film is much less than the energy relaxation time τ_ε. Spin relaxation processes are also not taken into account. We discuss their influence below. Then it can be obtained from Eqs. (1)–(4) that the distribution function in the film takes the form

$$\varphi_\sigma = \tanh\frac{\varepsilon + \sigma eV}{2T}. \tag{7}$$

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 leads are HM: The electrons from the spin-up (spin-down) subband can flow only to and from the left (right) lead. This one-step form is very essential for the existence of the effect. In principle, the superconductivity recovering can be also observed if one takes strong ferromagnets instead of half metals, but in this case the nonequilibrium distribution function inside the film is represented by a sum of the distribution functions coming from the left and right leads, weighted by factors depending on the interface transparencies (this is a double-step structure). This would lead to only partial recovery of superconductivity, or even to the absence of the effect. So, in order to provide the appropriate distribution function in the film, the resistances of the ferromagnets and S/F interfaces should obey quite strict conditions.

Substituting Eqs. (6) and (7) [$\tilde\varphi$ is obtained making use of the symmetry relation $\tilde\varphi_{\uparrow,\downarrow}(\varepsilon) = -\varphi_{\downarrow,\uparrow}(-\varepsilon)$] into Eq. (5), we come to the following self-consistency equation:

$$\frac{1}{\lambda} = \int_{-\omega_D}^{\omega_D} \frac{d\varepsilon}{4} \left\{ \mathrm{Re}\left[\frac{\mathrm{sgn}(\varepsilon + h_{\mathrm{eff}})}{\sqrt{(\varepsilon + i\Gamma + h_{\mathrm{eff}})^2 - \Delta^2}} \right] \tanh\frac{\varepsilon + eV}{2T} \right.$$
$$\left. + \mathrm{Re}\left[\frac{\mathrm{sgn}(\varepsilon - h_{\mathrm{eff}})}{\sqrt{(\varepsilon + i\Gamma - h_{\mathrm{eff}})^2 - \Delta^2}} \right] \tanh\frac{\varepsilon - eV}{2T} \right\}. \tag{8}$$

From Eq. (8) it is obvious how superconductivity in the film is recovered under the simultaneous influence of the exchange field and the spin-dependent quasiparticle distribution. At $h_{\mathrm{eff}} = eV$ for each of the subbands we have practically the same situation as for the equilibrium nonmagnetic film corresponding to $eV = h_{\mathrm{eff}} = 0$. It is worth noting here that in the framework of the simplified weak-coupling model with a constant pairing potential the maximal value of the Zeeman field, which does not destroy the superconductivity, is equal to the cutoff energy ω_D. However, for high enough exchange fields this simplified model is inapplicable. For concreteness let us discuss the phonon-mediated superconductivity. In this case our consideration fails for exchange fields of the order of the Debye energy and the correct calculation should be carried out in the framework of the particular phonon model.

The resulting Δ as a function of applied voltage eV is plotted in Fig. 1(a) for different values of h_{eff}. It is clearly seen that the effect of superconductivity recovery only takes place if the exchange field h_{eff} and the spin accumulation potential eV are very close to each other: Their difference should be less than Δ_0. Here Δ_0 denotes the value of the
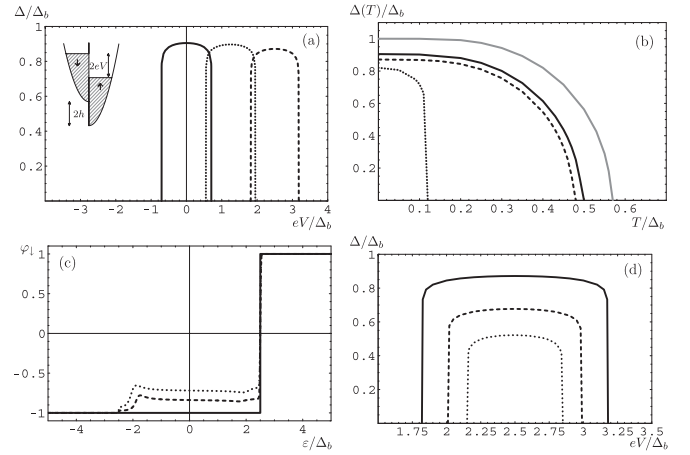

FIG. 1. (a) The dependence of $\Delta(T = 0)$ on eV. Solid line: $h_{\mathrm{eff}} = 0$; dotted line: $h_{\mathrm{eff}} = 1.25$; dashed line: $h_{\mathrm{eff}} = 2.5$. All the quantities are measured in units of the bulk order parameter Δ_b taken at $1/\tau_{\mathrm{sf}} = 0$ and $T = 0$. Inset: Population of majority and minority subbands for the given quasiparticle distribution. (b) The dependence of Δ on temperature. Gray solid line: $\Delta_b(T)$; black solid line: $\Delta_0(T) \equiv \Delta(T, h_{\mathrm{eff}} = eV = 0)$; dashed line: $\Delta(T, h_{\mathrm{eff}} = eV = 2.5)$; dotted line: $\Delta(T, h_{\mathrm{eff}} = 2.5, eV = 3.1)$. For (a) and (b) $1/\tau_{\mathrm{sf}} = 0$. (c) The distribution function $\varphi_\downarrow(\varepsilon)$ for different spin relaxation rates: $1/\tau_{\mathrm{sf}} = 0$ (solid line), 0.01 (dashed), and 0.02 (dotted). $h_{\mathrm{eff}} = eV = 2.5$. $\varphi_\uparrow(\varepsilon) = -\varphi_\downarrow(-\varepsilon)$. (d) The dependence of Δ on eV for $h_{\mathrm{eff}} = 2.5$ and different spin relaxation rates [the same as (c)]. For all the panels, $\Gamma = 0.1$.

superconducting order parameter in the film at $h_{\mathrm{eff}} = 0$. The physical reason for superconductivity recovery can be easily caught even without solving the self-consistency equation, already on the level of consideration of the Cooper's problem of one electron pair. It can be shown that in the presence of the exchange field, the lowest-energy level ε_0 for a pair of electrons with $\mathbf{p}_1 = -\mathbf{p}_2$ and opposite spins gets lower upon increasing the spin accumulation potential. Finally, ε_0 becomes exactly equal to its value for the zero exchange field at $eV = h_{\mathrm{eff}}$. It is worth noting here that in the absence of the exchange field the spin accumulation potential also destroys superconductivity at $|eV| \sim \Delta_0$. This effect has been studied in the literature theoretically as well as experimentally.[19–21]

At $eV = 0$ and $T,\Gamma \to 0$, the self-consistency equation (8) has a nonzero spatially uniform solution for $h_{\mathrm{eff}} < \Delta_0$. However, it is well known[1] that the uniform solution becomes metastable even earlier, at $h_{\mathrm{eff}} = \Delta_0/\sqrt{2}$ (Pauli limiting field) because of the fact that the paramagnetic state is more energetically favorable for higher exchange fields. In the considered case the paramagnetic state cannot be realized because the distribution function is created and supported by the external conditions in such a way that the populations of majority and minority subbands in the film remain equal. This is illustrated in the inset of Fig. 1(a). The uniform superconducting state is obviously more favorable than the normal one due to the condensation energy. However, there can exist another possibility: In principle, under a spin-dependent quasiparticle distribution an inhomogeneous superconducting state (analogous to the LOFF state) can occur and be more favorable than the homogeneous one at some ranges of parameters. This issue is a prospect for future work.

For the resonance value of $eV_{res} = h_{eff}$ the dependence of the superconducting order parameter on temperature is very similar to the original BCS one, giving practically the same ratio $2\Delta(T = 0)/T_c$, as illustated in Fig. 1(b). As it is also represented in Fig. 1(b), when eV deviates from eV_{res} the temperature suppresses the order parameter more sharply.

Now we turn to the discussion of the spin relaxation influence on the effect. We assume spin-flip scattering from magnetic impurities to be the dominant spin relaxation process inside the superconducting film at low temperatures. It can be taken into account by adding the corresponding self-energy term $[-(1/2\pi\tau_{sf})\check{\sigma}\check{g}\check{\sigma}, \check{g}]$ to the left-hand side of Eq. (1). Here $\check{\sigma} = [\boldsymbol{\sigma}(1 + \tau_3)/2 + \boldsymbol{\sigma}^*(1 - \tau_3)/2]\rho_0$. As it is well known, the influence of the spin-flip scattering is twofold. First, it "works" as a depairing factor by destroying the coherence peaks and reducing the critical temperature of the superconductor.[22] Second, the spin-flip scattering influences directly the distribution function by reducing the difference $\varphi_\uparrow - \varphi_\downarrow$. The reduction can be roughly estimated as $\varphi_\uparrow^{sf} - \varphi_\downarrow^{sf} = (\varphi_\uparrow - \varphi_\downarrow)/(1 + \tau_{esc}/\tau_{sf})$. Here $\varphi_\uparrow^{sf} - \varphi_\downarrow^{sf}$ is the difference in the presence of spin relaxation processes, while $\varphi_\uparrow - \varphi_\downarrow$ is defined by Eq. (7) and τ_{sf} is the characteristic spin relaxation time. The results

of the exact calculation of the distribution function slightly deviate from this rough estimate, especially in the region of the coherence peaks. They are represented in Fig. 1(c). Obviously, the "damage" to the distribution function due to spin relaxation also suppresses the effect of Δ recovery. This suppression can be roughly viewed as the effective reduction of the coupling constant $\lambda \to \lambda_{eff} = \lambda(1 + \tau_{esc}/\tau_{sf})^{-1}$. The resulting influence of the spin-flip scattering on the order parameter is demonstrated in Fig. 1(d).

In summary, we have theoretically shown that the creation of spin-dependent quasiparticle distribution in a superconductor can fully compensate the pair-breaking effect of the Zeeman field. Thus, superconductivity can be recovered for exchange fields well exceeding the Pauli limiting field if the spin accumulation potential $eV_\downarrow - eV_\uparrow \approx 2h_{eff}$ is generated in the superconductor. It is proposed that this effect can be experimentally realized on the basis of a voltage-biased junction consisting of a thin superconducting film sandwiched between two half metals.

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