Anomalous proximity effect in spin-valve superconductor/ferromagnetic metal/ferromagnetic metal structures

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We investigate superconductor (S) and ferromagnetic metal (F) SFF structures with noncollinear magnetizations of F films with arbitrary FF interface transparency. We show the existence of phase slips at both the SF and FF interfaces, which manifest themselves in the anomalous dependence of the spin-triplet correlations on the misorientation angle between the magnetization vectors in the F layers. We discuss how these effects can be observed in experiments with Josephson π junctions.

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I. INTRODUCTION

There is considerable interest in structures composed of superconducting (S) and ferromagnetic (F) layers.¹⁻³ The possibility of π states in SFS Josephson junctions due to the oscillatory nature of superconducting order parameters induced into a ferromagnet was predicted theoretically⁴⁻⁶ and has been convincingly demonstrated by experiments.⁷⁻²⁵ It was also shown recently that both classical and quantum circuits^{26–30} can be realized using SFS sandwich technology.⁷ A number of new phenomena were predicted in junctions with more than one magnetically ordered layer. Particularly interesting are equal-spin triplet superconducting correlations, which can penetrate a ferromagnet on a long-range scale.³¹⁻³⁹ These states are generated if spin rotation symmetry is broken and are therefore expected to become most important when the angle α between magnetization vectors of ferromagnetic layers is close to $\pi/2$. Long-range triplets were recently realized experimentally in Josephson junctions in a number of geometries and material combinations^{40–44} and in SFF spin valves.45-47

Golubov *et al.*⁴⁸ also predicted that in Josephson junctions with several ferromagnetic layers it is possible to realize π states even when the F layers are so thin that order parameter oscillations cannot develop, but that phase slips occur at SF interfaces with finite transparency. This effect was predicted for SFIFS junctions where two SF bilayers are decoupled by an insulating barrier, I. In this case, phase shifts $\delta\phi$ occur at each of the SF interfaces and saturate at $\delta\phi = \pi/2$ with the increase of the exchange field. As a result, the total phase shift across the junction equals π .

Recently, structures where two F layers are coupled to a superconductor (FSF or SFF) attracted much attention, because they may serve as superconducting spin valves where transition temperature is controlled by the angle α between magnetization directions of the F layers. SFF structures with fully transparent interfaces were studied theoretically by Fominov *et al.*,⁴⁷ who showed that the critical temperature T_c in such trilayers can be a nonmonotonic function of the angle α .

In this paper we address the important issue of the influence of interface transparency on singlet and triplet correlations in SFF structures and show that interface phase slips can lead to a number of peculiar new phenomena. First, the magnitudes of singlet and long-range triplet components generated in SFF structures with a varying angle α between the F-layer magnetizations have an anomalous dependence on α . Specifically, contrary to previous knowledge based on analysis of symmetric FSF or SFFS structures, the triplet component in SFF structures reaches maximum not in the vicinity of $\alpha = \pi/2$ and can even be zero for this configuration of magnetization vectors. Second, a π state in the SFFIS Josephson junction can be realized for parallel orientations of magnetizations in the F layers as a result of phase shifts at the interfaces.

II. RESULTS AND DISCUSSION

To prove the above statements, we consider the SFF structure presented in Fig. 1.

This structure consists of two identical single-domain ferromagnetic films, which may differ only by a value of exchange energy H_1 and H_2 for lower and upper layers, respectively. The magnetization vector of the lower F film is directed along the y axis, while in the upper film it may be deflected by angle α from this direction in the yz plane. We will also suppose that a dirty limit condition is valid for all the films and that the transparency of the SF interface is small enough to provide the opportunity to use a linearized Usadel equation in the form given by Bergeret *et al.*^{3,31} Thus, for the upper F film,

$$\xi_F^2 \nabla^2 f_0 - \Omega f_0 - i h_2 \cos \alpha f_3 = 0,$$

$$\xi_F^2 \nabla^2 f_3 - \Omega f_3 - h_2 \sin \alpha f_1 - i h_2 \cos \alpha f_0 = 0, \quad (1)$$

$$\xi_F^2 \nabla^2 f_1 - \Omega f_1 + h_2 \sin \alpha f_3 = 0.$$

Here, index i = 0,1,3 represents triplet condensate functions with 0 and ± 1 spin projections and a singlet condensate function, $\xi_F^2 = (D_F/2\pi T_c)$; D_F is the diffusion coefficient of the F material; $\Omega = \omega/(\pi T_c)$ are Matsubara frequencies; and $h_2 = H_2/(\pi T_c)$. The system of Usadel equations for the condensate functions p_i for the lower F film has the same



FIG. 1. SFF structure.

form as Eq. (1) with $\alpha = 0$ and where $h_1 = H_1/(\pi T_c)$ takes the place of h_2 .

Usadel equations must be supplemented by boundary conditions. At the FF interface (x = 0), they have the form⁴⁹

$$\gamma_B \xi_F \frac{\partial}{\partial x} f_i + f_i = p_i,$$

$$\frac{\partial}{\partial x} p_i = \frac{\partial}{\partial x} f_i,$$

$$i = 0, 1, 3.$$
 (2)

Here we consider the arbitrary transparency of the FF interface, described by suppression parameter γ_B (Ref. 1). At the SF interface ($x = d_F$)

$$\xi_F \frac{\partial}{\partial x} p_3 = \frac{\Delta}{\gamma_{BS} \sqrt{\Omega^2 + \Delta^2}},$$

$$\frac{\partial}{\partial x} p_{0,1} = 0,$$
(3)

where suppression parameter γ_{BS} (Ref. 1) describes the SF interface and Δ is the magnitude of order parameter in the S film, normalized on πT_c . Large values of γ_{BS} permit us to neglect the suppression of order parameter in the S film and consider Δ in Eq. (3) only as a temperature-dependent value.

For simplicity, we consider the limit of thin F films $(d_F/\xi_F \ll 1)$. In this limiting case, Green's functions in the first approximation of d_F/ξ_F are independent of space coordinate constants and can be similar to Karminskaya and Kupriyanov's⁵⁰ findings,

$$p_{1} = -\Gamma \frac{h_{2}\gamma_{BN}\sin(\alpha)S}{u^{2} \left(h_{2}^{2}\gamma_{BN} + v\right) \left(h_{1}^{2}\gamma_{BN} + v\right) - S^{2}},$$

$$p_{0} = -i\Gamma\gamma_{BN} \frac{h_{2}\cos(\alpha)S - h_{1}u^{2} \left(v + h_{2}^{2}\gamma_{BN}\right)}{u^{2} \left(h_{2}^{2}\gamma_{BN} + v\right) \left(h_{1}^{2}\gamma_{BN} + v\right) - S^{2}},$$

$$p_{3} = \Gamma \frac{\gamma_{BN}uv \left(h_{2}^{2}\gamma_{BN} + v\right)}{u^{2} \left(h_{2}^{2}\gamma_{BN} + v\right) \left(h_{1}^{2}\gamma_{BN} + v\right) - S^{2}},$$

$$f_{0} = -i\Gamma \frac{\gamma_{BN}u \left[h_{2}\cos(\alpha)S - h_{1} \left(v + \gamma_{BN}h_{2}^{2}\right)\right]}{u^{2} \left(h_{2}^{2}\gamma_{BN} + v\right) \left(h_{1}^{2}\gamma_{BN} + v\right) - S^{2}},$$
(4)

$$f_3 = -\Gamma \frac{\gamma_{BN} v S}{u^2 \left(h_2^2 \gamma_{BN} + v\right) \left(h_1^2 \gamma_{BN} + v\right) - S^2},$$

$$f_1 = P_1 u,$$

where parameter $\Gamma = \frac{\Delta \xi_F}{\gamma_{BS} d_F \sqrt{\Omega^2 + \Delta^2}}, \quad \gamma_{BN} = d_F \gamma_B / \xi_F$ describes transparency of the FF interface, $S = h_1 h_2 \gamma_{BN} \cos(\alpha) - \Omega(u+1), u = \Omega \gamma_{BN} + 1$, and $v = \Omega(u+1)$.

In the case of collinear orientation of magnetization vectors, long-range triplet components p_1 and f_1 are zero and it is



FIG. 2. $\operatorname{Re}(p_+)$ (solid line) and $\operatorname{Re}(f_+)$ (dashed line) vs parameter γ_{BN} for misorientation angle $\alpha = 0$, $\Omega = 0.5$, and h = 10.

convenient to deal with complex condensate functions $p_+ = p_3 + p_0$ and $f_+ = f_3 + f_0$. In the Matsubara representation, singlet components p_3 and f_3 are real and short-range triplet components p_0 and f_0 are purely imaginary quantities.

For antiparallel orientation of magnetization vectors in both ferromagnetic films ($\alpha = \pi$), functions p_3 and f_3 have the same sign for any value of transparency of the FF interface, due to compensation of magnetizations in the F layers. This property is clearly seen in Eq. (4).

For parallel orientation of magnetizations ($\alpha = 0$), the situation is more complex, since parameter *S* in Eq. (4) can change sign as a function of γ_{BN} . Figure 2 shows the dependencies of real parts of condensate functions for middle ferromagnet $\text{Re}(p_+) = p_3$ (solid line) and for upper ferromagnet $\text{Re}(f_+) = f_3$ (dashed line) on the parameter γ_{BN} when $h_1 = h_2 = h$. For the highly transparent interface $p_3 = f_3$, the real part of the condensate function in the upper ferromagnet changes sign at $\gamma_{BN} = \frac{2\Omega}{h_1h_2-\Omega^2}$ with the increase of γ_{BN} , due to the behavior of phases of complex functions p_+ and f_+ . In Fig. 3 the dependencies of these phases, $\text{Arg}(p_+)$ and $\text{Arg}(f_+)$, are shown vs the exchange field h.

In the high-transparency regime $\gamma_{BN} = 0$, proximity coupling of the upper and middle ferromagnetic films is strong and the phases of functions p_+ and f_+ coincide. However, for nonzero γ_{BN} , the films can become effectively separated at large values of h. Specifically, with an increase of h, the imaginary parts of the condensate functions p_0 and f_0 increase and phase slips at both interfaces are generated. In accordance with the result of Golubov *et al.*⁴⁸ for a single SF bilayer, the phase slips reach $-\pi/2$ at a large h. As a result, the phase of the upper F film shifts by $-\pi$ with respect to S,



FIG. 3. Arg(p_+) and Arg(f_+) vs value of exchange field h for misorientation angle $\alpha = 0$, $\Omega = 0.5$. $\gamma_{BN} = 1$ (solid line) and $\gamma_{BN} = 0.01$ (dashed line).



FIG. 4. Singlet components $\text{Re}(p_+)$, $\text{Re}(f_+)$ and triplet components $\text{Im}(p_+)$, $\text{Im}(f_+)$, p_1 , f_1 vs misorientation angle α at $h_1 = 10$ and $h_2 = 30$, $\Omega = 0.5$, and $\gamma_{BN} = 0$, 0.01 (solid and dashed lines, respectively).

while the phase of middle film is saturated on $-\pi/2$ at a large h. The point where the upper F film's phase crosses the value $-\pi/2$ corresponds to the sign change of the singlet component f_3 in this film.

As a result, for a sufficiently strong exchange field, the singlet component f_3 has opposite signs in parallel and antiparallel configurations. Therefore, an f_3 sign change should occur at some intermediate angle α . The dependencies of condensate functions in upper and middle ferromagnetic layers on angle α given by Eq. (4) are presented in Fig. 4 for $h_1 = 10, h_2 = 30$, and $\Omega = 0.5$ for two cases: $\gamma_{BN} = 0$ and $\gamma_{BN} = 0.01$.

It can be seen from Eq. (4) and Fig. 4 that singlet condensate function in the upper F film f_3 equals zero at an angle

$$\alpha_{in} = \pm \arccos\left(\frac{\Omega^2 + \Omega/\gamma_{BN}}{h_1 h_2}\right),\tag{5}$$

where the p_3 component in the lower F film has a finite value.

This influences the behavior of triplet components p_1 and f_1 . If the singlet component vanishes on at least one side of the FF interface, the triplet components p_1 and f_1 are not generated in the system, while triplet components p_0 and f_0 can still be nonzero. As a result, triplet condensate functions p_1



FIG. 5. I_C of SFFIS junction vs misorientation angle α , for $h_1 = 10$, $h_2 = 30$, and $T = 0.5T_C$. $\gamma_{BN} = 0$ (solid line) and $\gamma_{BN} = 0.1$ (dotted line).

and f_1 are zero not only at angles $\alpha = 0$, π but also at some intermediate angle α_{in} given by Eq. (5), if the FF interface transparency has a finite value.

It is clearly seen that the sign-reversal effect is absent in the limiting case of a transparent FF interface

$$f_{0} = p_{0} = i\Gamma \frac{h_{2}\cos(\alpha) + h_{1}}{h_{1}^{2} + h_{2}^{2} + 2h_{1}h_{2}\cos(\alpha) + 4\Omega^{2}},$$

$$f_{3} = p_{3} = \Gamma \frac{2\Omega}{h_{1}^{2} + h_{2}^{2} + 2h_{1}h_{2}\cos(\alpha) + 4\Omega^{2}},$$

$$f_{1} = p_{1} = \Gamma \frac{h_{2}\sin(\alpha)}{h_{1}^{2} + h_{2}^{2} + 2h_{1}h_{2}\cos(\alpha) + 4\Omega^{2}}.$$
(6)

Another interesting effect is a significant enhancement of the magnitudes of singlet components for some range of values of parameter γ_{BN} and related enhancement of triplet components with respect to the transparent FF interface.

These effects should lead to observable features in the critical current of an SFFIS Josephson junction consisting of an SFF trilayer coupled to a superconductor S across tunnel barrier I. In this case, the current-phase relation is sinusoidal, with the Josephson critical current given by the simple expression

$$I_C = \frac{\pi T}{eR_I} \sum_n f_3 \frac{\Delta}{\sqrt{\Delta^2 + \Omega^2}},$$

where R_I is the resistance of the interface I. The resulting dependence of the critical current on angle α is presented in Fig. 5 for two different values of suppression parameter $\gamma_{BN} = 0$ and $\gamma_{BN} = 0.1$.



FIG. 6. I_C of SFFIS junction vs temperature T for $\alpha = 0$, $h_1 = 10$, and $h_2 = 30$. $\gamma_{BN} = 0.005$ (dotted line), $\gamma_{BN} = 0.009$ (solid line), and $\gamma_{BN} = 0.014$ (dashed line).

It is seen that the critical current changes sign at some intermediate angle for a structure with nonzero γ_{BN} on the FF interface (dotted line), in contrast with zero γ_{BN} (solid line). The dependence of the critical current on the angle corresponds to dependence of singlet condensate function f_3 (Fig. 4); however, the angle at which the critical current changes sign does not coincide with the one given by Eq. (5) because of summation over Matsubara frequencies in the expression for critical current. To summarize, the $0-\pi$ transition may take place in the SFFIS junction as function of misorientation angle α , if the FF interface has finite transparency.

Interestingly, the $0-\pi$ transition may also occur at zero α as a function of temperature, as shown in Fig. 6. The low-temperature critical current is very sensitive to the magnitude of γ_{BN} : It is seen that the critical current changes sign at low temperatures in certain ranges of γ_{BN} , while at temperatures near T_C , the critical current is still positive (see the solid line in Fig. 6). Temperature-induced $0-\pi$ transition was observed by Ryazanov *et al.*,⁷ in long SFS junctions where a π state is realized due to the oscillatory nature of the condensate function with thin ferromagnetic layers, there are no oscillations of the condensate functions in the F layer. When the magnetic layers, while the π state is realized due to accumulation of phase shifts at SF and FF

interfaces. With further increase of γ_{BN} , SFFIS junction is in the π state at all temperatures (see the dashed line in Fig. 6).

III. CONCLUSION

In conclusion, we have investigated the proximity effect in SFF structures with finite FF interface transparency. We have shown that due to phase shift at the FF interface, long-range triplet pair correlations vanish not only at collinear orientations of magnetizations in both layers $\alpha = 0$ and π , but also at some intermediate angle α . This angle depends on parameters of the structure and typically is close to $\pi/2$, where triplet correlations in symmetric FSF structure are strongest. Moreover, maximum amplitudes of long-range triplet and singlet pair correlations are achieved at a finite transparency of the FF interface, not at an ideal transparency, as expected. The predicted effects manifest themselves in SFFIS Josephson junctions, where the peculiarities of the proximity effect in the SFF trilayer lead to the possibility of realizing a π state.

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