

Long-range spin-triplet proximity effect in Josephson junctions with multilayered ferromagnets

Luka Trifunovic and Zoran Radović

Department of Physics, University of Belgrade, P.O. Box 368, 11001 Belgrade, Serbia

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We study the proximity effect in SF'(AF)F'S and SF'(F)F'S planar junctions, where S is a clean conventional (*s*-wave) superconductor, while F' and middle layers are clean or moderately diffusive ferromagnets. Middle layers consist of two equal ferromagnets with antiparallel (AF) or parallel (F) magnetizations that are not collinear with magnetizations in the neighboring F' layers. We use fully self-consistent numerical solutions of the Eilenberger equations to calculate the superconducting pair amplitudes and the Josephson current for arbitrary thickness of ferromagnetic layers and the angle between in-plane magnetizations. For moderate disorder in ferromagnets, the triplet proximity effect is practically the same for AF and F structures, like in the dirty limit. Triplet Josephson current is dominant for $d' \approx \hbar v_F / 2h'$, where d' is the F' layer thickness and h' is the exchange energy. Our results are in a qualitative agreement with the recent experimental observations [T. S. Khaire, M. A. Khasawneh, W. P. Pratt, and N. O. Birge, *Phys. Rev. Lett.* **104**, 137002 (2010)].

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In hybrid systems containing superconducting and ferromagnetic metals, triplet correlations are induced.¹⁻⁴ In the case of homogeneous magnetization, the triplet pair amplitude has zero total spin projection on the magnetization axis: $F_{t_0}(t-t') \sim |\uparrow(t)\downarrow(t')\rangle + |\downarrow(t')\uparrow(t)\rangle$. For equal time $t=t'$, this function vanishes, in agreement with the Pauli principle. Therefore, F_{t_0} is an odd function of the time difference or equivalently, odd in frequency. The exchange field mixes the usual spin singlet pairing correlations, $F_s \sim |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$, and the spin triplet F_{t_0} because the wave functions $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$ acquire relative phase difference in the ferromagnet.⁵

Amplitudes F_s and F_{t_0} are short-ranged and oscillate spatially in the ferromagnet, both in the clean and the dirty limit.³ In the clean limit, they decay algebraically in the ferromagnet with $\hbar v_F / h$, where h is the exchange energy and v_F is the Fermi velocity. In the dirty limit, they decay exponentially with the characteristic length $\sqrt{\hbar D} / h$, where $D = v_F l / 3$ is the diffusion coefficient and l is the electron mean-free path in the ferromagnet. However, this is not true for the clean single-channel junctions where all pairing correlations are long ranged and their spatial decay in the ferromagnet is independent of the exchange field.⁴

The situation is quite different for inhomogeneous magnetization: not only F_s and F_{t_0} amplitudes exist but also odd triplet pair amplitude F_{t_1} with ± 1 total spin projection on the magnetization axis emerges in the ferromagnet.¹⁻³ The triplet component F_{t_1} is not suppressed by exchange interaction and penetrates into the ferromagnet over large distance on the order of $\sqrt{\hbar D} / k_B T$ in the dirty limit ($l < \hbar v_F / h$), and, likewise, over the distance $\hbar v_F / k_B T$ in the clean limit ($l > \hbar v_F / k_B T$).

It is not difficult to understand why F_{t_1} triplet component is induced in inhomogeneous ferromagnets. Consider a system where inhomogeneity is represented by two ferromagnetic layers with noncollinear magnetizations and angle α between them. As we have already pointed out, in each ferromagnetic layer the exchange field generates F_{t_0} from F_s . When F_{t_0} correlation penetrates into neighboring ferromagnetic layer it mixes with F_{t_1} having nonzero total spin projection due to different orientation of magnetizations. This

implies that F_{t_1} triplet component is generated in the presence of inhomogeneous magnetization and is proportional to $F_{t_0} \sin \alpha$ at the interface between two ferromagnetic layers. Therefore, for fully developed triplet proximity effect one of two layers should be sufficiently thin to provide large F_{t_0} at the interface between the layers.^{6,7}

Another possibility for dominant long-range triplet component was suggested in Refs. 8–10. In that approach, the role of thin ferromagnetic layers is replaced by spin active interfaces described by scattering matrix. The elements of the scattering matrix can be considered as phenomenological parameters. Purely microscopic approach that retains quasi-particle information at the atomic scale, with spin-dependent scattering potentials at the interfaces, was considered in Ref. 11.

Besides the impact on the Josephson current, another signature of odd-frequency triplet pair correlations has been proposed recently. The density of states in the presence of the odd-frequency pairing is enhanced and acquires a zero-energy peak.^{12,13}

Experimental results that may support the existence of long-range triplet amplitudes have already been obtained.¹⁴⁻¹⁶ However, in these experiments it was not possible to tune the junction parameters, and the transition from usual singlet to long-range triplet proximity effect has not been observed.

Quite recently, long-range triplet proximity effect has been observed in SF'(AF)F'S junctions with F' layer thickness as a controllable parameter.¹⁷ Here F' is a weak ferromagnetic layer (PdNi or CuNi) and AF is synthetic antiferromagnet: an exchange-coupled trilayer Co/Ru/Co. Previously, a similar arrangement (with homogeneous middle layer F) has been analyzed theoretically and proposed as a good candidate for the long-range triplet effect.^{7,13}

Junctions with F and AF middle layers were analyzed recently for the case when F' layer thickness is much smaller than $\sqrt{\hbar D} / h'$.¹⁸ However, within this approximation the triplet component is not dominant and consequently results are not applicable to the experiment (Ref. 17). More interesting case was considered by the same authors, when F' layer

thickness is arbitrary but the middle layer is homogeneous, as in Ref. 7. In the dirty limit (and moderately disordered ferromagnets as we will show), results are practically the same for F and AF structures of the middle layer. The situation is different for the case of clean ferromagnets.

In this Rapid Communication, we study the proximity effect in clean or moderately diffusive SF'(AF)F'S and SF'(F)F'S planar junctions, where S is a conventional (*s*-wave) superconductor, F' and middle layers are ferromagnets. Middle layer consists of two equal ferromagnets with parallel (F) or antiparallel (AF) magnetizations, and magnetizations in F' layers are not collinear in general with magnetizations in the neighboring layers.

To calculate the Josephson current and pair correlations in the clean limit and for moderately diffusive ferromagnets, we use the Eilenberger equations¹⁹ for a junction along the *x* axis,

$$\hbar v_x \partial_x \check{g} + [\omega_n \hat{\tau}_3 - i\check{V} + \check{\Delta} + \hbar \langle \check{g} \rangle / 2\tau, \check{g}] = 0 \quad (1)$$

with normalization condition $\check{g}^2 = \check{1}$. Disorder is characterized by the average time $\tau = l/v_F$ between scattering on impurities. We indicate by $\hat{\cdot}$ and $\check{\cdot}$ 2×2 and 4×4 matrices, respectively. Here, θ is an angle between the Fermi velocity and the *x* axis, $\hat{\tau}_i$ are the Pauli matrices in the particle-hole space, the brackets $\langle \cdot \cdot \rangle$ denote angular averaging over the Fermi surface (integration over θ) and $[\cdot \cdot \cdot]$ denotes a commutator. The quasiclassical Green's functions,

$$\check{g} = \begin{pmatrix} g_s + \mathbf{g}_t \cdot \hat{\boldsymbol{\sigma}} & (f_s + \mathbf{f}_t \cdot \hat{\boldsymbol{\sigma}}) i \hat{\sigma}_y \\ -(\tilde{f}_s + \tilde{\mathbf{f}}_t \cdot \hat{\boldsymbol{\sigma}}^*) i \hat{\sigma}_y & -(g_s + \mathbf{g}_t \cdot \hat{\boldsymbol{\sigma}}^*) \end{pmatrix}, \quad (2)$$

where $\hat{\boldsymbol{\sigma}} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ are the Pauli matrices in spin space. With this parametrization,⁹ it is clear that under rotation of magnetizations g_s and f_s remain unchanged while \mathbf{g}_t and \mathbf{f}_t transform like ordinary vectors. The conjugate Green's functions satisfy $\tilde{f}_s(\omega_n) = f_s^*(-\omega_n)$ and $\tilde{f}_t(\omega_n) = -\mathbf{f}_t^*(-\omega_n)$.²⁰ The exchange field term is given by $\check{V} = \text{Re}[\mathbf{h}(x) \cdot \hat{\boldsymbol{\sigma}}] + i \hat{\tau}_3 \text{Im}[\mathbf{h}(x) \cdot \hat{\boldsymbol{\sigma}}]$ and the pair potential $\check{\Delta} = (\hat{\tau}_+ \Delta + \hat{\tau}_- \Delta^*) \hat{\sigma}_y$, $\hat{\tau}_\pm = \hat{\tau}_x \pm i \hat{\tau}_y$. The exchange field \mathbf{h} has the following *x* dependence:

$$\mathbf{h} = \begin{cases} h'(-\sin \alpha y + \cos \alpha z) & -d - d' < x < -d \\ hz & -d \leq x < 0 \\ \pm hz & 0 \leq x < d \\ h'(\sin \alpha y + \cos \alpha z) & d \leq x < d + d', \end{cases} \quad (3)$$

where d' and $2d$ are F' and (F) or (AF) layer thickness, respectively. The angle between magnetizations in F' and neighboring ferromagnetic layers is α , the plus (minus) sign is for F (AF) middle layer. In the absence of out-of-plane magnetization, the amplitude $(\mathbf{f}_t)_x = 0$.

The supercurrent flowing through the junction is given by the normal Green's function,

$$I(\phi) = \pi e N(0) S k_B T \text{Im} \sum_{\omega_n} \langle v_x g_s(v_x) \rangle, \quad (4)$$

where ϕ is the macroscopic phase difference across the junction, $N(0)$ is the density of states per spin at the Fermi sur-

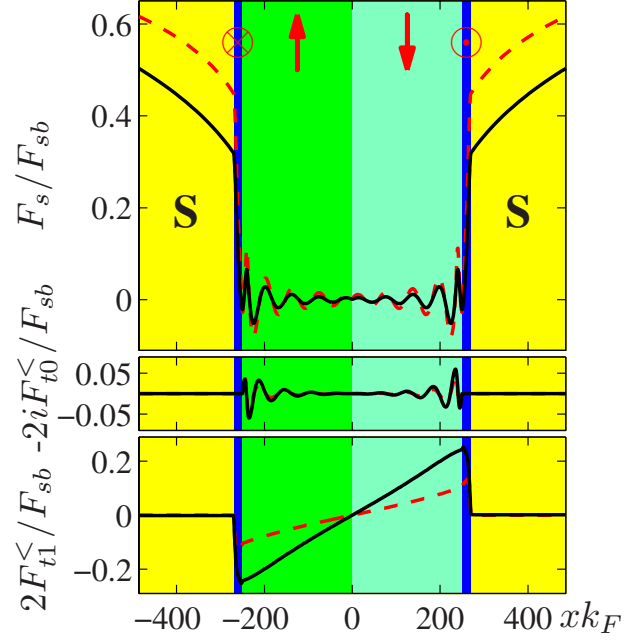


FIG. 1. (Color online) Spatial dependence of singlet and triplet pair amplitudes F_s , $F_{t0}^<$, and $F_{t1}^<$, normalized to the bulk singlet amplitude F_{sb} , for $T/T_c=0.1$, $h'/E_F=0.05$, $h/E_F=0.1$, $2d=500k_F^{-1}$, $d'=20k_F^{-1}$, $\alpha=\pi/2$ and two values of the mean-free path: $l=\infty$ (dashed curves) and $l=200k_F^{-1}$ (solid curves). All amplitudes are calculated for the ground state, $\phi=0$. The SF'(AF)F'S junction geometry is shown in the background; arrows and circles show orientation of magnetizations for each layer.

face, and S is the area of the junction. Equal-time pair amplitudes are defined in terms of anomalous Green's functions as

$$F_s = -i\pi N(0) k_B T \sum_{\omega_n} \langle f_s \rangle, \quad (5)$$

$$F_{t0}^< = \pi N(0) k_B T \sum_{\omega_n < 0} \langle (f_t)_z \rangle, \quad (6)$$

$$F_{t1}^< = -i\pi N(0) k_B T \sum_{\omega_n < 0} \langle (f_t)_y \rangle. \quad (7)$$

Equal-time triplet amplitudes identically vanish according to the Pauli principle, hence we defined auxiliary functions using summation over negative frequencies only. The spatial variation of the time-dependent triplet pair amplitudes is qualitatively the same as for auxiliary functions.

We consider the case of fully transparent interfaces, i.e., strong proximity effect. The opposite case of low transparency was considered in Ref. 18. Using continuity of the Green's functions at interfaces, Eq. (1) is solved iteratively with the self-consistency condition $\Delta = \lambda F_s$. Iterations are performed until self-consistency is reached, starting from the stepwise approximation for the pair potential $\Delta = \Delta(T) [e^{-i\phi/2} \Theta(-x-d-d') + e^{i\phi/2} \Theta(x-d-d')]$. The temperature dependence of the bulk pair potential is given by

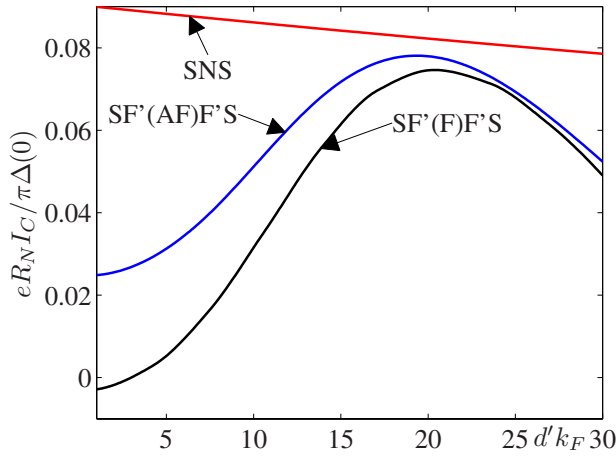


FIG. 2. (Color online) Dependence of the Josephson critical current on the F' ferromagnetic layer thickness d' , for $2d=500k_F^{-1}$, $T/T_c=0.1$, $h'/E_F=0.05$, $h/E_F=0.3$, $l=200k_F^{-1}$, and for three types of junctions: SNS, SF'(AF)F'S, and SF'(F)F'S.

$\Delta(T)=\Delta(0)\tanh(1.74\sqrt{T_c/T-1})$. For arbitrary mean-free path in ferromagnets, we employ the iterative procedure starting from the clean limit.

Figure 1 shows the spatial variation of the pair amplitudes for the SF'(AF)F'S junction geometry with magnetizations in F' layers orthogonal to the neighboring middle layers, $\alpha=\pi/2$. In this case, the 0 state is the ground state. Transition to the π state can be tuned with relative orientation of magnetizations in F' layers (π state is the ground state for parallel magnetizations in the two F' layers). For corresponding SF'(F)F'S junctions, pair amplitudes are practically the same. For $\phi=0$, the singlet and the long-range triplet amplitudes, F_s and F_{l1} , are real while the short-range triplet amplitude F_{l0} is imaginary.

The F' layer thickness $d'=20k_F^{-1}$ is chosen to give the maximum triplet current for moderate disorder in ferromagnets ($l=200k_F^{-1}$). This explains the large difference between amplitudes of the long-range triplet component in the clean and moderately disordered case, Fig. 1.

We observe oscillatory decay of F_s and $F_{l0}^<$ amplitudes, dependent on the exchange field. In contrast, long-range triplet component, $F_{l1}^<$, is monotonic in the middle layer and its decay length is independent of the exchange field magnitude. Thin F' layers are considered as weak ferromagnets, $h'/E_F=0.05$, and thick middle layers, $2d=500k_F^{-1}$, represent strong ferromagnets. For illustration of pair amplitudes, $h/E_F=0.1$ is used in order to have larger period of spatial oscillations, although $h/E_F=0.3$ is used in other illustrations. All amplitudes are normalized to the bulk singlet pair amplitude $F_{sb}=2\pi N(0)k_B T \sum_{\omega_n} \Delta / \sqrt{\omega_n^2 + \Delta^2}$.

Next we examine the Josephson critical current dependence on F' layer thickness. This quantity is actually measured in the experiment.¹⁷ Figure 2 illustrates $I_C(d')$, normalized to the resistance $R_N=2\pi^2\hbar/Se^2k_F^2$, for three types of junctions: SF'(F)F'S, SF'(F)F'S, and SNS, where N is the corresponding normal nonmagnetic metal ($h=h'=0$). Here, we take mean-free path $l=200k_F^{-1}$ in ferromagnetic and N metals.

In the clean limit, $I_C(d')$ curves for SF'(AF)F'S and the

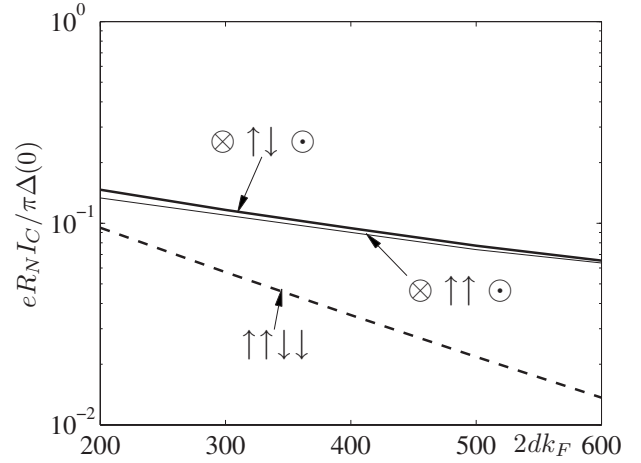


FIG. 3. Dependence of the Josephson critical current on the middle layer thickness $2d$, for $d'=20k_F^{-1}$, $T/T_c=0.1$, $h'/E_F=0.05$, $h/E_F=0.3$, $l=200k_F^{-1}$, and for two values of the misorientation angle: $\alpha=\pi/2$ (thick and thin solid lines for AF and F structures) and for $\alpha=0$ (AF structure, dashed line). Arrows and circles show orientation of magnetizations in ferromagnetic layers.

corresponding SNS junctions coincide⁵ while in the dirty limit, $I_C(d')$ curves for SF'(AF)F'S and SF'(F)F'S curves are practically the same.¹⁸ For intermediate disorder in ferromagnets, the critical current is always larger in SF'(AF)F'S than SF'(F)F'S junctions, Fig. 2.

We find for moderate disorder in ferromagnets that the largest critical current is almost as big as for the corresponding SNS junction. Note that in the dirty limit, maximum triplet critical current is only 12% of the critical current of the corresponding SNS junction.¹⁸ The position of maxima of $I_C(d')$ strongly depends on h' , $d'_{max} \approx \hbar v_F / 2h'$. It is practically independent of h and d , in agreement with the results of Ref. 18.

The critical Josephson current dependence on the middle layer thickness is shown in Fig. 3. The F' layer thickness is set to $d'_{max}=20k_F^{-1}$. We consider I_C dependence on $2d$ for two values of misorientation angle: $\alpha=\pi/2$ (fully developed triplet proximity effect, solid lines) and $\alpha=0$ (no long-range triplet component, dashed line). For thick middle layer, $2d=600k_F^{-1}$, in both AF and F geometries, I_C is an order of magnitude larger when the triplet component F_{l1} is present. These results, Figs. 2 and 3, are in a qualitative agreement with experimental observation.¹⁷

The current-phase relation is almost harmonic for a moderately disordered SF'(AF)F'S junction ($l=200k_F^{-1}$, Fig. 4. Here, the F' layer thickness is optimal for long-range triplet Josephson effect, i.e., the usual singlet Josephson critical current is an order of magnitude smaller. For the in-plane magnetizations considered here, we did not find any unusual $I(\phi)$ dependence, in agreement with Refs. 7 and 18.

However, we expect unusual $I(\phi)$ dependence for the samples used in the experiment (Ref. 17). Supercurrent could be observed even for $\phi=0$, if one takes into account the possibility of inhomogeneous out-of-plane magnetizations in PdNi layers and the fact that transport properties of the minority and majority electrons at the Fermi surface in Co layers are very different. Quasiclassical approximation we used

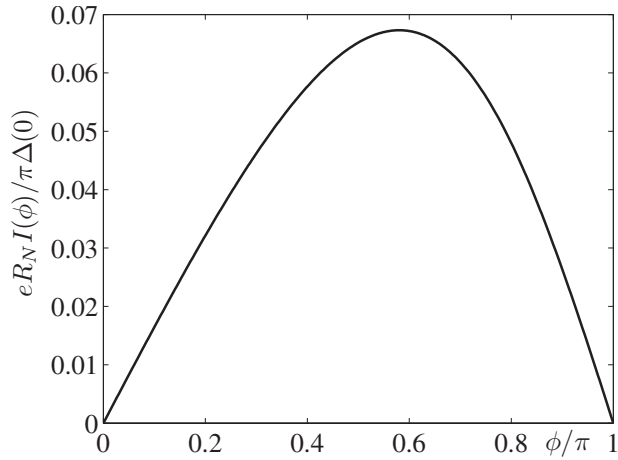


FIG. 4. The current-phase relation $I(\phi)$ for SF'(AF)F'S junction: $d' = 20k_F^{-1}$, $2d = 600k_F^{-1}$, $T/T_c = 0.1$, $h'/E_F = 0.05$, $h/E_F = 0.3$, $l = 200k_F^{-1}$, and $\alpha = \pi/2$.

here does not allow for the latter possibility; this is the principal limitation of our approach.

A new long-range triplet component, $(f_t)_x = (f_{\uparrow\uparrow} - f_{\downarrow\downarrow})/2$, is generated in the case of nonzero relative longitude angle χ

between magnetizations in two F' layers (i.e., inhomogeneous out-of-plane magnetization). This can be readily seen from Eq. (1) for $(f_t)_x$ component. The presence of $(f_t)_x$ implies different triplet Josephson current flow for majority (I_{\uparrow}) and minority (I_{\downarrow}) electrons. In the circuit theory approximation,¹³ it was obtained that $I_{\uparrow,\downarrow} = C_{\uparrow,\downarrow} \sin(\phi \pm \chi)$, where $C_{\uparrow,\downarrow}$ are proportional to densities of states and Fermi velocities. Although the case of very different Fermi velocities of the subbands is not accessible within the circuit theory approximation, it is reasonable to assume that similar current-phase relations are valid. Consequently, there is a finite supercurrent at zero phase difference, as was predicted for the half-metallic middle layer.¹³

It would be very interesting to measure the $I(\phi)$ dependence for the samples used in the experiment (Ref. 17) since the out-of-plane magnetizations in thin PdNi layers are their typical feature.²¹ A nonzero supercurrent for $\phi=0$ could provide an unambiguous evidence of the triplet proximity effect.

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