

Long-Range Superharmonic Josephson Current

Luka Trifunovic

Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

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We consider a long superconductor-ferromagnet-superconductor junction with one spin-active region. It is shown that an odd number of Cooper pairs cannot have a long-range propagation when there is only one spin-active region. When the temperature is much lower than the Thouless energy, the coherent transport of two Cooper pairs becomes the dominant process and the superharmonic current-phase relation is obtained ($I \propto \sin 2\phi$).

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The interplay between superconducting and ferromagnetic ordering has been the subject of intensive theoretical and experimental research [1–9]. It has been predicted that the hybrid systems containing superconductors (S) and ferromagnets (F) allow the realization of Josephson π junctions, $I \propto \sin(\phi + \pi)$ [1]. The spectral decomposition of the Josephson current-phase relation (CPR) gives $I = I_1 \sin\phi + I_2 \sin 2\phi \dots$. At the transition between the 0 and the π phases, I_1 vanishes, and it is possible to obtain dominant second harmonic (I_2) in the CPR. Unfortunately, the 0- π coexistence is very sensitive to the temperature changes and interface roughness [2].

The experimental realizations of π junctions remained elusive for a long time; the breakthrough came with the fabrication of weak ferromagnets [3]. Indeed, it has been recognized that the proximity effect in a ferromagnet is short-ranged. The electron and hole excitations acquire a nonzero relative phase in the ferromagnet between the scatterings from the two superconductor-ferromagnet (SF) interfaces [1]; the different orbital modes acquire different phases that add up destructively after summation. A detailed analysis demonstrates that the proximity effect in a superconductor-ferromagnet-superconductor (SFS) junction is suppressed algebraically in the ballistic regime, and exponentially in the diffusive one [4].

Quite recently, it was proposed that a SFS junction with an inhomogeneous magnetization in the F layer can generate a triplet pairing and can support a long-range Josephson current [4]. Subsequent theoretical and experimental research showed that in order to have dominant triplet pairing a SFS junction with two spin-active regions is required [5–8].

Motivation for this Letter is our previous numerical calculation [10], where clean and moderately disordered SFS junctions were considered (with a one spin-active interface), and the dominant second harmonic was obtained.

In this work, we consider a long SFS junction in the ballistic regime. Assuming the presence of a spin-active region on only one SF interface, we show that only the phase coherent transport of an even number of Cooper pairs is not suppressed by the exchange field. In particular,

the dominant contribution to the Josephson current stems from the transport of two Cooper pairs. As a consequence, the two times smaller flux quantum is obtained, leading to more sensitive quantum interferometers (SQUIDs) [11] and the half-integer Shapiro steps that can be experimentally observed [9]. Another interesting property is the coexistence of integer and half-integer fluxoid configurations in SQUIDs, corresponding to the minima of the triple-well potential energy [11]; this can be potentially useful for experimental study of the quantum superposition of macroscopically distinct states [12]. Also, junctions with a nonsinusoidal current-phase relation are shown to be promising for realization of “silent” phase qubits [13]. Last but not least, this result enables robust realization of so-called φ junctions [14].

It should be stressed that in contrast to the case of the 0- π transition the discovered effect is very robust: it is insensitive to a weak disorder [10], temperature changes, and the interface roughness. Nevertheless, relatively transparent interfaces are required in order to observe the effect.

Before we proceed with a quantitative analysis of the aforesaid effect, let us first give a simple and intuitive description. Note that when a SF interface is spin-active, there are two possibilities for Andreev reflection: the normal Andreev reflection (the spin projections of an electron and the reflected hole are opposite) and the anomalous Andreev reflection (an electron and the reflected hole have the same spin projections) [15]. We consider separately the phase coherent transport of one (I_1) and two (I_2) Cooper pairs. The transport of a single Cooper pair is suppressed by the exchange field because the electron and the Andreev reflected hole have opposite spin projections (see Fig. 1). On the other hand, the transport of two Cooper pairs has a long-range contribution stemming from two normal and two anomalous Andreev reflections.

We consider a simple model of a ballistic SFS junction consisting of two conventional (s -wave) superconductors, a uniform single-domain ferromagnet, and only one spin-active region. Andreev reflection requires relatively transparent SF interfaces; thus, for simplicity we assume them to be fully transparent. The spin-active region consists of a

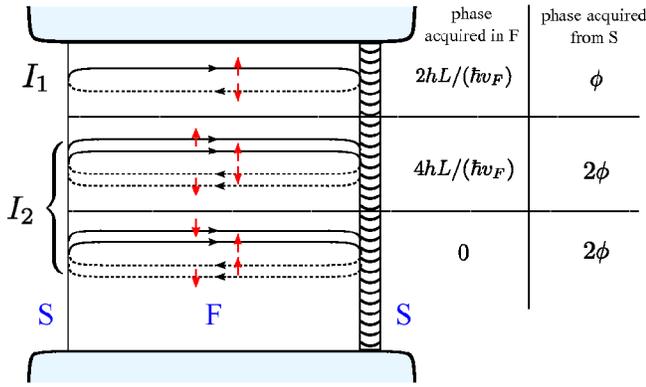


FIG. 1 (color online). The first two harmonics in the Josephson current-phase relation. The first one (I_1) consists of two normal Andreev reflections from SF interfaces. The second one (I_2) has two contributions: the one with four normal Andreev reflections (short-range), the other with two normal and two anomalous Andreev reflections (long-range, total phase acquired in F layer is zero). The solid (dotted) lines represent electron (hole) excitations in the F layer. The red (vertical) arrows represent spin projections, while the black (horizontal) arrows denote the excitation velocity direction. The right SF boundary (hatched) is spin-active.

ferromagnetic spacer layer with the magnetization noncollinear to that of the F layer.

The Josephson current is calculated using the scattering approach [16]. The knowledge of scattering matrices (S matrices) of both SF interfaces is sufficient to obtain the Josephson current in the ballistic regime. Each of these matrices relates the amplitudes of the excitations propagating towards the corresponding SF interface to the excitation propagation away from it. There is no orbital channel mixing in the ballistic regime, but only mixing of different spin channels (due to the spin-active region) and the particle-hole mixing (due to superconductors). Ergo, the dimension of the S matrix is 4×4 —we write all matrices in the Kronecker product of particle-hole and spin spaces. The Josephson current is given by [16]

$$I = -\frac{2ek_B T}{\hbar} \frac{d}{d\phi} \sum_{n=0}^{\infty} \ln \det[1 - \mathcal{R}(i\omega_n) \mathcal{R}'(i\omega_n)], \quad (1)$$

where \mathcal{R}' and \mathcal{R} are S matrices of the two SF interfaces, while the phases acquired upon propagation through the F region are included in one of these matrices. $\omega_n = (2n+1)\pi k_B T$ are the Matsubara frequencies and ϕ is the phase difference between the two superconductors.

In order to calculate the S matrix, one has to solve the Bogoliubov–de Gennes equation for each SF interface. Here we take a simpler approach and express the S matrix (\mathcal{R}) in terms of a S matrix of a SN interface [2,15], where N stands for a normal-nonferromagnetic layer. By so doing, we neglect the difference in the number of spin-up and -down modes in the ferromagnet. This approximation is also assumed in the quasiclassical approach and is justified for a weak exchange field in the ferromagnet [17].

The S matrix of a transparent SN interface reads

$$\mathcal{R}_{\mathcal{A}}(\varepsilon, \phi) = \alpha(\varepsilon) \begin{pmatrix} 0 & i\sigma_2 e^{i\phi} \\ -i\sigma_2 e^{-i\phi} & 0 \end{pmatrix}, \quad (2)$$

where $\alpha(\varepsilon) = e^{-i \arccos(\varepsilon/\Delta)}$ and σ_2 is the second Pauli matrix. For the ferromagnetic spacer layer (spin-active region), the S matrix is

$$S_F = \begin{pmatrix} 0 & U \\ U & 0 \end{pmatrix}, \quad U = e^{i(\eta + \rho \mathbf{m} \cdot \boldsymbol{\sigma})/2}, \quad (3)$$

where $\mathbf{m} = (\sin\theta \cos\psi, \sin\theta \sin\psi, \cos\theta)$ is the magnetization orientation in the ferromagnetic spacer layer. The difference of the phase shifts of spin-up and spin-down electrons upon propagating through the ferromagnetic spacer layer is denoted by $\rho = \nu_{\uparrow} - \nu_{\downarrow}$, while $\eta = \nu_{\uparrow} + \nu_{\downarrow}$. Here the spin-up (spin-down) is defined with respect to the magnetization axis in the F layer. These phases depend on the orbital channel index μ , but for the sake of notational simplicity we have suppressed the index. We also assume that the ferromagnetic spacer layer thickness is much smaller than the superconducting coherence length ($L' \ll \xi_S$), so that the energy dependence of ρ and η can be ignored. Combining these scattering matrices we obtain the scattering matrix of the SF interface with the spin-active region

$$\mathcal{R}(\varepsilon) = \alpha(\varepsilon) \begin{pmatrix} 0 & -\hat{r}_{\text{he}}^* e^{i\phi} \\ \hat{r}_{\text{he}} e^{-i\phi} & 0 \end{pmatrix}, \quad (4)$$

with

$$\hat{r}_{\text{he}} = \begin{pmatrix} -ie^{i\psi} \sin\theta \sin\rho & -\cos\rho + i \cos\theta \sin\rho \\ \cos\rho + i \cos\theta \sin\rho & ie^{-i\psi} \sin\theta \sin\rho \end{pmatrix}. \quad (5)$$

For the SF interface without the spin-active region, we also include the phases acquired in the F layer and obtain

$$\mathcal{R}'(\varepsilon) = T(\varepsilon) \mathcal{R}_{\mathcal{A}}(\varepsilon, 0) T(\varepsilon), \quad (6)$$

with $T = e^{i \text{diag}[k_{\mu,\uparrow}(\varepsilon), k_{\mu,\downarrow}(\varepsilon), -k_{\mu,\uparrow}(-\varepsilon), -k_{\mu,\downarrow}(-\varepsilon)]L}$; $k_{\mu,\uparrow\downarrow}$ is the longitudinal component of the wave vector in the orbital mode μ (spin-up or -down), and L is the F layer thickness.

In order to perform the integration over orbital modes, we put $\rho_{\mu} = Z'/\cos\Theta$, where $Z' = 2h'L'/(\hbar v_F)$; h' is the exchange energy in the spacer layer which is assumed to be small ($h' \ll E_F$), and Θ is the angle between the excitation velocity and the junction axis. Also, in the Andreev approximation $k_{\mu,\uparrow\downarrow}(\varepsilon) = (k_F \pm h/\hbar v_F + \varepsilon/\hbar v_F)/\cos\Theta$, where h is the exchange energy in the F layer ($h \ll E_F$). Setting $L' = 0$ (no spin-active layer) recovers the result previously obtained from the quasiclassical approach [11], where the following expression relates the scattering to the quasiclassical approach (see also Ref. [18]):

$$\sum_{\sigma} \frac{g_{\sigma}(\Theta) - g_{\sigma}(-\Theta)}{2} = \frac{e^2 R_N}{i\pi\hbar} \frac{d}{d\phi} \ln \det(1 - \mathcal{R}\mathcal{R}'), \quad (7)$$

where R_N is the normal resistance and g_σ is the normal Green function in the ferromagnet [11].

In the general case—with one spin-active region—upon inserting Eqs. (4) and (6) into Eq. (1) the formula for the Josephson current is obtained. We introduce a new variable $u = \operatorname{arcsinh}(\omega/\Delta)$, and integrate over orbital modes

$$I = \frac{2\pi T}{eR_N} \sum_{\sigma, \omega_n > 0} \int_0^{\pi/2} d\Theta \sin\Theta \cos\Theta \operatorname{Im} \tanh \left[u + \Delta \sinh u \frac{L + L'}{\hbar v_F \cos\Theta} + i\sigma \frac{\chi}{2} + i \frac{\varphi}{2} \right], \quad (8)$$

with $\chi = \arccos(\operatorname{Re}[r_{\text{he}}^{\uparrow\downarrow} e^{-iz/\cos\Theta}])$ and $Z = 2hL/(\hbar v_F)$; $r_{\text{he}}^{\uparrow\downarrow}$ denotes the element (2,1) of the matrix in Eq. (5).

We now consider a long *SFS* junction, and show that only even harmonics in the CPR are long-range. In this case ($L \gg \xi_S$), the first term in the argument of the hyperbolic tangent (u) can be neglected. At zero temperature, the summation over ω_n can be replaced by an integration; the expression for the Josephson current reads

$$I = \frac{4\hbar v_F}{eR_N L} \sum_{k=1}^{\infty} (-1)^k I_k \sin(k\phi). \quad (9)$$

In the last equation, the spectral weights I_k quantify the contribution to the current coming from the phase coherent transport of k Cooper pairs across the barrier

$$I_k = - \int_1^{\infty} \frac{T_k(\operatorname{Re}[r_{\text{he}}^{\uparrow\downarrow} e^{-izx}])/k}{x^4} dx, \quad (10)$$

where T_k is the Chebyshev polynomial of the first kind and $x = 1/\cos\Theta$. We have used the identity $\operatorname{Im} \tanh z = 2 \sum_{k=1}^{\infty} (-1)^k \operatorname{Im} e^{-2kz}$ for obtaining Eqs. (9) and (10) [19].

In order to avoid cumbersome expressions, we concentrate on the case with mutually orthogonal magnetizations in the *F* and the ferromagnetic spacer layer ($\theta = \pi/2$). Later we will show that all our conclusions are valid for arbitrary (but not too small) θ . Now, $r_{\text{he}}^{\uparrow\downarrow} = \cos(Z'x)$ and the expression for the spectral weights reads

$$I_k = \int_1^{\infty} \frac{dx}{x^4} \sum_{n=0}^{\lfloor k/2 \rfloor} \sum_{m=0}^n \frac{(-1)^{n+m+1}}{k} \binom{k}{2n} \binom{n}{m} [\cos(Zx) \times \cos(Z'x)]^{k-2(n-m)}, \quad (11)$$

where $\lfloor x \rfloor$ denote the largest integer not greater than x .

We perform the integration in Eq. (11) by expanding the integrand as a sum of cosines. When k is odd, every term in that sum depends on Z ; for a long *SFS* junction and for a reasonably strong exchange energy ($h > \Delta$), we have $Z \gg 1$ and $\int_1^{\infty} \cos(Zx)/x^4 dx = -\sin Z/Z + O(1/Z^2)$. Hence, odd harmonics are suppressed by the factor $1/Z$. On the other hand, for even k we find terms that are independent of Z (the exchange energy in the *F* layer). Thus we write $I_{2k} = I_{2k}^{\text{LR}} + O(1/Z)$. The long-range component of even harmonics (I_{2k}^{LR}) is given by

$$I_{2k}^{\text{LR}} = \int_1^{\infty} \frac{dx}{x^4} \sum_{n=0}^k \sum_{m=0}^n \frac{(-1)^{n+m+1}}{2^{2(k-n+m)}} \frac{1}{2k} \binom{2(k-n+m)}{k-n+m} \times \binom{2k}{2n} \binom{n}{m} \cos(Z'x)^{2(k-n+m)}. \quad (12)$$

This result shows that even harmonics dominate in a long junction. It should be noted that even harmonics can dominate for any junction length if the parameters of the spin-active region are chosen in such a way that normal Andreev reflection vanishes on one *SF* interface (i.e., $r_{\text{he}}^{\uparrow\downarrow} = 0$). This can be directly seen from Eqs. (9) and (10).

Figure 2 shows the dependence of the first two long-range harmonics (I_2 and I_4) on Z' . For large values of Z' both curves converge to the constant values— \bar{I}_2 and \bar{I}_4 , respectively. Hence, we see that for a certain range of the ferromagnetic spacer layer parameters [$1 \ll L'h'/(\hbar v_F) \ll h'/\Delta$] the Josephson current reads

$$I = \frac{4\hbar v_F}{eR_N L} \sum_{k=1}^{\infty} \bar{I}_{2k}^{\text{LR}} \sin(2k\phi) + O\left(\frac{\hbar v_F}{hL}, \frac{\hbar v_F}{h'L'}\right), \quad (13)$$

where \bar{I}_{2k}^{LR} are independent of the junction parameters

$$\bar{I}_{2k}^{\text{LR}} = \sum_{n=0}^k \sum_{m=0}^n \frac{(-1)^{n+m+1}}{2^{4(k-n+m)}} \frac{1}{6k} \binom{2(k-n+m)}{k-n+m}^2 \times \binom{2k}{2n} \binom{n}{m}. \quad (14)$$

Equations (13) and (14) assert that for a long *SFS* junction with one spin-active region, the long-range part of the Josephson current depends only on the Thouless energy. Consequently, the long-range part of the current is not suppressed by fluctuations of the ferromagnetic barrier thickness (interface roughness). The current-phase relation is depicted in the inset of Fig. 3.

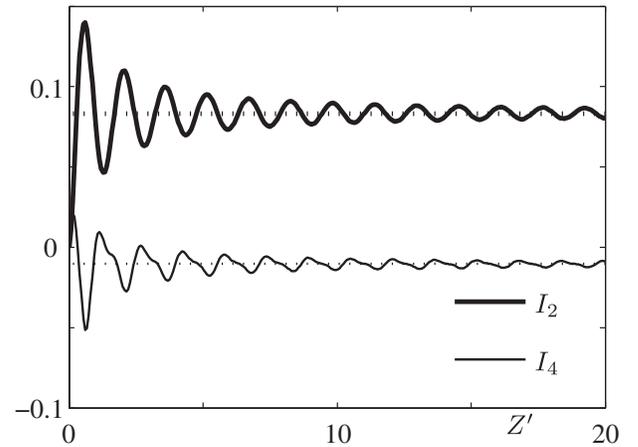


FIG. 2. The long-range component of the second (thick solid curve) and the fourth (thin solid curve) harmonic of the Josephson current-phase relation. For $Z' \gg 1$, both the curves converge to constant values (dotted lines), \bar{I}_2 and \bar{I}_4 .

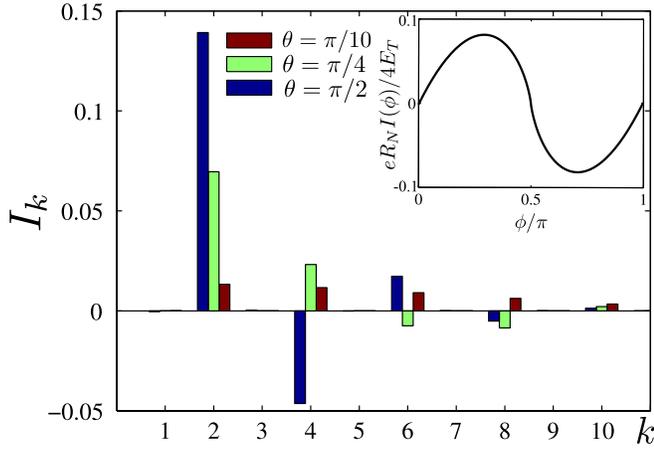


FIG. 3 (color online). The spectral weights of the current-phase relation for $Z' = 0.5$ and $Z = 1000$, and for three values of the relative angle between magnetizations: $\theta = \pi/10$, $\pi/4$, $\pi/2$. The inset depicts the generic CPR given by Eq. (13). The Josephson current is normalized by $eR_N/(4E_T)$, where $E_T = \hbar v_F/L$ is the Thouless energy.

In the general case, when the value of Z' is arbitrary (but $L' \ll \xi_S$), the supercurrent dependence on Z' is given by Eq. (12). Again, even harmonics are dominant. The free energy of the Josephson junction is given by $F(\phi) \propto \int_0^\phi I(\tilde{\phi}) d\tilde{\phi}$; the ground state of the junction is degenerate: the 0 and π states have the same energy. The degeneracy is lifted only by algebraically small factors ($\sim 1/Z$).

The dependence of the spectral weights on the misorientation angle is depicted for three values of θ in Fig. 3. We observe that even harmonics dominate also for $\theta < \pi/2$. As the relative angle between magnetizations approaches zero, the amplitudes of even harmonics lessen and eventually the odd and even harmonics become comparable. Therefore, one can tune the ratio I_2/I_1 by changing the angle θ , while the sign of I_1 can be changed by adjusting the thickness of the F layer [2]. This enables robust realization of φ junctions [14], contrary to previously discussed realizations with the 0- π transition.

At finite temperatures a new length scale appears—the normal metal coherence length $\xi_N = \hbar v_F/(2\pi T)$. For high temperatures, $T \gg \hbar v_F/L = E_T$ (i.e., $L \gg \xi_N$), only the first term ($n = 0$) in the summation over Matsubara frequencies contributes to the current given by Eq. (8). After performing the integration over orbital modes, assuming $h \gg T$, we obtain

$$I = -\frac{8\pi T}{eR_N} \left[\frac{\sin(Z - Z')}{Z} \cos^2 \frac{\theta}{2} + \frac{\sin(Z + Z')}{Z} \sin^2 \frac{\theta}{2} \right] \times \frac{\Delta^2}{(\pi T + \sqrt{\Delta^2 + \pi^2 T^2})^2} e^{-L/\xi_N} \sin \phi. \quad (15)$$

We conclude that the first harmonic always dominates in the high-temperature limit, because the higher harmonics are suppressed by the factor $e^{-k\xi_N/L}$ ($k > 1$) [20]; in this case the supercurrent has only the short-range part [5].

In conclusion, we have shown that SFS junctions with one and two spin-active SF interfaces are qualitatively different. In the case of two spin-active interfaces, all harmonics in the Josephson current-phase relation are long-range (and the first one is dominant) [8], while in the case of one spin-active interface, we find that only even harmonics are long-range (and the second one is dominant). Some repercussions of the discovered effect are half-integer Shapiro steps [9], the coexistence of integer and half-integer fluxoid SQUID configurations [11], and robust realization of the φ junctions [14].

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