

Superharmonic Long-Range Triplet Current in a Diffusive Josephson Junction

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(Received 5 March 2013; published 21 May 2013)

We study the Josephson current through a long ferromagnetic bilayer in the diffusive regime. For noncollinear magnetizations, we find that the current-phase relation is dominated by its second harmonic, which corresponds to the long-range coherent propagation of two triplet pairs of electrons.

DOI: [10.1103/PhysRevLett.110.217004](https://doi.org/10.1103/PhysRevLett.110.217004)

PACS numbers: 74.45.+c, 74.20.Rp, 74.50.+r, 75.70.Cn

The interplay between ferromagnetism and superconductivity in hybrid structures is an active area of research [1,2]. In a homogeneous ferromagnet (F) adjacent to a conventional superconductor (S), superconducting correlations between pairs of electrons are induced in both the spin singlet channel and the triplet channel without spin projection along the magnetization axis. This proximity effect is short-ranged due to the dephasing of electrons with opposite spins induced by the ferromagnetic exchange field. On the other hand, in the presence of a noncollinear magnetic configuration, a long-range proximity effect can be induced in the triplet channels with parallel electron spins, as no such dephasing occurs in that case [3,4].

Early proposals to observe this effect suggested measuring the critical current in a Josephson junction through a ferromagnetic *trilayer* with noncollinear magnetizations [5,6]. In this geometry, the external F layers convert singlet Cooper pairs into triplet pairs which have a nonvanishing projection onto the channels with parallel electron spins along the (tilted) magnetization of the central F layer and, thus, may propagate coherently over long distances. A maximal critical current with amplitude comparable to that of a normal metallic Josephson junction with the same length is obtained when the external layers have a thickness comparable to the ferromagnetic coherence length ξ_F , and the magnetizations in successive layers are orthogonal. (Similarly, long-range triplet correlations can be induced by spin-active interfaces between a ferromagnet and the superconducting leads [7–9].) Indeed, recent experiments observed a strong enhancement of the Josephson current through a ferromagnetic multilayer when the layers were ordered noncollinearly [10–12].

Here we are interested in the question whether three layers (or, equivalently, spin-active interfaces on both sides of the junction) are necessary to observe a long-range triplet Josephson current. Recently, it has been shown theoretically that a long-range triplet proximity effect may also develop in ballistic *bilayer* ferromagnetic Josephson junctions with noncollinear magnetizations [15,16]. In this case, a superharmonic Josephson relation is generated by the long-range propagation of an even number of parallel-spin triplet pairs which may then recombine into singlet Cooper pairs. It is important to know whether this effect is

robust to disorder which is believed to be present in the experiments, i.e., whether it exists in diffusive systems as well. In this Letter, we show that this is indeed the case. We find that the amplitude of the critical current determined by the second harmonic of the Josephson relation—corresponding to the coherent transfer of two Cooper pairs—decays algebraically with length. The maximal current is smaller than in trilayers due to fact that two parallel-spin triplet pairs with opposite spin directions have to recombine into singlet Cooper pairs, a process that takes place only within a distance $\sim \xi_F$ near the F/S interface.

We consider a Josephson junction formed of two superconducting leads contacted through two ferromagnetic layers in series, with thicknesses d_L and d_R , respectively; cf. Fig. 1. The F layers are assumed to have the same properties but different orientations of their magnetizations. As we are interested in the long-range proximity effect, moreover, we assume that the right layer of length d_R is much thicker than the diffusive ferromagnetic coherence length $\xi_F = \sqrt{D/\hbar}$, where D is the diffusion constant and \hbar is the amplitude of the exchange field.

Within the quasiclassical diffusive theory [17], the equilibrium supercurrent flowing through the junction can be expressed through the quasiclassical Green function g , which is a 4×4 matrix in the particle-hole and spin spaces, and obeys normalization conditions $g^2 = 1$ and $\text{Tr}g = 0$. It solves the nonlinear Usadel equation

$$-D\partial_y(g\partial_y g) + [(\omega + i\hbar(y) \cdot \boldsymbol{\sigma})\tau_z, g] = 0, \quad (1)$$

where y is a coordinate along the junction, τ_i and σ_j ($i, j = x, y, z$) are Pauli matrices in the particle-hole and spin spaces, respectively, and $\omega = (2n + 1)\pi T$ (n integer) is a Matsubara frequency at temperature T . The orientations of the exchange fields are characterized by a tilt angle:

$$\hbar(y) = \hbar[\sin\theta(y)\hat{x} + \cos\theta(y)\hat{z}], \quad (2)$$

where $\theta(y) = \theta$ for $-d_L < y < 0$ and $\theta(y) = 0$ for $0 < y < d_R$. Note that the orbital effect is neglected in Eq. (1) by assuming that the magnetic flux through the junction is much smaller than the flux quantum.

Equation (1) has to be supplemented by boundary conditions at each interface. We assume that the interface

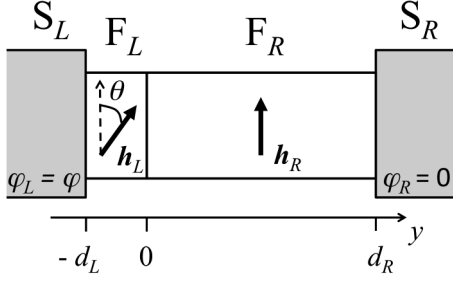


FIG. 1. Setup of the junction. The superconducting leads are coupled through two ferromagnetic layers in series, with thicknesses d_L and d_R , respectively. The magnetizations of the layers are tilted with respect to each other by an angle θ .

resistance between the two F layers is much smaller than the resistance of each layer. Therefore, g and its derivative are continuous at $y = 0$. Moreover, assuming a rigid boundary condition at the interfaces with the leads as well as negligible resistances of the F/S interfaces, we impose the continuity conditions $g(-d_L) = g_L$ and $g(d_R) = g_R$, where $g_k = [\omega\tau_z + \Delta(\cos\varphi_k\tau_x - \sin\varphi_k\tau_y)]/\sqrt{\omega^2 + \Delta^2}$ ($k = L, R$) are the Green functions of the leads. Here Δ is the amplitude of the superconducting gap and φ_k its phase. For convenience, we choose $\varphi_L = \varphi$ and $\varphi_R = 0$ such that φ denotes the superconducting phase difference.

The supercurrent is then related to the Green function g through

$$I = \pi e \nu D A T \sum_{\omega > 0} \text{ImTr}[\tau_z g \partial_y g], \quad (3)$$

where ν is the density of states (per spin) at the Fermi level and A is the cross section of the junction. Because of current conservation, Eq. (3) may be evaluated at any position along the junction.

To proceed further, we introduce the parametrization

$$g = \begin{pmatrix} \sqrt{1 - F\tilde{F}} & F \\ \tilde{F} & -\sqrt{1 - \tilde{F}F} \end{pmatrix}, \quad (4)$$

which automatically satisfies the normalization conditions. Here, the anomalous functions F and $\tilde{F} = \sigma_y F^* \sigma_y$ are 2×2 matrices in spin space and odd in Δ . With the parametrization (4), the Usadel equation (1) takes the form

$$-D\partial_y \left[\sqrt{1 - F\tilde{F}} \partial_y F - F \partial_y \sqrt{1 - \tilde{F}F} \right] + \{\omega + i\mathbf{h}(y) \cdot \boldsymbol{\sigma}, F\} = 0, \quad (5)$$

while the current (3) simplifies to

$$I = 2\pi e \nu D A T \sum_{\omega > 0} \text{ImTr}[F \partial_y \tilde{F}]. \quad (6)$$

Note that, as F and \tilde{F} are odd functions of Δ , the current is even in Δ .

At temperatures slightly below the superconducting critical temperature T_c , the gap vanishes as $\Delta \propto \sqrt{T_c(T_c - T)}$.

Thus, one may solve Eq. (5) perturbatively around the normal state solution, $F = 0$. To this end, we expand F in the small parameter [18] Δ/ω , i.e., $F = (\Delta/\omega)F^{(1)} + (\Delta/\omega)^3 F^{(3)} + \dots$, and solve Eq. (5) order by order. To obtain the current up to the fourth order in Δ/T_c , it is sufficient to compute $F^{(1)}$ and $F^{(3)}$.

Let us start with the leading order. Then, Eq. (5) yields the linear differential equation

$$-D\partial_y^2 F^{(1)} + 2\omega F^{(1)} + i\{\mathbf{h}(y) \cdot \boldsymbol{\sigma}, F^{(1)}\} = 0. \quad (7)$$

Upon performing a unitary transformation, $\mathcal{F}^{(1)}(y) = e^{i\sigma_y \theta(y)/2} F^{(1)}(y) e^{-i\sigma_y \theta(y)/2}$, the general solution in each layer takes the form $\mathcal{F}^{(1)} = \mathcal{F}_0^{(1)} + \mathcal{F}_x^{(1)} \sigma_x + \mathcal{F}_z^{(1)} \sigma_z$, where

$$\mathcal{F}_{L/R,s}^{(1)} = A_s^{L/R} e^{-p_s y} + B_s^{L/R} e^{p_s y} \quad (8)$$

with $s = \pm$, x and $\mathcal{F}_{\pm}^{(1)} = \mathcal{F}_0^{(1)} \pm \mathcal{F}_z^{(1)}$. Here $\mathcal{F}_{\pm}^{(1)}$ with $p_{\pm} = \sqrt{2(\omega \pm ih)/D}$ correspond to the short-range singlet and triplet correlations, while $\mathcal{F}_x^{(1)}$ with $p_x = \sqrt{2\omega/D}$ corresponds to an equal superposition of the long-range triplet correlations. For typical ferromagnets, $h \gg T_c$ and, therefore, $p_{\pm} \approx (1 \pm i)/\xi_F$.

The coefficients $A_s^{L/R}$ and $B_s^{L/R}$ that enter Eq. (8) are determined by the boundary conditions. Assuming $d_R \gg \xi_F$, we find $A_{\pm}^R = e^{i\varphi} a_{\pm}$, $B_{\pm}^R = e^{-p_{\pm} d_R}$, and $A_x^R e^{-p_x d_R} = -B_x^R e^{p_x d_R} = i e^{i\varphi} a_x$ (up to exponentially small corrections in d_R/ξ_F) with

$$a_+ = a_-^* = \frac{\alpha - \beta}{|\alpha|^2 - |\beta|^2}, \quad (9a)$$

$$a_x = \frac{\tan\theta}{2 \sinh p_x d} \text{Im}[(p_+ d_L + 1)a_+], \quad (9b)$$

and

$$\alpha = \cos^2 \frac{\theta}{2} e^{p_- d_L} + \frac{\sin^2 \theta}{2 \cos \theta} (p_- d_L + 1) \frac{\cosh p_- d_L \sinh p_x d_R}{\sinh p_x d}, \quad (10a)$$

$$\beta = \sin^2 \frac{\theta}{2} (\cosh p_+ d_L - i \sinh p_+ d_L) - \frac{\sin^2 \theta}{2 \cos \theta} (p_- d_L + 1) \frac{\cosh p_+ d_L \sinh p_x d_R}{\sinh p_x d}, \quad (10b)$$

where $d = d_L + d_R$ is the total length of the junction. We do not give the explicit results in the left layer, because, in the following, we choose to evaluate the current (6) in the right layer.

Note that the ratio d_L/ξ_F may be arbitrary, while Eqs. (9) and (10) have been simplified with the assumption $d_L \ll \xi_N$, where $\xi_N = \sqrt{D/(2\pi T_c)}$ ($\xi_N \gg \xi_F$) is the normal coherence length close to T_c . For a noncollinear configuration of the layers, long-range correlations are present ($\mathcal{F}_x^{(1)} \neq 0$). Nevertheless, they do not contribute to the first harmonic of the current-phase relation, as we show now.

Inserting the solution (8) for $\mathcal{F}_R^{(1)}$ into Eq. (6), we obtain the first harmonic I_1 of the current-phase relation, $I_1(\varphi) = I_1 \sin\varphi$, where $I_1 \propto \text{Re}[p_+ e^{-p_+ d_R} a_+]$. It takes the form

$$I_1 = \frac{\pi G \Delta^2}{\sqrt{2} e T_c} \frac{d}{\xi_F} e^{-d/\xi_F} Y_1\left(\frac{d_L}{\xi_F}, \frac{d_R}{\xi_F}, \theta\right), \quad (11)$$

where $G = 2e^2 \nu DA/d$ is the conductance of the junction in the normal state and Y_1 is a scaling function.

The exponential reduction factor e^{-d/ξ_F} in Eq. (11) shows that I_1 is always short-ranged. The scaling function Y_1 is shown in Fig. 2 as a function of the length of the left ferromagnet and the tilt angle between the magnetizations. Its maximal absolute value is of the order of 1 and its sign oscillates, thereby displaying transitions between 0 states (when $I_1 > 0$) and π states (when $I_1 < 0$). Such oscillations have indeed been observed in $S/F/S$ junctions [19–21].

Simple analytic expressions for the function Y_1 in Eq. (11) can be obtained in various regimes. For a parallel alignment, $\theta = 0$, the results for a long ($d \gg \xi_F$) monodomain $S/F/S$ junction are reproduced with $Y_1 \simeq \sin(d/\xi_F + \pi/4)$ [22]. Moreover, as long as $d_L \ll \xi_F$, this result remains valid for arbitrary angles. In the opposite limit, $d_L \gg \xi_F$, we find $Y_1 \simeq \sqrt{2} \cos(d_L/\xi_F) \cos(d_R/\xi_F)$ for $\delta\theta < \theta < \pi - \delta\theta$ and $Y_1 \simeq \sqrt{2} \cos[(d_L - d_R)/\xi_F]$ for an antiparallel alignment, $\theta = \pi$. Here $\delta\theta \sim \xi_F / \min(d_L, d_R)$ is the crossover scale over which Y_1 varies smoothly between its respective values at $\theta = 0$, at intermediate angles, and at π . These results generalize those of Refs. [23–25] obtained at $d_L = d_R$ [26].

We now turn to higher-order contributions in Δ/T_c to the current-phase relation. The fourth-order terms consist in a small (short-range) correction to the amplitude I_1 of the first harmonic, as well as a second harmonic $\propto \sin(2\varphi)$. Here we concentrate on the second harmonic. In collinear structures, its amplitude I_2 is short-ranged with a suppression factor e^{-2d/ξ_F} . Thus, it becomes important only near

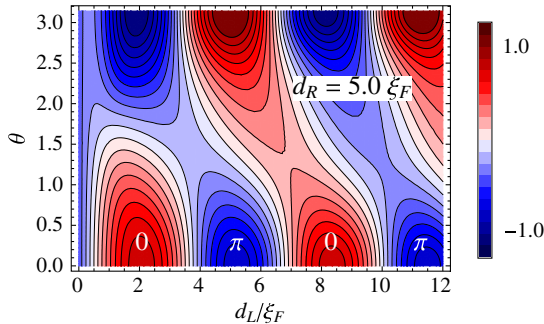


FIG. 2 (color online). Dependence of the short-range first harmonic I_1 of the current-phase relation on the thickness d_L/ξ_F and the angle θ at fixed $d_R/\xi_F = 5$. Here I_1 is measured in units of $I_{10} = \pi G \Delta^2 d / (\sqrt{2} e T_c \xi_F) e^{-d/\xi_F}$.

$0/\pi$ transitions [29–31] when I_1 vanishes. In noncollinear structures, however, I_2 is long-ranged as we show now.

For concreteness, we will assume $d_R \gg d_L$ and evaluate the current (6) in the right layer. Long-range contributions originate from the propagation of parallel-spin triplet pairs described by F_x , namely, $I_2^r \propto \text{ImTr}[F_{R,x}^{(1)} \partial_y \tilde{F}_{R,x}^{(3)} + F_{R,x}^{(3)} \partial_y \tilde{F}_{R,x}^{(1)}]$. Using Eq. (5), we find that $F_{R,x}^{(3)}$ obeys the equation

$$-D \partial_y^2 F_{R,x}^{(3)} + 2\omega F_{R,x}^{(3)} = \frac{D}{2} \partial_y \{ [(F_{R,x}^{(1)})^2 + F_{R,+}^{(1)} F_{R,-}^{(1)}] \partial_y \tilde{F}_{R,x}^{(1)} + [F_{R,+}^{(1)} \partial_y \tilde{F}_{R,+}^{(1)} + F_{R,-}^{(1)} \partial_y \tilde{F}_{R,-}^{(1)}] F_{R,x}^{(1)} \}. \quad (12)$$

Being interested in the second harmonic, we need only to retain contributions to $F_{R,x}^{(3)}$ that are proportional to $e^{-i\varphi}$. Thus, we may decompose $F_{R,x}^{(3)} = f_x^{(3)} e^{-i\varphi} + \Phi_{R,x}^{(3)}$, where $\Phi_{R,x}^{(3)}$ does not contain any contributions $\propto e^{-i\varphi}$. The function $f_x^{(3)}$ then obeys a differential equation obtained from Eq. (12) by keeping on the right-hand side the terms $\propto e^{-i\varphi}$ only. In particular, such terms arise from the contribution $F_{R,+}^{(1)} F_{R,-}^{(1)} \partial_y \tilde{F}_{R,x}^{(1)}$ yielding

$$-D \partial_y^2 f_x^{(3)} + 2\omega f_x^{(3)} = -\frac{2Dq}{\xi_F} \frac{\Delta^3}{\omega^3} a_x e^{2(y-d_R)/\xi_F}, \quad (13)$$

where we used that $\xi_F \ll \xi_N$. Note that the source term $F_{R,+}^{(1)} F_{R,-}^{(1)} \partial_y \tilde{F}_{R,x}^{(1)}$ involves the short-range correlations $F_{R,\pm}^{(1)}$ existing close to the F/S interface; thus, $f_x^{(3)}$ contains the conversion between short-range and long-range pairs. A general solution that satisfies Eq. (13) together with the boundary condition $f_x^{(3)}(d_R) = 0$ at the right F/S interface reads

$$f_x^{(3)} = \frac{p_x \xi_F}{2} \frac{\Delta^3}{\omega^3} a_x \{ e^{2(y-d_R)/\xi_F} - \cosh p_x (d_R - y) \} + C \sinh p_x (d_R - y). \quad (14)$$

Here C is a constant which should be determined from the boundary condition at the interface between the left and right ferromagnets. It turns out, however, that it does not contribute to the current and, thus, will not need to be determined. We obtain

$$I_2 = 4\pi e \nu D A T \sum_{\omega > 0} \frac{\Delta^4}{\omega^4} p_x^2 \xi_F a_x^2, \quad (15)$$

which evaluates to

$$I_2 = \frac{\pi G \Delta^4}{192 e T_c^3} \frac{\xi_F}{d_R} Y_2\left(\frac{d_L}{\xi_F}, \theta\right) \times \begin{cases} 1, & d_R \ll \xi_N, \\ \frac{384 d_R^2}{\pi^4 \xi_N^2} e^{-2d_R/\xi_N}, & d_R \gg \xi_N. \end{cases} \quad (16)$$

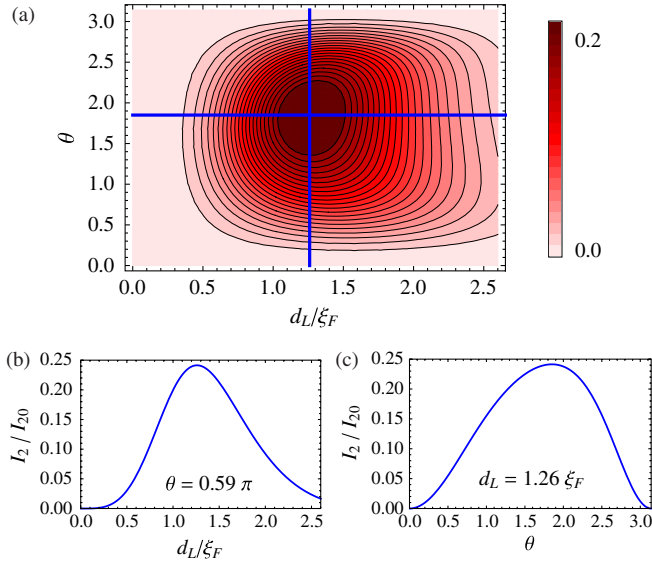


FIG. 3 (color online). Dependence of the long-range second harmonic I_2 of the current-phase relation on the thickness d_L/ξ_F and the angle θ . Here I_2 is measured in units of $I_{20} = \pi G \Delta^4 \xi_F / (192 e T_c^3 d_R)$. The plots (b) and (c) are taken along the cuts indicated by the blue lines in (a), at $\theta = 0.59\pi$ and $d_L/\xi_F = 1.26$, respectively. Both cuts include the maximal value $I_2^{\max} \approx 0.24 I_{20}$.

The second harmonic is long-ranged: It depends on d_R/ξ_F as a power law. The suppression factor ξ_F/d_R is due to the conversion of two parallel-spin triplet pairs into two singlet Cooper pairs which takes place on a distance ξ_F from the F/S interface only. The dependence of the function Y_2 that appears in Eq. (16) on the tilt angle θ and the thickness $d_L \ll d_R$ of the short layer is shown in Fig. 3. Y_2 is maximal for lengths $d_L \sim \xi_F$ and angles $\theta \sim \pi/2$. As expected, it vanishes for collinear structures, $\theta \rightarrow 0, \pi$ of arbitrary length. In noncollinear structures, it simplifies to $Y_2 \simeq (d_L/\xi_F)^4 \sin^2 \theta$ for a short F layer, $d_L \ll \xi_F$, while it vanishes exponentially

$$Y_2 = \frac{4 \sin^2 \theta (\cos \frac{d_L}{\xi_F} \cos^2 \frac{\theta}{2} - \sin \frac{d_L}{\xi_F})^2 e^{-2(d_L/\xi_F)}}{(\sin^2 \theta + a_\theta \frac{\xi_F}{d_L})^2} \quad (17)$$

for a long F layer, $d_L \gg \xi_F$. Here a_θ is relevant only in a vicinity $\delta\theta$ near $\theta = 0, \pi$, where $a_0 = 2$ and $a_\pi = 1$, respectively.

As in noncollinear structures the second harmonic I_2 is long-ranged (in contrast to the first harmonic I_1), we expect it to dominate as soon as d_R exceeds a few times ξ_F . In particular, for an optimal thickness $d_L \sim \xi_F$, we find the ratio $I_2/I_1 \sim (\Delta/T_c)^2 (\xi_F/d_R)^2 e^{d_R/\xi_F}$ close to T_c . The effect is expected to be robust at lower temperatures [32], further enhancing the ratio I_2/I_1 when $\Delta \sim T_c$. More generally, odd and even harmonics will be short-ranged and long-ranged, respectively. However, as triplet pairs need to recombine into singlet Cooper pairs on a distance ξ_F from

the F/S interface, we conjecture that the amplitude I_{2n} of the even harmonics $\propto \sin(2n\varphi)$ will contain a small factor $(\xi_F/d_R)^n$; see Eq. (16) for the case $n = 1$. Thus, the current-phase relation could be dominated by the second harmonic at all temperatures.

Let us now discuss the measurability of our prediction. The long-range proximity effect has been observed in trilayers [10,11], where already the first harmonic is long-ranged. The amplitude of the second harmonic for bilayers predicted in our work is smaller only by a factor ξ_F/d_R as compared to the first harmonic in trilayers. Thus we believe that it should be well within the sensitivity of present-day experiments. Its specific phase dependence may be detected by a direct measurement of the current-phase relation [33] or through the appearance of fractional Shapiro steps in the current-voltage characteristics under microwave irradiation [34].

Note that our prediction relies on the coherent propagation of parallel-spin triplet pairs with opposite spin directions. Therefore, I_2 should be proportional to $1 - P^2$, where P is the spin polarization of conduction electrons in the ferromagnet ($|P| \ll 1$ in the quasiclassical theory that we used). In particular, the effect should be robust in ferromagnets with intermediate spin polarization, while it would be suppressed in half-metals where $|P| \rightarrow 1$.

In conclusion, we predicted that the current-phase Josephson relation through a long diffusive ferromagnetic bilayer with noncollinear magnetizations is dominated by a superharmonic contribution $\propto \sin(2\varphi)$ when one of the layers has a thickness comparable to the ferromagnetic coherence length while the other layer is much thicker. The second harmonic can be viewed as the minimal Josephson current [35] that can flow between the conventional *even-frequency* superconductor in one lead and the effectively *odd-frequency* superconductor [2,36] generated by the long-range triplet proximity effect at the extremity of the ferromagnetic bilayer attached to the other lead. Measuring the dependence of the Josephson current on the thicknesses of the ferromagnetic layers and the angle between their magnetizations would provide further evidence for the long-range triplet proximity effect. Similar experimental studies have been performed for the critical temperature of a thin superconducting film in contact with a ferromagnetic spin valve [37]. Furthermore, detecting the π periodicity of the current-phase relation through phase-sensitive measurements would be a strong indication of the odd-frequency nature of the long-range proximity effect.

Part of this research was supported through ANR Grants No. ANR-11-JS04-003-01 and No. ANR-12-BS04-0016-03 and an EU-FP7 Marie Curie IRG.

Note added.—We recently learned of the experimental detection of a significant second harmonic in the Josephson relation through a ferromagnetic bilayer with intermediate length [38], which is possibly due to the effect discussed in our work.

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