Superharmonic Josephson relation at 0-/π-junction transition

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Critical current was recently measured near the transition from 0 to π-contact in superconductor/ferromagnet/superconductor Josephson junctions. Contrary to expectations, it does not vanish at the transition point. It shows instead a tiny, though finite, minimum. The observation of fractional Shapiro steps reenforces the idea that the vanishing of the main sinusoidal term in the Josephson relation gives room to the next harmonics. Within quasiclassical approach we calculate the Josephson relation taking into account magnetic scattering. We find that the observed minimum is compatible with the value of the second harmonics expected from the theory.

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According to textbooks equilibrium supercurrent, I, in a tunnel-barrier Josephson junction depends sinusoidally on the phase difference, ϕ, between the superconducting leads: $I = I_c \sin\phi$, where $I_c > 0$ is the so-called critical current of the junction. In his seminal work of late seventies Bulaevskii *et al* predicted [1] that the sign of I_c can change (or, equivalently, a shift of π appear in the argument of the sine) in the presence of magnetic impurities within the tunnel barrier. Soon after, Buzdin *et al.* [2, 3] suggested that such a junction, conventionally called now the π-junction, can be realized in a hybrid structure where the tunnel barrier is replaced with the ferromagnetic metal. While predicted theoretically, experimental realizations of π-junctions remained long unobserved. Indeed, superconductivity and magnetism compete; thus conventional ferromagnets would strongly suppress supercurrent. The first recent successful realization of a π-junction [4, 5] utilized the so-called *weak* ferromagnets, and the observation of a non-monotonic dependence of I_c as a function of the temperature [4, 6] and on the thickness of the ferromagnetic layer [5] served as the first evidences of the actual realization of π-junctions. Moreover in the former case, the existence of the temperature T^*, where I_c reached a minimum with the vanishing magnitude, allowed for a precise identification of the transition point.

These spectacular observations of π-junction behaviors remarkably confirmed original predictions of [1, 2, 3] and yet posed new puzzling questions. The first was that the observed amplitude of the current in the junctions appeared two orders of magnitude smaller than that expected from the theory. H. Sellier *et al* [6] proposed that magnetic impurities in the ferromagnet could be the origin of this effect, and indeed it was recently shown [7] that magnetic impurities can lead to a noticeable reduction of the critical current if one assumes somewhat artificial uniaxial distribution of magnetic disorder. It remains however to understand the effect of a more realistic isotropic disorder distribution.

Another puzzle concerns the form of the phase-current relation at the transition point T^*. Quite generally, the phase relation has to be periodic in ϕ. This does not rule out the possibility of the second or even higher harmonics: $I = I_1 \sin\phi + I_2 \sin 2\phi + \ldots$, which indeed appear in Josephson junctions formed by point contacts, constrictions, or normal metals [8, 9]. However the amplitude of higher order components in the magnetic Josephson junctions was long considered too low for being observed. Note now that at T^* the coefficient of the first harmonics (I_1) vanishes and, therefore, the higher harmonics become dominant. In Ref. [12] the measured critical current (the maximum of the absolute value of the current-phase relation) does not vanish at T^*, but passes through a minimum. This fact, together with the observation of fractional Shapiro steps, indicates that the observed current is in fact the I_2 component. On the other hand no such component was detected in Ref. [13].

In this Letter we develop a theory enabling the quantitative derivation of the full current-phase relation in the regimes corresponding to actual experiments of Refs. [6, 12, 13]. For this purpose we generalize the approach of Ref.[7] which assumes uniaxial magnetic disorder, and solve the resulting equations numerically without restriction on the values of the parameters. Extracting the exchange field and magnetic scattering time from the published data on the temperature dependence of I_c, we estimate the expected magnitude of I_2. We find that the predicted values agree favorably with those observed in Ref. [12], while the expected magnitude of I_2 for the sample of Ref. [13] is too small to be observable. A powerful and microscopic approach to superconductivity in disordered metals is offered by the quasiclassical theory in a form described in [14, 15]. The theory can also describe ferromagnetism, by inclusion of an exchange field acting on conduction electrons. This was done for instance in Refs. [16, 17], where the spin-orbit coupling with impurities was also included. However, for the weak ferromagnet Cu_xNi_{1-x} used in experiments of [4, 6, 12, 13], the spin-orbit coupling is expected to play a minor role. The more important effect should come from the strong inhomogeneities of the magnetic field on both the microscopic- (magnetic impurities) and mesoscopic scales (randomly oriented magnetic domains). Our theory takes this effect into account.

The metallic ferromagnet is described by the following Hamiltonian:

$$H = \int d\boldsymbol{r} \sum_{ss'} \psi_s^\dagger \left[\left(-\frac{\boldsymbol{\nabla}^2}{2m} - \mu + U \right) \delta_{ss'} - \boldsymbol{h}.\boldsymbol{\sigma}_{ss'} \right] \psi_{s'}, \tag{1}$$

where $\psi_s(\boldsymbol{r})$ and $\psi_s^\dagger(\boldsymbol{r})$ are annihilation and creation operators for electrons having spin projection s along the $\hat{z}$ direction, m is the effective electron mass, and μ is the Fermi energy ($\hbar = 1$). The disorder potential $U(\boldsymbol{r})$ describes the interaction of electrons with nonmagnetic impurities and is characterized by the correlation function:

$$\overline{U(\boldsymbol{r})U(\boldsymbol{r'})} = \frac{1}{2\pi\nu\tau}\delta(\boldsymbol{r}-\boldsymbol{r'}), \tag{2}$$

where τ is the elastic mean free time and ν is the density of state at the Fermi level per spin. The upper bar stands for disorder averaging. The exchange field $\boldsymbol{h}(\boldsymbol{r})$ acting on the electron spins may originate, for instance, from contact interaction between conduction electrons and localized impurity spins. We do not consider the question of the microscopic origin of $\boldsymbol{h}(\boldsymbol{r})$, but restrict ourselves to setting its statistical properties only. Namely, we take its average to be spatially uniform: $\overline{\boldsymbol{h}(\boldsymbol{r})} = h\,\hat{z}$, with h proportional to the magnetization of the ferromagnet. The fluctuating part is characterized by correlation functions:

$$\overline{(h_\alpha(\boldsymbol{r}) - \overline{h}_\alpha)(h_\beta(\boldsymbol{r'}) - \overline{h}_\beta)} = \frac{1}{2\pi\nu\tau_m^\alpha}\delta_{\alpha\beta}\delta(\boldsymbol{r}-\boldsymbol{r'}), \tag{3}$$

for $\alpha, \beta = x, y, z$. Here τ_m^α characterizes mean free time due to magnetic impurities. In the following, we also assume rotational symmetry around $\hat{z}$, thus $\tau_m^x = \tau_m^y$.

In order to describe the proximity effect in the ferromagnet, it is convenient to introduce thermal Green's functions

$$\mathcal{G}_{ns,n's'}(\boldsymbol{r},\boldsymbol{r'},\tau) = -\langle T_\tau \Psi_{ns}(\boldsymbol{r},\tau)\Psi^\dagger_{n's'}(\boldsymbol{r'},0)\rangle \tag{4}$$

in the Nambu(n)-spin(s) space, where $\Psi_{1s} = \psi_s$ and $\Psi_{2s} = \psi^\dagger_{-s}$. The equation of motion for the Matsubara-transformed disorder-averaged Green's function, $\overline{\mathcal{G}}$, is derived from the Hamiltonian (1) and reads:

$$\left[i\omega_n - \left(-\frac{\boldsymbol{\nabla}^2}{2m} - \mu - h\sigma_z \right)\tau_z - \Sigma_1 - \Sigma_2 \right] \overline{\mathcal{G}} = \hat{1}. \tag{5}$$

Here, ω_n are Matsubara frequencies at temperature T. The self-energies Σ_1 and Σ_2 are due to nonmagnetic and magnetic disorder, respectively. Using Eqs. (2) and (3) we derive the self-consistent equations for Σ_1 and Σ_2:

$$\begin{aligned} \Sigma_1(\boldsymbol{r},\omega_n) &= (2\pi\nu\tau)^{-1}\tau_z\overline{\mathcal{G}}(\boldsymbol{r},\boldsymbol{r},\omega_n)\tau_z \\ \Sigma_2(\boldsymbol{r},\omega_n) &= \sum_{\alpha=x,y,z}(2\pi\nu\tau_m^\alpha)^{-1}\tau_z S_\alpha \overline{\mathcal{G}}(\boldsymbol{r},\boldsymbol{r},\omega_n) S_\alpha \tau_z\,. \end{aligned}$$

Here $\boldsymbol{S} = (\sigma_x, \sigma_y, \sigma_z\tau_z)$, σ_α and τ_α are Pauli matrices in spin and Nambu spaces, respectively.

Now we define the quasiclassical Green's function

$$g(\boldsymbol{r},\omega_n) = \frac{i}{\nu\pi}\tau_z\overline{\mathcal{G}}(\boldsymbol{r},\boldsymbol{r},\omega_n)\,, \tag{6}$$

which, in the diffusive limit, obeys the equation

$$-D\boldsymbol{\nabla}(g\boldsymbol{\nabla}g) + [\omega_n\tau_z - ih\tau_z\sigma_z + \sum_\alpha \frac{1}{2\tau_m^\alpha}S_\alpha g S_\alpha, g] = 0, \tag{7}$$

with D the diffusion coefficient, and the normalization condition $g^2 = 1$. Symmetry properties of the Hamiltonian further constrain the form of g. Specifically: (i-a) By the invariance under the rotation around the average magnetization axis $\hat{z}$, we find that g is block diagonal in spin space; we thus define the two matrices in Nambu space, g_+ and g_-, as the two non-vanishing upper and lower components, respectively. (i-b) From the invariance under the rotation over an angle π around the $\hat{x}$ (or $\hat{y}$) axis and simultaneously change of sign of h, we find $g_+(h) = \tau_z g_-(-h)\tau_z$. ($ii$) By time-reversal symmetry, we find $g_\pm(\boldsymbol{r},\omega_n) = -\tau_z g_\pm(\boldsymbol{r},-\omega_n)^\dagger\tau_z$. Finally the 16 correlation functions introduced in Eq. (4) are not independent, since the representation is redundant. This gives: (iv) $g_+(\boldsymbol{r},\omega_n) = -\tau_x g_-(\boldsymbol{r},\omega_n)^*\tau_x$.

Exploiting these properties, we parameterize the complete Green's function in terms of g_+, which in its turn is completely determined by complex functions, θ and η:

$$g_+ = \begin{pmatrix} \cos\theta & \sin\theta e^{i\eta} \\ \sin\theta e^{-i\eta} & -\cos\theta \end{pmatrix}. \tag{8}$$

Then, Eq. (7) yields:

$$0 = D\nabla(\sin^2\theta\nabla\eta) + \frac{2}{\tau_m^x}\sin\theta\sin\theta^*\sin(\eta-\eta^*) \tag{9a}$$

$$\begin{aligned} 0 = &-D\nabla^2\theta + D\cos\theta\sin\theta(\nabla\eta)^2 \\ &+2(\omega_n - ih)\sin\theta + \frac{2}{\tau_m^z}\sin\theta\cos\theta \\ &+\frac{2}{\tau_m^x}[\sin\theta\cos\theta^* + \cos\theta\sin\theta^*\cos(\eta-\eta^*)] \end{aligned} \tag{9b}$$

These equations constitute the main analytical result of our work. Note that in non ferromagnetic superconductors, symmetry properties (i-b) and (iv) for $h = 0$ imply that θ and η are real. Then Eqs. (9) only depend on the effective magnetic scattering time $1/\tau_m = 1/\tau_m^z + 2/\tau_m^x$,

model	order	h (meV)	$1/h\tau_m$	ρ (%)	RI_2 (nV)
	1	4.6	9.3	25	1.08
$\tau_m^\alpha = 3\tau_m$	2	49	1.1	8	0.25
(isotropic)	3	83	0.3	10	0.07
$\tau_m^z = \tau_m$	1	15	2.7	12	0.10
$\tau_m^x = \infty$	2	61	0.8	0	0.16
(uniaxial)	3	86	0.2	8	0.08

TABLE I: Parameters of the fit (shown in Fig. 1 for the isotropic case). We took for the sample $\Delta = 1.3$ meV, $D/L^2 = 1.13$ meV for $L = 17$ nm, T^*=1.1 K. $\rho = \sigma_f/(2\sigma_f + \gamma_b L)$ is the ratio of the barrier resistance to the total resistance R of the junction in its normal state.

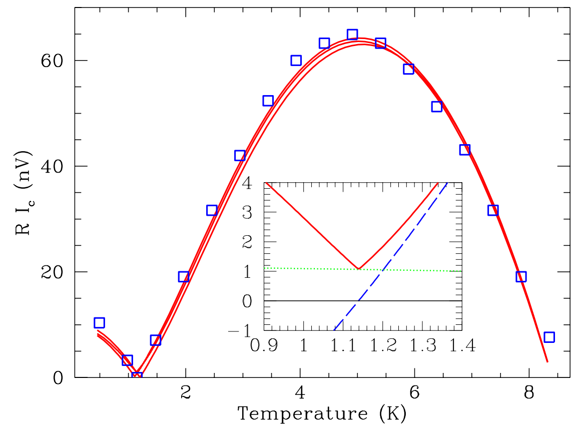

FIG. 1: Fit to experimental data for the critical current of Ref. [12] (boxes), for the three solutions reported in the table of the isotropic model. The three solutions gives nearly indistinguishable curves. Inset: temperature dependence near T^* of the calculated $I_c = \max_\phi(|I(\phi)|)$, I_1 (dashed), and I_2 (dotted), for the first solution. The minimum of I_c coincides with I_2.

in agreement with Abrikosov-Gor'kov theory for magnetic impurities [18]. By contrast, in ferromagnetic superconductors, magnetic disorder can be characterized by two scattering times: $\tau_m^x = \tau_m^y$ and τ_m^z [19, 20]. In Ref. [7] the uniaxial disorder was considered. In our notation this corresponds to $\tau_m^x = \tau_m^y = \infty$, $\tau_m^z = \tau_m$. This hypothesis simplifies greatly the solution of Eq. (9), since θ and η are no more coupled to θ^* and η^*. The physical reason for this simplification is that magnetic scattering does not couple the spin up and spin down populations. However it seems more realistic that the magnetic disorder is also able to flip the spin of conduction electrons. In this sense the opposite limit is to consider a completely isotropic disorder: $\tau_m^x = \tau_m^y = \tau_m^z = 3\tau_m$. In the following we thus concentrate on this case. We shall also discuss briefly the uniaxial one for comparison.

In order to determine the Josephson relation, we assume that the ferromagnet is a layer of the length L along the $\hat{x}$-axis. We thus need to solve Eqs. (9) with appropriate boundary conditions [21] at $x = \pm L/2$:

$$\sin\theta\nabla\eta = \mp\frac{\gamma_b}{\sigma_f}\frac{\Delta}{\sqrt{\omega_n^2+\Delta^2}}\sin(\eta\mp\frac{\phi}{2}), \tag{10a}$$

$$\nabla\theta = \mp\frac{\gamma_b}{\sigma_f}\frac{\omega_n\sin\theta - \Delta\cos\theta\cos(\eta\mp\frac{\phi}{2})}{\sqrt{\omega_n^2+\Delta^2}} \tag{10b}$$

Here, σ_f is the conductivity of the ferromagnet, and γ_b is the barrier resistance per unit area at the contacts (taken to be identical for simplicity) between the ferromagnet and the superconducting leads. We also assume that temperature dependence of the superconducting gap in the leads, Δ, is simply given by conventional BCS theory. The supercurrent is given by

$$I = \frac{2\pi G_f L T}{e}\sum_{\omega_n>0}\mathrm{Re}\left[\sin^2\theta\nabla\eta\right], \tag{11}$$

where G_f is the conductance of the ferromagnetic metal.

We use now the above equations for fitting the experimental data of Ref. [12, 13]. For a given set of parameters, namely the Thouless energy D/L^2, $\Delta(T=0)$, T, γ_b, h, τ_m, we obtain the current-phase relation by solving numerically the system of differential equations (9) and calculating the current through Eq. (11). One can then extract the first two harmonics, I_1 and I_2. (Higher harmonics near T^* are much smaller than I_2). We begin with the data of Ref. [12], Fig. 2, concerning a sample with a ferromagnetic layer of $L = 17$ nm. The length L, the superconducting gap, and the temperature are known experimentally. The interface resistance is more difficult to measure. The authors of Ref. [12] give an estimate of 30% of the total resistance of the junction in its normal state, R [6].

For a given value of γ_b one can find the pairs of values (h, τ) that satisfy the two equations $I_1(h, \tau_m, T^*) = 0$ and $I_1(h, \tau_m, T_1) = I_1^{exp}$, where T_1 is a temperature different from T^* and I_1^{exp} is the corresponding experimental value for I_1. We find that only three pairs of values satisfy this constraint. We order them by increasing value of h. One can show that the n-th solution refers to a junction where, as a function of the length of the sample, other $n-1$ zeros are predicted for $L < 17$ nm. We optimize then this first estimate of the parameters by including γ_b as a fitting parameter for the full experimental curve. The solutions are given in Table I. For comparison fitting parameters for uniaxial magnetic disorder are also given.

In all cases we have a good fit to data (see Fig. 1). Thus, the quality of the fit is not a sufficient criterium to discriminate between the three possibilities (for each model). One argument in favor of the first solution (for isotropic model of magnetic disorder) is the agreement of the fitting parameter γ_b with the estimated value in the experiment. A second one is the dependence of T^* on

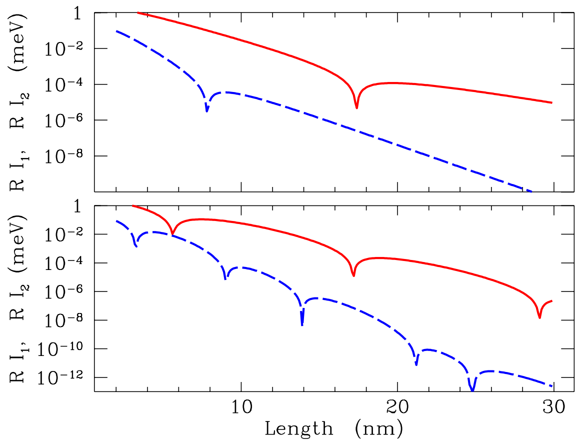

FIG. 2: Length dependence at $T = 4.2$ K of I_c (full line) and I_2 (dashed line) for the parameters obtained with the fit with the isotropic model. We show the solution 1 (upper panel) and 2 (lower panel). The zero at $L = 17$ nm is visible in both cases. Solution 2 displays a second zero for $L < 17$ nm.

the length that increases with the order of the solution. The range of L for which $0 < T^* < T_c$ is about 1 nm for the first solution, and about $0.4 - 0.3$ nm for the second and third one. The first case compares better with the experiment [12], where incertitude on L is about 1 nm, and π-contacts were observed for $L = 17 - 19$ nm [6].

We consider now the second component, I_2.

In Ref. [12] the minimum value of RI_2 is 0.5 nV, and it falls between the first and the second predicted value for isotropic model (cf. Tab. I). In both cases we thus find that the amplitude of the second harmonics is compatible with the observed one. We also find a strong temperature dependence of I_2, the values presented in the table at $T^* = 1.1$ K, are reduced by a factor 10 at 5 K. This dependence can explain the much smaller value for RI_2 observed in the 19 nm sample of Ref. [12]. In comparison, the uniaxial model gives a much smaller value for $RI_2 \approx 0.1$ nV for all three solutions.

We repeated the fitting procedure on the data of Frolov *et al.* [13], where no second harmonics is observed at T^*. We found again that the data can be compatible with either a second or a first zero, but in both cases $RI_2 < 10^{-10}$ mV, thus below the observation threshold.

We finally discuss the length dependence of the first and second harmonics for the fitted values of the parameters (see Fig. 2). As anticipated, $I_1(L)$ displays an oscillating behavior. One can clearly see in Fig. 2 that I_1 for solution 1 and 2 vanishes once and twice, respectively, for $L \leq 17$ nm (solution 3 is not shown). A more unexpected result is the oscillatory behavior for $I_2(L)$, that shows a remarkable doubled periodicity with respect to $I_1(L)$: Between two zeros of the first harmonics we *always* observed two zeros of the second harmonics. This means that the sign of I_2 remains always positive when I_1 vanishes. Therefore we find that the transition from 0- to π-contact is always discontinuous. We cannot rule out, however, that I_2 may be negative at T^* in some other region of the parameters space. This would imply that the transition from 0 to π-contact is continuous as a function of the temperature [3].

In conclusion, we have presented a development of the quasiclassical theory of superconductivity taking into account magnetic scattering in the presence of an exchange field. We have used our model to extract the exchange field and scattering times from the temperature dependence of the critical current in superconductor/ferromagnet/superconductor junctions. With these parameters we have calculated the second harmonics at the vanishing value of the first component which agree favorably with the experimental findings.

Note added: after the completion of this work we became aware of related work by A. Buzdin [22] where I_2 is calculated near the critical temperature.

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