Double-reversal thickness dependence of critical current in superconductor-ferromagnet-superconductor Josephson junctions.

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We report the first experimental observation of the two-node thickness dependence of the critical current in Josephson junctions with a ferromagnetic interlayer. Vanishings of the critical current correspond to transitions into π-state and back into conventional 0-state. The experimental data allow to extract the superconducting order parameter oscillation period and the pair decay length in the ferromagnet. We develope a theoretical approach based on Usadel equations, which takes into account the spin-flip scattering. Results of numerical calculations are in good agreement with the experimental data.

One of the exciting topics in studying the coexistence of superconductivity (S) and ferromagnetism (F) is proximity-induced sign-reversal superconductivity in a ferromagnet close to an SF-interface \([1, 2]\). The superconducting order parameter does not simply decay into the ferromagnet but also oscillates. An undoubted evidence of sign-reversal spatial oscillations of the superconducting order parameter in a ferromagnet was the observation of the π-state in SFS Josephson junctions \([3, 4, 5, 6]\). ‘π-junctions’ \([3]\) are weakly coupled superconducting structures which demonstrate π-shift of the macroscopic phase difference in the ground state. The relation between the superconducting current \(I_s\) and the phase difference \(\varphi\) in a Josephson junction is described by a 2\(\pi\)-periodic function. In the simplest case of a tunnel barrier or a barrier, made of dirty normal metal, one finds \(I_s = I_c \sin \varphi\). The Josephson π-junction has an anomalous current-phase relation \(I_s = I_c \sin(\varphi + \pi) = -I_c \sin \varphi\), i.e. it is characterized (nominally) by the negative critical current \([4]\). Spatial oscillations of the superconducting order parameter in a ferromagnet close to an SF-interface was predicted in Ref. \([3]\). A physical origin of the oscillations is the exchange splitting of the spin-up and spin-down electron subbands in a ferromagnet. It was discussed in Refs. \([3, 5, 6, 7]\) that in order to observe manifestations of the transition into the π-state one should fabricate an SFS sandwich with the F-layer thickness \(d_F\) close to integer numbers of half-periods of the order parameter spatial oscillations \(\lambda_{ex}/2\). The period is \(\lambda_{ex} = 2\pi \xi_{F2}\), where the oscillation (or “imaginary”) length \(\xi_{F2}\) can be extracted from the complex coherence length \(\xi_F\) in a ferromagnet: \(I_c = I_c (\frac{1}{\xi_{F2}^2} + i \frac{1}{\xi_{F1}^2})\). In the simplest case the imaginary length \(\xi_{F2}\) and the order parameter decay length \(\xi_{F1}\) are equal \([2]\): \(\xi_{F1} = \xi_{F2} = \sqrt{D/E_{ex}}\), where \(D\) is the diffusion coefficient for electrons in a ferromagnet and \(E_{ex}\) is the exchange energy responsible for sign-reversal superconductivity in a ferromagnet. Temperature changes of the coherence length related to the thermal energy contribution to pair-breaking processes were introduced in Ref. \([3]\), in which temperature \(0 - \pi\)-transition was observed for the first time. The expressions for \(\xi_{F1}\) and \(\xi_{F2}\) are the following:

\[
\xi_{F1,2} = \sqrt{\frac{hD}{\sqrt{(\pi k_B T)^2 + E_{ex}^2}}} \pm \pi k_B T \approx \sqrt{\frac{hD}{E_{ex} (1 + \frac{\pi k_B T}{2E_{ex}})}}
\]

The latter approximation corresponds to the case \(E_{ex} \gg k_B T\), which is valid for experiments discussed below.

Detailed experimental studies of the critical current thickness dependence for \(Nb-Cu_{0.47}Ni_{0.53} - Nb\) Josephson junctions has been started by us in Ref. \([6]\). A very large decay of the critical current and its sharp reentrant behavior for thicknesses close to 23 nm have been observed. An analysis of the experimental data and their comparison with the model described below have shown that the observed deep minimum is probably the reverse transition from the π- into the 0-state at the F-layer thickness close to full oscillation period while the first node of the dependence has to be at the thickness of about 10 nm. Thus, the presented work is devoted to finding of the two-node behavior of the SFS junction critical current as well as to discussion of mechanisms of the strong order parameter decay in a ferromagnetic CuNi interlayer.

In fact a nonmonotonic \(I_c(d_F)\) dependence close to \(0 - \pi\)-transition was observed for the first time in Ref. \([3]\) and has been presented there as a number of \(I_c(T)\) curves for different thicknesses \(d_F\). Later Kontos et al \([7]\) for \(Nb-Pd_{0.9}Ni_{0.1} - Nb\) and then Sellier et al \([8]\) for \(Nb-Cu_{0.52}Ni_{0.48} - Nb\) junctions measured detailed reentrant \(I_c(d_F)\) curves for F-interlayer thicknesses close to \(0 - \pi\)-transition. In this work we have investigated the thickness dependence of the SFS junction critical current density in a wide thickness range for sandwiches fabricated as described in Ref. \([6]\). All junctions had their lateral sizes smaller than the Josephson length and uni-
form current distribution. To do this the junctions with F-layer thicknesses of less than 17 nm were made with the contact area $10 \times 10 \times 10^{-6}$ and all the rest had the area $50 \times 50 \times 10^{-6}$. Weakly-ferromagnetic $Cu_{0.47}Ni_{0.53}$-interlayers had the Curie temperature of about 60 K. In the thickness interval $8 \sim 28$ nm we had about 6 orders of the critical current density change with vanishings at two $d_F$ values as it is presented in Fig. 1. One can see that undoubtly the curve demonstrates both direct $\pi$-transition and reverse transition from $\pi$- to 0-state. In transition points the critical current $I_c(d_F)$ is equal to zero and then should formally change its sign. Since in real experiments we could measure only magnitude of the critical current, the dependence $I_c(d_F)$ between two sharp cusps is the negative (corresponding to the $\pi$-state) branch of the curve which is reflected into the positive region. Due to slight temperature dependence of the order parameter oscillation period in our weak ferromagnet (described by $\xi_F$) we could pass through the transition points using samples with critical F-layer thicknesses 11 nm and 22 nm by means of temperature decrease. Temperature $0 \sim \pi$- and $\pi \sim 0$-transitions are presented in middle panels of Fig. 1. Upper and lower panels show the critical current temperature behavior for samples with close F-layer thicknesses. One can see that we lost a possibility to detect temperature transitions changing the thickness only by $1 \sim 2$ nm. This implies that the temperature decrease from 9 K down to 1 K is accompanied by the decrease of $1 \sim 2$ nm in the spatial oscillation period and by the decrease of about 0.3 nm in the oscillation length. In this temperature range the change of $\xi_{F1}$ is about $0.2$ nm as it has been estimated from $I_c(d_F)$ curves at different temperatures. At the same time simple evaluations of $\xi_{F1}$ (obtained from the slope of the $I_c(d_F)$ envelope) and $\xi_{F2}$ (estimated from the interval between two minima) show a large difference between these two lengths ($1.3$ nm and $3.5$ nm, correspondingly) that can not be explained by the thermal contribution described by $\xi_F$.

So, to carry out a theoretical analysis of the results obtained, we need to specify the nature of additional de-pairing processes that increase $\xi_{F2}$ and decrease $\xi_{F1}$. As the F layer is an alloy, a role of the magnetic scattering may be quite important. Magnetic inhomogeneity is related above all to Ni-rich clusters arising in $Cu_{1-x}Ni_x$ ferromagnet for $x$ close to 0.5. In the region of these concentrations when the Curie temperature is small, we may expect that the inverse spin-flip scattering time $\hbar\tau^{-1}$ could be of the order of the average exchange field $E_{ex}$ or even larger. This circumstance strongly modifies the proximity effect in the SF systems. A role of spin-orbit scattering should be neglected for the $CuNi$ alloy since it is substantial only in ferromagnets with large atomic number $Z$. To take into account the exchange field and the magnetic scattering in the framework of Usadel equations it is necessary just to substitute Masubara frequencies by $\omega \sim \omega + iE_{ex} + G\hbar/\tau_s$, where $G$ is the normal Green’s function. Note that this procedure assumes a presence of the relatively strong uniaxial magnetic anisotropy which prevents mixing of spin-up and spin-down Green functions.

To have some idea about the influence of the magnetic scattering on the proximity effect we may start with the linearized Usadel equation for the anomalous Green’s function in a ferromagnet

$$[\omega + iE_{ex} \operatorname{sgn}(\omega) + \frac{\hbar}{\tau_s}]F_j - \frac{\hbar D}{2} \frac{\partial^2 F_j}{\partial x^2} = 0.$$  

The exponentially decaying solution has the form

$$F_j(x, \omega > 0) = A \exp(-x(k_1 + ik_2)), $$  

with

$$k_1 = \frac{\hbar}{\xi_F \sqrt{1 + \left(\frac{\omega}{E_{ex}} + \frac{\hbar}{E_{ex} \tau_s}\right)^2}},$$

$$k_2 = \frac{\hbar}{\xi_F \sqrt{1 + \left(\frac{\omega}{E_{ex}} - \frac{\hbar}{E_{ex} \tau_s}\right)^2}}.$$

Here $\xi_F = \sqrt{\hbar D/E_{ex}}$ and $\xi_{F1,2} = 1/k_{1,2}$. The anomalous Green’s function $F_j$ at $\omega \sim k_B T_c$ gives us an idea about the spatial variation of the Cooper pair wave function. In the limit of the vanishing magnetic scattering and $k_B T_c << E_{ex}$ the decaying ($\xi_{F1}$) and the oscillating ($\xi_{F2}$) lengths are practically the same. However, if the spin-flip scattering time becomes relatively small $E_{ex} \tau_s/\hbar \lesssim 1$, the decaying length could be substantially

![FIG: The F-layer thickness dependence of the critical current density for Nb-$Cu_{0.47}Ni_{0.53}$-Nb junctions at temperature 4.2 K. Open circles present experimental results, solid and dashed lines show model calculations discussed in the second part of the paper.](image-url)
smaller than the oscillating length. This results in much stronger decrease of the critical current in SFS junctions with increase of the F layer thickness.

We have seen it experimentally \[6\] that the form of \(I_c(d_F)\) dependence varies a little with temperature, so a good idea about this dependence may be already obtained from the temperature region near \(T_c\). Using \(k = k_1 + i k_2\) in the form

\[
k = \sqrt{2 \left( |\omega| + i E_{ex} \text{sgn}(\omega) + \frac{h}{\tau_s} \right) / h D},
\]

we can obtain (see Ref. \[11\]) for the case of good SF-interface transparency and \(d_F \gg \xi_{F1}\) the following expression for the critical current:

\[
j_c \sim e^{-d_F/\xi_{F1}} (\cos d_F / \xi_{F2} + (\xi_{F1}/\xi_{F2}) \sin d_F / \xi_{F2}),
\]

where \(\xi_{F1,2}\) are taken in the limit of \(\omega \ll E_{ex}, h/\tau_s\).

Now we shall address the question of the exact thickness and temperature dependence of the critical current in SFS junctions. To deal with the complete set of the Usadel equations it is convenient to apply the usual parametrization of Green’s functions: \(G_f = \cos \Theta(x)\) and \(F_f = \sin \Theta(x)\). Then for \(\omega > 0\) the Usadel equation is written as

\[
\left( \omega + i E_{ex} + \frac{h \cos \Theta}{\tau_s} \right) \sin \Theta - \frac{h D}{2} \frac{\partial^2 \Theta}{\partial x^2} = 0.
\]

If the temperature variation of the exchange field is negligible at \(T < T_c\), the most direct way how the temperature could interfere is through the Matsubara frequencies. The presence of the magnetic scattering provides another mechanism of the critical current temperature dependence - through the normal Green’s function \(G_f = \cos \Theta\). The important range of the Matsubara frequencies variation is of the order of \(k_B T_c\) for superconductivity. Then in the case of the relatively strong magnetic scattering \(h \tau_s^{-1} >> k_B T_c\) the second mechanism of the temperature dependence will be predominant.

In the limit of relatively large F-layer thicknesses \(d_F > \xi_{F1}\) and rigid boundary conditions we may obtain an analytical solution of Eq. \(6\) and the expression for the critical current density reads

\[
j_c(d_F, T) = \frac{64\sigma_n \pi k_B T_c}{e \xi_F} \text{Re} \left( \sum_{n>0} \frac{F(n) q \exp(-qy)}{\sqrt{1-p^2}} F(n) + 1 \right)^2
\]

with the function

\[
F(n) = \frac{(\Delta/(2\pi k_B T))}{\left[ n + 1/2 + \sqrt{(n+1/2)^2 + (\Delta/(2\pi k_B T))^2} \right]^2}
\]

and \(y = d_F/\xi_F\), \(q = \sqrt{2\alpha + 2\alpha + 2\omega} \), where \(\alpha = h/(\tau_s E_{ex})\), \(\omega = \omega/E_{ex} = 2\pi(n+1/2)/[\xi_{ex}/k_B T_c]\) and \(1 - p^2 = (i + \omega)/(|\alpha + i + \omega|)\).

In the limit \(\alpha \to 0\) and \(k_B T_c << E_{ex}\) Eq. \(7\) coincides with that obtained previously in Ref. \[13\]. The theoretical fit of our experimental results which is based on Eq. \(7\) is presented in Fig. \(1\) by the solid line and in Fig. \(2\) by dashed lines. Besides the dashed line in Fig. \(1\) shows calculations made using Eq. \(6\). One can see a good agreement obtained with the following parameters: \(E_{ex}/k_B \approx 850 K\), \(h/\tau_s \approx 1.33 E_{ex}\), \(\xi_F = 2.16 nm\). The fitting also yields considerable value of ‘dead’ layers \(d_0; 2d_0 \approx 4.3 nm\), which do not take part in creating of the ‘sign-reversal’ superconductivity. The dead layer may arise due to not full correspondence of the theoretical approach and the real system. On the other hand, other experiments \[14\] also demonstrate the existence of large enough (2-3 nm) nonmagnetic layers at SF-surfaces.

A final remark concerns the real transparency of SF-surfaces in our SFS sandwiches. In modern theories an interface transparency is characterized by a parameter \(\gamma_B = (R_B S/\rho F \xi)^*\), where \(R_B\) is interface resistence per unit area, \(S\) is the SFS junction area, \(\rho F\) is F-layer resistivity and \(\xi^* = \sqrt{\hbar D/2\pi k_B T_c}\). To estimate \(R_B\) and \(\rho F\) we have carried out detailed measurements of SFS-junctions IV-characteristics. The upper inset in Fig. \(8\) shows that IV-characteristics are described by the expression \(V = R\sqrt{T^2 - T_c^2}\). The linear approximation presented in Fig. \(8\) has given \(R_B \approx 30 \mu\Omega\) for junctions with the area of \(10 \times 10 \mu\text{m}^2\) and \(\rho F \approx 62 \mu\Omega \cdot \text{cm}\). It allows to estimate following ferromagnet parameters:
the electron mean free path \( l \approx 1 \text{ nm} \), the diffusion coefficient \( D \approx 5.2 \text{ cm}^2/\text{s} \) and the characteristic spatial scale \( \xi^* \approx 9.4 \text{ nm} \). Values obtained determine the good enough transparency parameter \( \gamma_B = 0.52 \) that confirms the validity of the approximation used.

An additional breakthrough of the work is fabrication of \( \pi \)-junctions with large enough critical current density. Solving of this problem has enabled detailed experimental investigations of 0–\( \pi \)-transition peculiarities, reliable detections of second harmonic in the current-phase relation [13] and the 0–\( \pi \)-coexistence [16]. High magnitude of the critical current also allows to use \( SFS \) \( \pi \)-junctions as stationary phase \( \pi \)-shifters in novel modifications of the digital and quantum logic [17]. In proposed logic circuits \( \pi \)-junctions are connected together with ordinary tunnel junctions and should not introduce themselves any noticeable phase shift during dynamical switchings in the rest of the circuit. This is possible only if the \( \pi \)-junction critical current is much larger then critical currents of other junctions. The \( Nb-CuNi-Nb \) \( \pi \)-junctions are based on the standard niobium thin film technology so they can be incorporated directly into existing architectures of the superconducting electronics.

Thus, both 0–\( \pi \) and reverse \( \pi \)-0 transitions have been detected in \( SFS \) (\( Nb-CuNi-Nb \)) junctions for the first time. The double-reversal-thickness dependence of the critical current is a most striking evidence of the superconducting order parameter spatial oscillations in a ferromagnet close to \( SF \)-interface. We have also observed that the oscillation length in the ferromagnetic \( CuNi \) alloy is considerably larger than the the pair decay length. We have presented a theoretical description of an extra mechanism of the order parameter decay in \( CuNi \), mainly related to the strong spin-flip scattering on magnetic inhomogeneity.

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FIG. 3: Resistance of \( SFS \)-sandwiches normalized to the junction area of 10 \( \times \) 10 \( \mu \text{m}^2 \) vs. the \( F \)-layer thickness. Insets show typical IV- and Fraunhofer \( (L_s(H)) \) dependences of \( SFS \) junctions with ideal fitting by well-known Josephson expressions.

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