Thickness Dependence of the Josephson Ground States of Superconductor-Ferromagnet-Superconductor 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. Nodes of the critical current correspond to the transitions into the π state and back into the conventional 0 state. From the experimental data the superconducting order parameter oscillation period and the pair decay length in the ferromagnet are extracted reliably. We develop a theoretical approach based on the Usadel equations taking into account the spin-flip scattering. Results of numerical calculations are in good agreement with experiments.

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One of the exciting topics in studying the coexistence of superconductivity (S) and ferromagnetism (F) is the proximity-induced sign-reversal superconductivity in ferromagnets close to SF interfaces [1,2] (see [3] as a review). The decay of the superconducting order parameter in the ferromagnet is accompanied by the sign-changing oscillations. The unambiguous evidence of sign-reversal spatial oscillations of the superconducting order parameter in a ferromagnet was provided by the observation of the π state in SFS Josephson junctions [4–7]. "π junctions" [8] are weakly coupled superconducting structures with the ground state phase difference of the macroscopic superconducting wave function $\varphi = \pi$. They are characterized by the anomalous current-phase relation $I_s = I_c \sin(\varphi + \pi) = -I_c \sin\varphi$ with negative critical current [8]. Spatial oscillations of the superconducting order parameter in a ferromagnet close to an SF interface were predicted in Ref. [2]. The physical origin of the oscillations is the exchange splitting of spin-up and spin-down electron subbands in ferromagnets. In order to observe the manifestations of the transition into the π state one should fabricate SFS sandwiches with the F-layer thicknesses d_F close to integer numbers of half periods of the order parameter spatial oscillations $\lambda_{\rm ex}/2$ [4–7]. The period is $\lambda_{\rm ex} = 2\pi\xi_{F2}$, where the oscillation (or "imaginary") length ξ_{F2} can be extracted from the complex coherence length ξ_F in a ferromagnet: $\frac{1}{\xi_F} = \frac{1}{\xi_{F1}} + i\frac{1}{\xi_{F2}}$. In the case of large exchange energy and negligible magnetic scattering in the F layer the imaginary length ξ_{F2} and the order parameter decay length ξ_{F1} are equal [2]: $\xi_{F1} = \xi_{F2} = \sqrt{\hbar D/E_{\rm ex}}$, where D is the diffusion coefficient for electrons in a ferromagnet and $E_{\rm ex}$ is the exchange energy responsible for sign-reversal superconductivity in a ferromagnet. The possibility of manipulating the coherence length by temperature was demonstrated in Ref. [4], in which the temperature-driven 0-π transition was observed for the first time. For the case $E_{\rm ex} \gg k_BT$ the expressions [4] for $\xi_{F1}(T)$ and $\xi_{F2}(T)$ are given by:

$$\xi_{F1,2} \simeq \sqrt{\frac{\hbar D}{E_{\rm ex}}}\left(1 \mp \frac{\pi k_B T}{2E_{\rm ex}}\right). \quad (1)$$

Detailed experimental studies of the critical current versus F-interlayer thickness dependence for Nb-$Cu_{0.47}Ni_{0.53}$-Nb Josephson junctions were started by us in Ref. [7]. A very rapid decay of the critical current and its sharp reentrant behavior for thicknesses close to 22–23 nm were observed. Subsequent analysis of the experimental data and its comparison to the theoretical model described below indicated that at $d_F \approx 22$ nm transition from the π into the 0 state at the F-layer thickness close to the full oscillation period is observed, while the first node of the dependence corresponding to transition to the π state has to be at the thickness of about 10 nm. In the present work we report on the discovery of the first node and demonstrate the two-node behavior of the SFS junction critical current. We also discuss the possible mechanisms of the strong order parameter decay in ferromagnetic CuNi interlayers.

The nonmonotonic $I_c(d_F)$ dependence close to 0-π transition was observed for the first time in Ref. [4] and has been presented there in the form of a series of $I_c(T)$ curves for different F-interlayer thicknesses d_F. More detailed reentrant $I_c(d_F)$ curves for interlayer thicknesses close to 0-π transition were measured in Refs. [5,6]. In this work we have investigated the thickness dependence of the SFS junction critical current density in a wide thickness range for sandwiches (see inset in Fig. 1) fabricated in a multistep process by optical lithography and magnetron sputtering as described in Refs. [7,9]. All junctions were prepared with lateral sizes smaller than the Josephson penetration depth to ensure uniform supercurrent distribution. Junctions with barrier thicknesses of less than 17 nm had areas of $10 \times 10\ \mu m^2$ while junctions with thicker barriers were $50 \times 50\ \mu m^2$ in area to achieve observable critical currents.

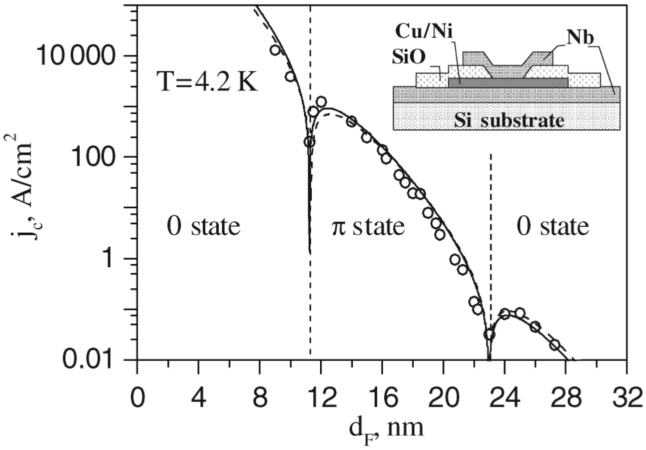

FIG. 1. The F-layer thickness dependence of the critical current density for Nb-$Cu_{0.47}Ni_{0.53}$-Nb junctions at temperature 4.2 K. Open circles represent experimental results; solid and dashed lines show model calculations discussed in the second part of the Letter. The inset shows a schematic cross section of our SFS junctions.

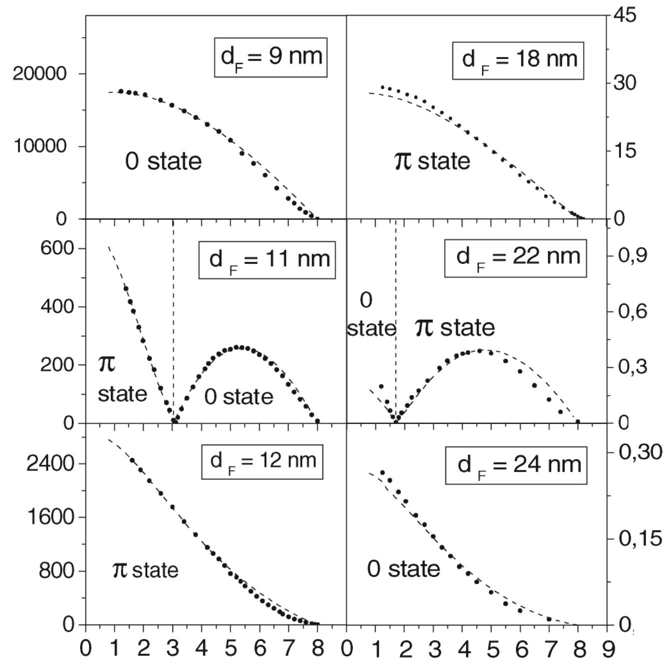

FIG. 2. Temperature dependences of the SFS junction critical current density at several F-layer thicknesses close to the critical ones. The dashed lines show calculation results based on Eq. (6).

Weakly ferromagnetic $Cu_{0.47}Ni_{0.53}$ interlayers had the Curie temperature of about 60 K. In the barrier thickness interval of 8–28 nm the critical current density varied by 6 orders of magnitude and had nodes at two d_F values as presented in Fig. 1.

One can see that the curve in Fig. 1 demonstrates both direct 0-π transition and reverse transition from π to 0 state. At transition points, $d_{c1,c2}$, the critical current $I_c(d_F)$ is equal to zero and then should formally change its sign. Since in our transport experiments we could measure only the magnitude of the critical current, the negative region of $I_c(d_F)$ between the two sharp cusps (that corresponds to the π state) is reflected into the positive region. Because of a slight temperature dependence of the order parameter oscillation period [described by Eq. (1)] and other temperature dependent processes discussed below, we could pass through the transition points using samples with critical F-layer thicknesses $d_{c1} = 11$ nm and $d_{c2} = 22$ nm by changing temperature. Temperature-driven 0-π and π-0 transitions are presented in the middle panels of Fig. 2. The upper and lower panels show the temperature dependences of the critical current for samples with F-layer thicknesses around $d_{c1,c2}$. For barrier thicknesses over 1–2 nm from $d_{c1,c2}$ 0-π transitions are not observed in the experimental temperature range. This implies that the temperature decrease from 9 K down to 1 K is accompanied by the decrease of 1–2 nm in the spatial oscillation period and by the decrease of about 0.3 nm in the imaginary length. In this temperature range the change of ξ_{F1} is about 0.2 nm as it has been estimated from $I_c(d_F)$ curves at different temperatures. However, values of $\xi_{F1} = 1.3$ nm and $\xi_{F2} = 3.5$ nm obtained from the slope of the $I_c(d_F)$ data and from the interval between the two minima of $I_c(d_F)$ correspondingly differ by almost a factor of 3, which cannot be explained simply by thermal terms in the Eq. (1) [10].

We propose a theoretical model that includes additional depairing processes which increase ξ_{F2} and decrease ξ_{F1}. Because our F layer is an alloy, the role of magnetic scattering in the junction barrier is important [6,7]. Magnetic inhomogeneities due to Ni-rich clusters [12,13] are known to exist in $Cu_{1-x}Ni_x$ films for x close to 0.5. For such Ni concentrations the Curie temperature is small, and we may expect that the inverse spin-flip scattering time $\hbar\tau_s^{-1}$ could be of the order of the average exchange field E_{ex} or even larger. This circumstance strongly modifies the proximity effect in SF systems. The role of spin-orbit scattering should be neglected for $Cu_{1-x}Ni_x$ alloys since it is only substantial in ferromagnets with large atomic numbers Z. To take into account the exchange field and the magnetic scattering in the framework of the Usadel equations it is necessary to substitute the Matsubara frequencies as follows: $\omega \to \omega + iE_{ex} + G\hbar/\tau_s$ [14], where G is the normal Green's function. Note that this procedure assumes the presence of relatively strong uniaxial magnetic anisotropy which prevents the mixing of spin-up and spin-down Green's functions [3].

To understand the influence of magnetic scattering on the proximity effect we start with the linearized Usadel equation [15] for the anomalous Green's function in a ferromagnet

$$\left[|\omega| + iE_{ex}\mathrm{sgn}(\omega) + \frac{\hbar}{\tau_s}\right]F_f - \frac{\hbar D}{2}\frac{\partial^2 F_f}{\partial x^2} = 0. \quad (2)$$

The exponentially decaying solution has the form $F_f(x, \omega > 0) = A\exp[-x(k_1 + ik_2)]$ with

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

Here $\xi_F = \sqrt{\hbar D/E_{\rm ex}}$ and $\xi_{F1,2} = 1/k_{1,2}$. The anomalous Green's function F_f at $\omega \sim k_B T_c$ contains information about the spatial variation of the superconducting order parameter. In the limit of vanishing magnetic scattering and $k_B T_c \ll E_{\rm ex}$ the decay (ξ_{F1}) and the oscillation (ξ_{F2}) lengths are nearly equal. However, if the spin-flip scattering time becomes relatively small $E_{\rm ex}\tau_s/\hbar \lesssim 1$, the decay length could be substantially smaller than the oscillation length. This results in much stronger decrease of the critical current in SFS junctions as the F layer thickness increases.

Previously, we have observed [7] that the shape of the $I_c(d_F)$ dependence varies very little with temperature; therefore, for the qualitative theoretical model we can consider the temperature region near T_c. Using $k = k_1 + ik_2$ in the form

$$k = \sqrt{2[|\omega| + iE_{\rm ex}\,{\rm sgn}\,(\omega) + \hbar/\tau_s]/\hbar D}, \tag{3}$$

we can obtain [see Ref. [3]] for the case of high SF-interface transparency and $d_F \gg \xi_{F1}$ the following expression for the critical current density:

$$j_c \sim e^{-d_F/\xi_{F1}}[\cos(d_F/\xi_{F2}) + (\xi_{F1}/\xi_{F2})\sin(d_F/\xi_{F2})], \tag{4}$$

where $\xi_{F1,2}$ are taken in the limit of $\omega \ll E_{\rm ex}, \hbar/\tau_s$.

In order to obtain a more exact expression for the thickness and temperature dependence of the critical current in SFS junctions it is necessary to consider the complete set of the Usadel equations. In this case, it is convenient to apply the usual parametrization of the Green's functions: $G_f = \cos\Theta(x)$ and $F_f = \sin\Theta(x)$. Then for $\omega > 0$ the Usadel equation is written as

$$\left(\omega + iE_{\rm ex} + \frac{\hbar\cos\Theta}{\tau_s}\right)\sin\Theta - \frac{\hbar D}{2}\frac{\partial^2\Theta}{\partial x^2} = 0. \tag{5}$$

If the temperature variation of the exchange field is negligible at $T < T_c$ the most direct way in which temperature can enter the solution is through the Matsubara frequencies. The magnetic scattering rate appears in the Eq. (5) together with the normal Green's function $\cos\Theta$ that makes the effective magnetic scattering temperature dependent. This provides an additional contribution to the temperature dependence of the critical current. The important range of the Matsubara frequencies is of the order of $k_B T_c$ for superconductivity. In the case of relatively strong magnetic scattering, such that $\hbar\tau_s^{-1} \gg k_B T_c$, the latter mechanism of the temperature dependence will be predominant. In the limit of large F-layer thicknesses $d_F > \xi_{F1}$ and rigid boundary conditions we may obtain the analytical solution of Eq. (5). The expression for the critical current density then reads

$$j_c(d_F, T) = \frac{64\sigma_n\pi k_B T_c}{e\xi_F} \times {\rm Re}\left(\sum_{n>0}^{\infty}\frac{\mathcal{F}(n)q\exp(-qy)}{[\sqrt{(1-p^2)\mathcal{F}(n)+1}+1]^2}\right) \tag{6}$$

with the function

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

and $y = d_F/\xi_F$, $q = \sqrt{2i + 2\alpha + 2\tilde{\omega}}$, where $\alpha = \hbar/(\tau_s E_{\rm ex})$, $\tilde{\omega} = \omega/E_{\rm ex} = \frac{2\pi(n+1/2)(T/T_c)}{E_{\rm ex}/k_B T_c}$, and $1 - p^2 = (i + \tilde{\omega})/(\alpha + i + \tilde{\omega})$.

In the limit $\alpha \to 0$ and $k_B T_c \ll E_{\rm ex}$, the Eq. (6) coincides with that obtained previously in Ref. [16]. The theoretical fit of our experimental results which is based on Eq. (6) is presented in Fig. 1 by the solid line and in Fig. 2 by the dashed lines. Besides the dashed line, Fig. 1 shows calculations based on Eq. (4). Good agreement was obtained with the following parameters: $E_{\rm ex}/k_B \approx 850$ K, $\hbar/\tau_s \approx 1.33E_{\rm ex}$, $\xi_F = 2.16$ nm. The fitting also yields a considerable offset thickness of "dead" layers d_0: $2d_0 \approx 4.3$ nm, which do not take part in the "oscillating" superconductivity. Thicknesses of dead layers cannot be calculated quantitatively in the framework of the proposed theoretical model. Other experiments [17] also demonstrate the existence of comparable (2–3 nm) nonmagnetic layers at SF interfaces.

In order to evaluate the relevance of the described model to our data, it is important to estimate the interface transparency parameter $\gamma_B = (R_B S/\rho_F\xi^*)$, where R_B is interface resistance per unit area, S is the SFS junction area, ρ_F is F-layer resistivity, and $\xi^* = \sqrt{\hbar D/2\pi k_B T_c}$. Both R_B and ρ_F can be estimated from the measurements of IV characteristics of SFS junctions. The upper inset in Fig. 3 shows that the IV characteristics are well described by the expression $V = R\sqrt{I^2 - I_c^2}$, with R values presented in the main panel. The linear approximation of the $R(d_F)$ dependence yields $R_B \approx 30\ \mu\Omega$ for junctions with the area of $10 \times 10\ \mu{\rm m}^2$ and $\rho_F \approx 62\ \mu\Omega$ cm. It allows us to estimate following parameters in our ferromagnet: the electron elastic mean free path $l \approx 1$ nm, the diffusion coefficient $D \approx 5.2\ {\rm cm}^2/{\rm s}$, and the characteristic spatial scale $\xi^* \approx 9.4$ nm. The transparency $\gamma_B = 0.52$ is sufficiently high, which allows the approximation above to be used.

Experiments on SFS junctions with thinner barriers, and hence higher critical current densities, may lead to several additional breakthroughs. In particular, the chances of observing the signatures of secondary Josephson tunneling are higher in junctions with thinner barriers due to reduced

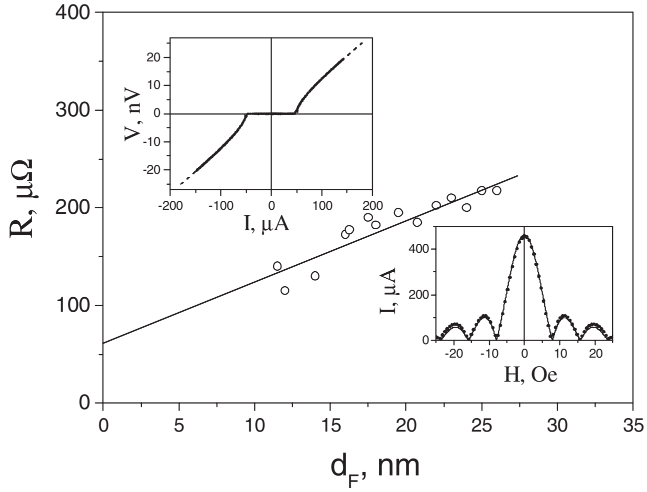

FIG. 3. Resistance of SFS sandwiches normalized to the junction area of $10 \times 10\ \mu m^2$ vs the F-layer thickness. Insets show typical IV and Fraunhofer [$I_c(H)$] dependences of our SFS junctions fitted to the conventional Josephson expressions.

order parameter damping [18]. Because of increased sensitivity of the critical current density to thickness variations near the first node of the $I_c(d_F)$ it becomes possible to study the 0-π junction regime [9] and search for the predicted 0-π coexistence [19]. Critical current densities of π junctions were enhanced by 3 orders of magnitude opening way for embedding SFS π junctions into digital and quantum circuits as stationary phase π shifters [20]. In the proposed logic circuits, π junctions are connected with tunnel junctions and should not introduce any noticeable phase shift during dynamical switchings in the rest of the circuit. This is possible only if the π junction critical currents are much larger than the critical currents of other junctions. The Nb-CuNi-Nb π junctions are based on the standard niobium thin film technology and can be incorporated directly into the existing architectures of superconducting electronics.

Thus, both 0-π and reverse π-0 transitions were detected in SFS (Nb-CuNi-Nb) junctions for the first time. The double-reversal thickness dependence of the critical current is the most striking evidence for the spatial oscillations of the superconducting order parameter in ferromagnets close to SF interface. We have also observed that the oscillation length in the ferromagnetic $Cu_{1-x}Ni_x$ alloy is considerably larger than the pair decay length. We have presented a theoretical description that accounts for additional order parameter decay due to strong spin-flip scattering on magnetic inhomogeneities.

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[10] We use here a model based on the Usadel equations that is justified by the diffusive regime of the electron motion in $Cu_{1-x}Ni_x$ alloy. In the ballistic regime (no impurities scattering in F layer) the critical F layer thickness of 0-π transition also slightly depends on the temperature [1,11]. This can lead to the I_c(T) dependence qualitatively similar to that presented in Fig. 2, which cannot explain our results quantitatively.

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