Oscillations of the Superconducting Critical Current in Nb-Cu-Ni-Cu-Nb Junctions

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We report on experimental studies of superconductor-ferromagnet layered structures. Strong oscillations of the critical supercurrent were observed with the thickness variation of the ferromagnet. Using known microscopic parameters of Ni, we found reasonable agreement between the period of oscillations and the decay of the measured critical current, and theoretical calculations.

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The interplay between superconductivity and ferromagnetism is an old subject which was studied extensively over decades [1–3]. The most striking effect in such systems is the formation of the so-called π phase junction in a superconductor-ferromagnet-superconductor (SFS) structure [1]. One of the manifestations of the π phase is a nonmonotonic variation of the critical temperature [3,4], T_c, with the variation of the ferromagnetic layer thickness, d. Oscillatory dependence of T_c vs d of SF multilayers has been observed by few groups [5–8], however, other groups [9–11] reported monotonic behavior in similar structures. Interpretations of these experiments suggested the π junction mechanism as well as some other origins of the effect [6,10,12]. The recent observation [13] of nonmonotonic behavior of the critical current *as a function of temperature* in a weak-ferromagnetic layer of Cu_xNi_{1-x} between two Nb layers is considered as an unambiguous proof of the π phase formation. Another interesting theoretical prediction [2,3,14,15] concerns nonmonotonic behavior of the critical current as a function of the thickness of the ferromagnetic layer d. According to the above predictions, the critical current I_c is expected to oscillate and decay as d is increased. To the best of our knowledge, such a behavior has not been reported so far.

In this paper, we present the experimental evidence of oscillatory behavior of the critical current vs thickness variation of ferromagnetic Ni layer. We also show a reasonable agreement between our data and the theories [2,15] in the appropriate limit of $E_{ex} \gg \hbar/\tau \gg k_B T_c$. Here, τ and E_{ex} are the electron relaxation time and the exchange energy of the ferromagnet, and T_c is the critical temperature of the superconductor.

We have studied temperature and thickness dependence of the critical current in Nb-Cu-Ni-Cu-Nb junctions. Additionally, we have studied the thickness dependence of critical current of similar junctions without a Ni layer. The junctions with $10 \times 10\ \mu m^2$ area were fabricated with the standard photolithography technique. The process contained three stages of lithography: liftoff of the bottom Nb-Cu layer, liftoff of the variable thickness Ni or Cu layers, and liftoff of the top Cu-Nb layer. Nb films were sputtered using a magnetron gun and *in situ* covered with the Cu layer by thermal evaporation, for preventing the Nb oxidation. The ferromagnet layers of Ni were e-gun evaporated in a separate vacuum chamber, and subsequently covered *in situ* by Cu. It is important to emphasize that all samples were prepared simultaneously. The variation of Ni thickness for Nb-Cu-Ni-Cu-Nb junction and variation of Cu for Nb-Cu-Nb junctions was obtained by a specially designed shutter, which exposed the samples in sequence, so that every sample was exposed to the evaporating Ni or Cu for additional fragments of time. This method guaranteed that all the interfaces between each layer in our multilayer structure are identical, and the only difference between the samples is their Ni or Cu thickness. The thickness of each Nb layer was 2000 Å. The total thickness of the Cu in the Nb-Cu-Ni-Cu-Nb junctions was 2400 ± 250 Å and the Ni thickness varied from 10 ± 1 Å to 90 ± 1 Å. In the other set of junctions, namely, Nb-Cu-Nb, the Cu thickness varied from 5000 to 10 000 Å. The structure of these junctions is shown schematically in the inset of Fig. 1.

Typical resistance of both types of junctions was around 100 $\mu\Omega$, independent of both Ni and Cu

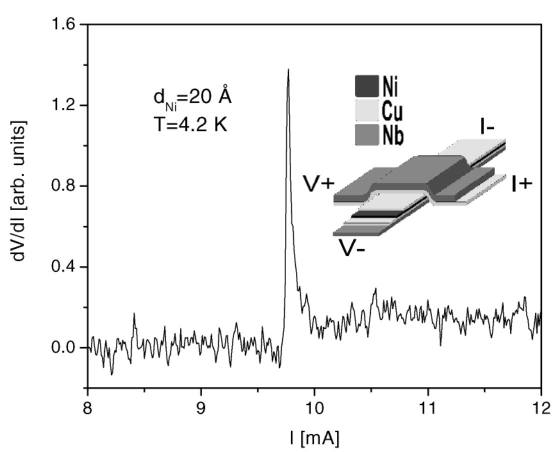

FIG. 1. dV/dI of one of the samples. The critical current was defined as the onset of the peak. In the inset: schematic picture of a Nb-Cu-Ni-Cu-Nb junction.

thicknesses. The critical temperature of the Nb was about 8.5 K. The measured resistivity of Ni layer $\rho = 31\ \mu\Omega$ cm at 4.2 K, together with the known value [16] $\rho l = 1.5 \cdot 10^{-11}\ \Omega\,\mathrm{cm}^2$, allowed us to establish the mean free path $l = 48$ Å.

The measurements were performed in ^{4}He cryostat in the range from 6.5 K down to 1.5 K. The critical current was measured by passing a dc current with a small ac modulation through the sample. The ac voltage which appeared above the critical dc current was picked up by lock-in amplifier operated in a transformer mode.

A typical differential resistance of both types of junctions vs dc current is plotted in Fig. 1, which clearly indicates on the sharp onset of dissipation at $I = I_c$. First, we show results of the Nb-Cu-Nb set (Fig. 2). As expected [17], the critical current I_c normalized to the length of the junction L, decreases exponentially with L. From the decay of the critical current we found the thermal length of the the Cu, $L_T = \sqrt{\hbar D/2\pi k_B T} = 1600$ Å at 4.2 K, and the diffusion constant $D = 860\ \mathrm{cm}^2/\mathrm{s}$.

Very different thickness dependence was found in the Nb-Cu-Ni-Cu-Nb junctions. Figure 3 shows the thickness dependence of the critical current in these junctions at $T = 4.2$ K. In spite of the large statistical error bars, the nonmonotonic variation of the critical current is quite evident, namely, the deviations of the data from the exponential decay surmounts by far the uncertainty of each measured point.

A further, and even a stronger evidence for the oscillatory behavior is provided by temperature dependence of the critical current. Figure 4 shows a family of I_c vs T curves for all Ni thicknesses, which are normalized to their values at 4.2 K. We define the slope α_T of temperature variation, namely $\alpha_T \equiv d[\ln I_c(T)]/dT$, and plot these values as a function of d in Fig. 3 (squares). Oscillations of α_T are very prominent, and are in antiphase with the oscillations of the critical current. Unlike the critical current, which had quite large experimental error bars, the slope of the temperature dependence had error bars of only a few percent. Such a behavior of α_T as a function of d is intimately related to the variation of the critical current oscillations amplitude with temperature. Although the amplitude of the oscillations decreases at high temperatures, their relative value increases, and on a semilogarithmic plot of Fig. 5, the oscillatory behavior of I_c is more pronounced at higher temperatures.

We would like to start our discussion with what is well known for superconductor-normal-superconductor (SNS) junctions. The dependence of critical current of such junctions on the thickness of N layer is exponential, $I_c \alpha e^{-L/L_T}$ for $L > L_T$ [17]. In the opposite limit, where $L < L_T$, I_c approaches a constant value. This behavior has been observed in few experiments for different material systems [18,19], including our sample presented in Fig. 2.

Nonmonotonic behavior of the critical current is not expected theoretically for long metallic junctions [20], and had never been observed experimentally in SNS structures. Since the thickness of Ni is the only parameter which is varied in our Nb-Cu-Ni-Cu-Nb samples, it is reasonable to assume that the oscillations observed in the data are due to the presence of Ni in the structure.

As mentioned in the introduction, oscillatory behavior of the critical current vs the thickness of the ferromagnetic layer is predicted theoretically [2,3,15]. The origin of these oscillations is the phase shift acquired by electron-hole pair upon entrance into the ferromagnet, due to their different spin orientations. Several expressions have been derived for the critical current in various limits of the strength of E_{ex}, thickness of the ferromagnet d and disorder. Since in our experiment we have determined only the magnitude of the critical current I_c, the formulas below will be written for the absolute value $|I_c|$. For the clean, thin and strong ferromagnetic layers

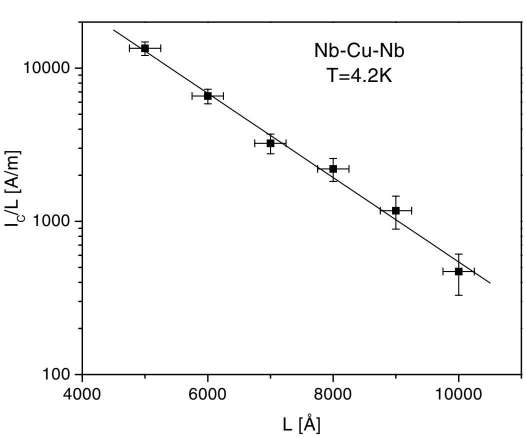

FIG. 2. The normalized critical current I_c/L of the Nb-Cu-Nb junction as a function of the Cu thickness L. The straight line represents the theoretical decay.

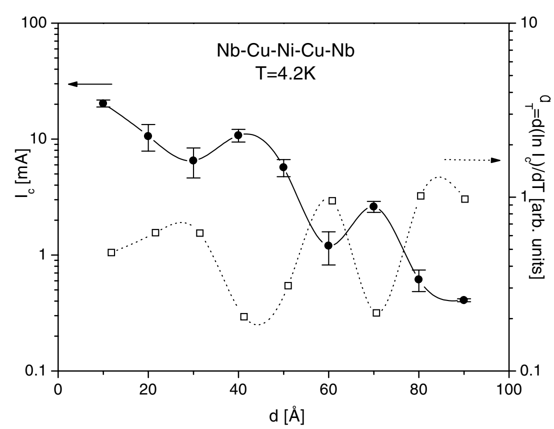

FIG. 3. Critical current of the Nb-Cu-Ni-Cu-Nb junctions as a function of the Ni layer's thickness d at 4.2 K (circles). The dependence of the slope α_T on d is represented by squares. The dashed and dotted lines are only for guiding the eye.

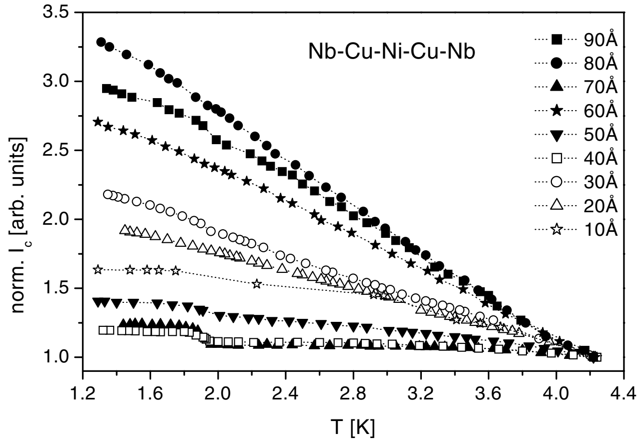

FIG. 4. Critical current as a function of temperature of the Nb-Cu-Ni-Cu-Nb junctions for different thicknesses of the Ni layer.

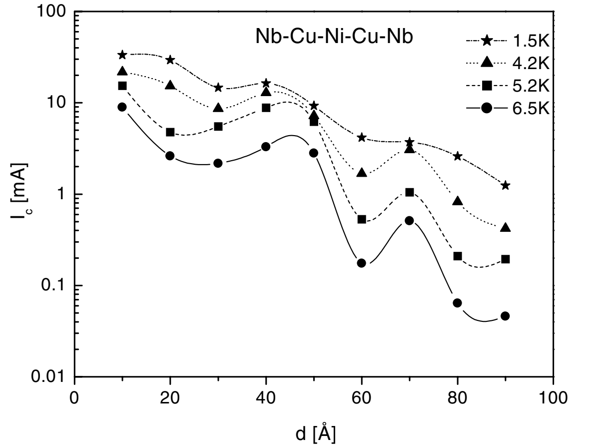

FIG. 5. Critical current as a function of the thickness of the Ni layer at different temperatures.

$l > \hbar v_f/E_{ex}$, d, the critical current should vary with the thickness as [2]:

$$I_c \sim |\sin(2E_{ex}d/\hbar v_f)|/(2E_{ex}d/\hbar v_f). \quad (1)$$

The dependence of absolute value of the critical current is shown on Fig. 6 by a dashed line. In the opposite limit, namely $l < d$, the following dependence was derived [3] (dotted line in Fig. 6):

$$I_c \sim ye^{-y}|\sin(y + \pi/4)| \qquad y = d\sqrt{4E_{ex}/\hbar D}. \quad (2)$$

Another expression for the critical current in the limit $l > \hbar v_f/E_{ex}$, is given by [15]:

$$I_c \sim \left| \mathrm{Re} \sum_{\omega_n > 0} \frac{\Delta^2}{\Delta^2 + \omega_n^2} \int_{-1}^{1} \frac{\mu d\mu}{\sinh(k_\omega d/\mu l)} \right|, \quad (3)$$

where $\omega_n = \pi T k_B(2n + 1)$ is the Matsubara frequency, n is an integer number, $k_\omega = (1 + 2|\omega_n|\tau/\hbar) - 2iE_{ex}\tau/\hbar$, $\mu = \cos\theta$, θ is the angle between the momentum and the normal to the SF interface, and Δ is the order parameter in the superconductor. The solid line of Fig. 6 represents the above expression. In fitting our data to the expressions for all limits, we have used [21] $v_f = 2.8 \times 10^5$ m/sec and $l = 48$ Å (based on the resistivity measurements of Ni). Therefore, the only fitting parameter apart from the numerical prefactor was the strength of the exchange interaction E_{ex}.

The periodicity of oscillations in Eq. (1), $L_{osc} \sim \pi\hbar v_f/E_{ex} \simeq 54$ Å best fits our data when $E_{ex} = 107 \pm 3$ meV. Note, that L_{osc} is twice as large as the periodicity observed in Fig. 6, due to the absolute value taken for I_c. In order to have similar periodicity in Eq. (2), we have used weaker exchange field $E_{ex} = 86$ meV. All values are close to the recently reported value [21] $E_{ex} = 115$ meV.

The curve given by Eq. (3) in Fig. 6 follows the decay and the phase of the oscillations of the measured I_c closer than the other two curves for large d. Furthermore, we estimate $\hbar v_f/E_{ex} = 17$ Å which is smaller than the measured mean free path $l = 48$ Å, and therefore, the validity of using Eq. (3), $l > \hbar v_f/E_{ex}$, is justified. However, since Eq. (3) is valid only for $d > l$, the data points should follow Eq. (1) for $d < 40$ Å. We thus conclude that our data are consistent with Eqs. (1) and (3) in their appropriate limits. Therefore, we give the fit of Eqs. (1) and (3) to our data for four temperatures in Fig. 7.

The oscillations of α_T (Fig. 3), which slightly smear the oscillations of I_c (Fig. 5), are not accounted by any of the mentioned theories and have only a minor effect on the overall quality of the fit presented in Fig. 7. A possible origin of this effect could be related to the presence of normal metal on both sides of the ferromagnetic layer. The structure we have used contains layers of Cu of a combined thickness $L = 2400$ Å, which is much larger than the thickness of the Ni layer. It is obvious that at high temperatures ($L_T < L$) the contribution of the

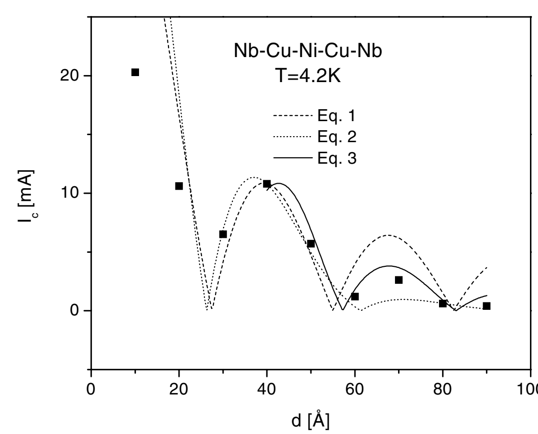

FIG. 6. Critical current of the Nb-Cu-Ni-Cu-Nb junctions at 4.2 K, and the theoretical fits. The dashed line represents Eq. (1), the dotted line Eq. (2), and the solid line Eq. (3).

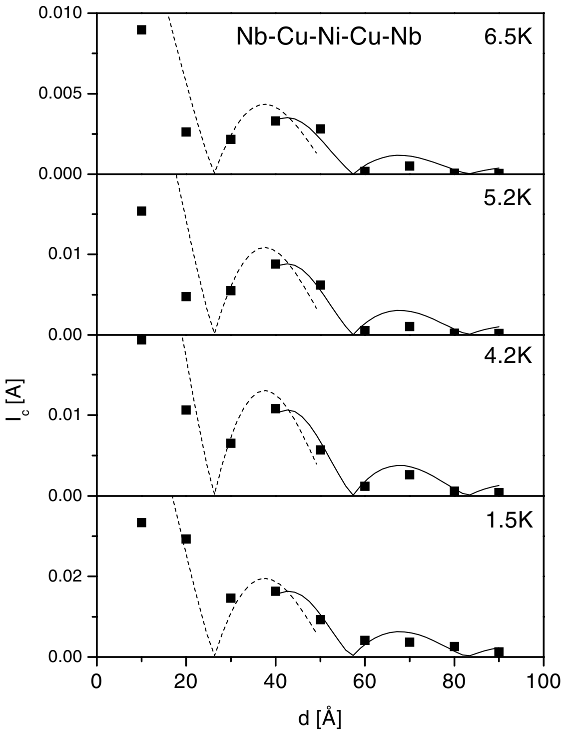

FIG. 7. Critical current of the Nb-Cu-Ni-Cu-Nb junctions at 1.5, 4.2, 5.2, and 6.5 K, and the theoretical fits according to Eq. (3) (solid line) and Eq. (1) (dashed line).

electron-hole pair propagating via short diffusion path is predominant, since paths which are much longer than L do not contribute to the supercurrent. At low temperatures, where $L_T > L$, the contribution of longer paths is not negligible. The latter implies that the impact of the trajectories crossing Ni layer several times is higher at low temperatures. The multiple crossing of the ferromagnetic layer could produce additional harmonics of the basic frequency of the oscillations, which will smear the nonmonotonic behavior. This multiple crossing has not been considered in the existing theories for S-F-S junctions, and therefore might be responsible for the observed temperature dependence of the oscillations in S-N-F-N-S junctions (Fig. 4). A different origin of the anomalous temperature dependence could be related to the nontrivial Fermi surface of the Ni [22], which is also not accounted in the theoretical models.

In summary, we have observed the oscillations and the decay of the critical current in Nb-Cu-Ni-Cu-Nb junctions upon the increase of the thickness of the Ni layer. We found a reasonable agreement with the recent theoretical calculations in the appropriate limit. The puzzling temperature dependence of the oscillations' amplitude remains unexplained.

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