Observation of periodic $\pi$-phase shifts in ferromagnet-superconductor multilayers

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We report complementary studies of the critical temperature and the critical current in ferromagnet (Ni) - superconductor (Nb) multilayers. The observed oscillatory behavior of both quantities upon variation of the thickness of the ferromagnetic layer is found to be in good agreement with theory. The length scale of oscillations is identical for both quantities and is set by the magnetic length corresponding to an exchange field of 200 meV in Ni. The consistency between the behavior of the two quantities provides strong evidence for periodic $\pi$ phase shifts in these devices.

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INTRODUCTION

Devices of superconducting materials as, for example, Josephson junctions, have proven exceptionally useful in many fields of physics. In bulk conventional superconductors, the spin degree of freedom is frozen out by the spin-singlet Cooper pair formation. The proximity to a ferromagnetic material, however, opens up the spin as an additional degree of freedom. Consequently, the construction of Josephson junctions of superconductor-ferromagnet-superconductor (S-F-S) materials has attracted considerable attention recently within the emerging field of spintronics.

One intriguing effect associated with the new spin degree of freedom in an S-F-S Josephson junction is the thermodynamic stability of a phase difference $\pi$ between the two superconductors for certain parameter ranges of the middle ferromagnetic material. The $\pi$ phase is a result of a peculiar superconducting proximity effect in the ferromagnet (F). The two spin species are split in energy by the exchange field $E_{\text{ex}}$, which leads to an oscillatory behavior of the proximity induced pair amplitude in the ferromagnet. As a result, properties such as the critical temperature $T_c$, and the critical current $I_c$, are nonmonotonic, oscillating, and decaying functions of increasing ferromagnetic thickness. For the Josephson junction, the free energy loss due to the energy splitting can be compensated for by a spontaneous appearance of a superconducting phase difference of $\pi$ over the junction. This additional degree of freedom leads to a series of $0 \rightarrow \pi$ and $\pi \rightarrow 0$ transitions, that can be observed as zero crossings of $I_c$ and as kinks and minima of $T_c$ with a varying ferromagnetic layer thickness.

Previous studies of these effects have been focused on ferromagnetic alloys sandwiched between two superconductors, because the corresponding oscillation wavelength is quite long and is easily resolved. It is important for the development of applications to also understand devices including strong ferromagnets as iron, cobalt, or nickel, but the short oscillation wavelength in these materials is harder to resolve. In previous reports on devices made of strong ferromagnets either the $T_c$ variations or the $I_c$ variations were considered as a function of the ferromagnetic layer thickness. However, sample preparation techniques make a direct comparison of the different experiments difficult. For a successful theoretical understanding a high control of material properties is required.

In this paper we report studies of both the critical current and the critical temperature variations as a function of the F layer thickness in S-F-S junctions made of one set of materials, namely Nb-Ni-Nb junctions prepared under identical conditions. We find that both quantities, $I_c$ and $T_c$, vary on the same scale, the magnetic length $L_M = \sqrt{\hbar D_F/E_{\text{ex}}}$ set by the properties of the ferromagnet only ($D_F$ is the diffusion constant in the ferromagnet). By a detailed comparison of our measurements with theory, we find consistent fits for an exchange field of $E_{\text{ex}} = 200$ meV in Ni.

SAMPLE PREPARATION

The samples for the $I_c$ measurements had a $10 \times 10 \, \mu m^2$ cross sectional area and were fabricated with a standard photolithography technique. The process contained three stages of lithography: Liftoff of the bottom Nb/Cu layers, liftoff of the variable thickness Ni layer, and liftoff of the top Cu/Nb layers. We fabricated two sets of nine Nb-Cu(Au)-Ni-Cu(Au)-Nb junctions with variable Ni thickness in the range of 35–75 Å in steps of 5 Å (set 2) and 4 Å (set 3). The thickness of each Nb layer is 2000 Å, while the total thickness of the Cu is 2400±250 Å (Au-500±50 Å). We show the layout of the junctions for the $I_c$ measurements in Fig. 1(a).

The Nb films were sputtered using a magnetron gun and were covered in situ with a Cu (set 1.2) or a Au (set 3) layer by thermal evaporation to prevent Nb oxidation upon removal from the vacuum chamber for the subsequent lithography stage. Prior to deposition of the ferromagnet layer, the normal metal was deposited again to refresh the surface. The contaminated interface was in this way kept inside the normal metal. We have two such interfaces in each sample, which most probably will determine the sample resistance.

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The ferromagnet layers of Ni were e-gun evaporated in a separate vacuum chamber at a pressure of 2·10⁻⁷ Torr and subsequently covered in situ by Cu or Au. The variation of the Ni thickness was achieved by a specially designed shutter, which exposed the samples in sequence, so that every sample was exposed to the evaporating Ni for additional fragments of time. Because all samples within one set were prepared simultaneously, all layer interfaces are nominally identical and the only difference between the samples is their Ni thickness. 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 a lock-in amplifier operated in a transformer mode. The measurements were performed in a ²He cryostat in the range from 4.2 K down to 1.5 K.

The samples for the $T_c$ studies were prepared by an in situ evaporation of Ni and Nb layers without photolithography, see Fig. 1(b). Two sets of structures, each containing 16 samples were fabricated. The first set of samples contains only a single layer of Nb and was obtained by sequential deposition of Ni(30 Å)-Nb(430 Å)-Ni(x), where Ni(x) was varied from 0 to 37 Å in steps of ~2.5 Å. The second set contains two Nb layers, namely Ni(30 Å)-Nb(430 Å)-Ni(x)-Nb(430 Å)-Ni(30 Å), and was prepared in a similar manner. The thicknesses of the bottom and top Ni layers, as well as the thicknesses of the Nb layers, were chosen such that the bulk $T_c$ of Nb was suppressed. This increases the sensitivity of $T_c$ to variations of the thickness of the center Ni layer.

For strong ferromagnets like Ni, the magnetic length $L_M$ is below 20 Å. The first $0 \to \pi$ transition is, therefore, expected to occur for very thin films. However, Ni films thinner than a few tens of Ångströms prepared by standard e-gun evaporation is not expected to be homogeneous or to perfectly cover a metallic surface. This is a problem for measurements of $I_c$ in S-F-S junctions with a very thin F-layer, since uncovered regions short-circuit the junction. For thermodynamic measurements, such as measurements of $T_c$, an inhomogeneous coverage is less of a problem. At the same time, the effect we address decays exponentially with the layer thickness and it is undesirable to have very thick films, in particular for thermodynamics measurements where the oscillations become negligible. With this in mind, we have chosen Ni-film thicknesses ranging from 0 to 35 Å for the $T_c$ measurements and thicknesses ranging from 35 to 75 Å for the $I_c$ measurements.

**FIG. 1.** (Color online) Schematic layout: (a) Of the junctions for $I_c$ experiments, (b) of planar samples for $T_c$ experiments.

CRITICAL CURRENT

The results of the critical current measurements are shown in Fig. 2(a). We have also included data published earlier (Refs. 13 and 15), here named set 1.

Since the sets were prepared separately and are somewhat different in their normal metal constituents, they are expected to have different interface properties reflected as a different transmission coefficient $T$ of the superconductor-ferromagnet interface, which enters the calculations of $I_c$ as a prefactor $T^2$ (see below). Therefore, we have normalized the values of $I_c$ by $T^2$ varying between the sets to minimize the deviations from the theoretical curve, as indicated in the legend of Fig. 2.

An expression for the critical current for a diffusive system $T_c \ll \hbar$ ($\tau$ is the momentum relaxation time) with a strong exchange field $E_{ex} \tau \gg \hbar$ was derived in Ref. 16 and has the form

$$I_c \sim \text{Re} \sum_{\omega_n > 0} \frac{\Delta^2}{\Delta^2 + \omega_n^2} \int_1^{T_c} \frac{\mu d\mu}{\sin(k_n\delta_N/\mu \ell)},$$

where $\omega_n = \pi k_BT(2n+1)$ is the Matsubara frequency, $n$ is an integer number, $d_N$ is the thickness of the ferromagnet, $\ell$ is the mean free path, $k_n = (1+2|\omega_n|/\tau \hbar) - 2iE_{ex}/\tau \hbar$, $\mu = \cos \theta$, $\theta$ is the angle between the quasiparticle momentum and the normal to the SF interface, and $\Delta$ is the order parameter deep inside the superconductor. In the limit where the ferromagnet layer thickness is large compared to the mean free path, $d_N \gg \ell$, Eq. (1) can be approximated by the following expression

$$I_c = \frac{eN^2}{2\hbar} \frac{\sin(2E_{ex}d_N/v_F)}{16} \frac{2E_{ex}d_N/v_F}{v_F} \times k_BT \sum_{\omega_n > 0} \frac{\Delta^2}{\Delta^2 + \omega_n^2} \exp\left[-\frac{d_N(1+2|\omega_n|/\tau)}{\ell}\right],$$

where $N = k_F^2/\pi^2$ is the number of channels, $S$ is the cross-sectional area of the junction, $v_F$ is the Fermi velocity, and $k_F$ is the Fermi vector. According to Eq. (2) the critical current oscillates with a period determined by the exchange energy, and decays exponentially at distances exceeding the mean free path of the ferromagnet. Note, that in contrast to the case...
OBSERVATION OF PERIODIC $\pi$-PHASE SHIFTS IN . . .

PHYSICAL REVIEW B 73, 174506 (2006)

FIG. 2. (Color online) (a) The critical Josephson current $I_c$ as a function of the Ni-layer thickness $d_Ni$. The solid line shows the theoretical curve with $E_{ex}=200$ meV used as a fit parameter. The dots are the experimental data for all 3 sets. (b) Oscillating part of $I_c$ for set 2.

of a weak ferromagnet it is the mean free path that determines the length scale of the decay rather than the magnetic length.

The theoretical curve for the critical current at $T=4.2$ K normalized by $T^2$ [solid line in Fig. 2(a)] was calculated with Eq. (1) using the following parameters: $\nu_F = 2.8 \times 10^7$ cm/s (Ref. 19), $T_c = 8.5$ K, $E_{ex}=200$ meV, $k_F = 1.18$ Å$^{-1}$, $S=10 \times 10$ $\mu$m$^2$ and $\ell=28$ Å. Figure 2(b) shows the oscillatory part of $I_c(d)$ for set 2 on a linear scale. The magnetic length in terms of these parameters is $L_M=10$ Å.

Our result for the exchange energy is somewhat higher than the results obtained from spin-resolved photoemission spectroscopy20 where the splitting between spin-up and spin-down bands at the Fermi energy ranged between $2E_{ex}=200$ and 350 meV. The theoretical predictions for $2E_{ex}$ are typically higher and range from 600 to 850 meV. Our value of $2E_{ex}=400$ meV falls in between the above experimental and theoretical values, and is consistent with the values in Ref. 11 of $E_{ex}=220$ meV (corresponding to a magnetic length of 8.8 Å), obtained from measurements of $T_c$ in Ni-Nb bilayers.

The theoretical prediction of the minima in Fig. 2 were obtained with Eq. (1). The oscillation period for large thicknesses is thus not equal to the thickness where the first minimum in $I_c$ is predicted by the theory. This is due to the fact that the first minimum occurs at a thickness smaller than the mean free path $\ell$. Equal spacing of the minima only takes place in the regime $d_{Ni} > \ell$. The theoretical prediction for the first minimum using the above fit is $d_{min}=17$ Å.

It was recently noted14 that the critical current of junctions with a given thickness of the ferromagnet Py scatter considerably. We confirm this effect in our Ni junctions. We believe that this phenomenon is related to the domain structure of the ferromagnet. It was recently shown22 that variations in the domain configuration lead to considerable variations in $I_c$, provided that the magnetic flux through typical domains is of the order of the flux quantum $\Phi_0$. By using a magnetization for Ni of $M_s=500$ Oe, we estimate for a 50 Å $\times$ 1 $\mu$m domain cross section a flux of approximately one flux quantum. In order to average the influence of domains, we have measured different junctions with the same thickness of the ferromagnet layer.

Another option would be to magnetize the Ni layer to eliminate the domain structure by applying a strong magnetic field, as reported in Ref. 23. However, this method is problematic since it implies cooling samples with residual flux in the junction which leads to a trapped flux in the Nb banks.

We use the Landauer formula to estimate a lower limit for $T_c$ under the assumption that the entire resistance of the junction arises from the two S-F interfaces. By using a typical resistance value $R=200$ $\mu$Ω for our junctions we get $T_{min}=0.012$. The values of $T$ used for all of our sets are indeed larger than the estimated lower limit. Set 3 which included Au films instead of Cu has a value of $T$ in between the values obtained for the sets 1 and 2 with Cu, indicating that the quality of the interfaces depended in our case more on a variety of processing parameters rather than the type of normal metal. Note that $T>T_{min}$ means that there are additional interfaces which determine the junction resistance, i.e., the interfaces inside the normal metals created in the manufacturing process described above.

We emphasize that irrespective of which theory our experimental data is compared with, every $15$ Å of Ni the phase over the Nb-Ni-Nb junction changes$^{15}$ from 0 to $\pi$ or from $\pi$ to 0. This implies that we should expect the first pronounced minimum in the variation of $T_c$ at a Ni layer thickness of $d_{min}=15$ Å.

CRITICAL TEMPERATURE

Figure 3 shows the variation of $T_c$ in the Ni-Nb-Ni$(x)$-Nb-Ni multilayer structure versus the thickness of the Ni$(x)$ layer. The data contains a pronounced minimum around $x_{min}=17$ Å, which is in excellent agreement with the
absence of a pronounced local minimum in the trilayer with side in the trilayer case while they are penetrating from two critical current measurements above. Figure 4 shows super-Ni-layer thickness and for a /H9266magnetic region of thickness cause superconducting correlations are penetrating the ferro-

ture was measured for the same range of Ni /H20849and for a /H20850phase of the pentalayer be-

In order to ensure that this minimum stems from a 0 → π transition, the variation of Tc in a Ni-Nb-Ni(x) structure was measured for the same range of Ni(x) thicknesses, see Fig. 3. Note that Tc is expected to vary twice as fast for the trilayer compared to the 0-phase of the pentalayer because superconducting correlations are penetrating the ferromagnetic region of thickness xNi in Fig. 1(b) only from one side in the trilayer case while they are penetrating from two sides and meet at the midpoint in the pentalayer case. The absence of a pronounced local minimum in the trilayer with a single Nb layer, therefore, undoubtedly indicates that the minimum of Tc at 17 Å of Ni observed for the pentalayer containing two Nb layers must arise from a 0 → π transition.

In order to compare our experimental results for Tc with theory we solve the gap equation and compute Tc of our systems with the quasiclassical Green’s function technique in the diffusive approximation. Near Tc, the order parameter Δ ≈ Tc, and the Usadel equation can be linearized. We have generalized the results for symmetric trilayers by Fominov et al. in Ref. 25 to asymmetric F1-S-F2 trilayers and symmetric F2-S-F1-S-F2 pentalayers. Instead of discretizing the spatial coordinate we Fourier-series expand the order parameter and find Tc by studying the resulting eigenvalues of the gap equation. With this technique, the accuracy as well as the speed of the numerics is improved immensely compared with previously used methods.

Several material parameters serve as input to the model: The exchange field Eex of Ni, the critical temperature Tc0 of Nb in the absence of the Ni layers, and the diffusion constants of Ni (Dρ) and Nb (Dφ). The boundary conditions at the Ni-Nb interface are expressed in terms of a normalized boundary resistance γρ and the conductivity mismatch γ between the Ni and Nb materials. We have considered these two quantities as free parameters.

Our fits of the experimental data for Tc as function of Ni layer thickness with the above theory are shown as curves in Fig. 3. We use as input parameters the exchange field Eex = 200 meV and the diffusion constants Dρ = 2.8 cm²/s and Dφ = 3.9 cm²/s (with D = 1/2 ρν l) obtained from the fit of Ic in Fig. 2. The fit parameters are the bulk Nb transition temperature Tc0, the interface resistance γρ, and the materials’ conductivity mismatch γ. The fits indicate that Tc0 of the batches of trilayers and pentalayers differ, while other sample characteristics remained essentially the same. Although all samples within the trilayer set and within the pentalayer set were prepared in situ, both sets were evaporated separately. We assign the difference in Tc0 to this fact.

We have calculated Tc as a function of xNi for the pentalayer for zero phase difference and for π phase difference between the two superconductors, using the same parameters. The corresponding curves are shown in Fig. 3 as full and dashed lines. The 0 → π transition takes place where the two Tc curves cross. We note, that the fit parameter Tc0 is determined by the small thickness data points, and the remaining fit parameters γ and γρ are determined by the fitting of the zero-phase curve. Having no additional fit parameter, the 0 → π crossing at xNi = 16 Å is in remarkable agreement with the experimental data for Tc, and with the prediction of the Ic data fit. We also fit the experimental data for the trilayer with very similar interface parameters.

We would like to mention that we expect corrections to the Usadel theory when the exchange field is large. From the fit of the Ic data, where such corrections were taken into account, we see that l ≈ L, while Usadel theory works well for l ≪ L. Nevertheless, we obtain a remarkably good fit for Tc as function of xNi. This is probably due to the fact that the Ni-layers for the Tc measurements are quite thin, in which case surface disorder is relevant and justifies the use of Usadel theory. We note that the surfaces are characterized in this case by strong disorder with a large number of point

FIG. 3. (Color online) Critical temperature Tc as a function of Ni-layer thickness xNi for the trilayer and pentalayer structures. The experimental data are shown as symbols, while the curves are fits to the data with the theory discussed in the text. For the pentalayer, the highest Tc is obtained for a zero junction (full line) for xNi < 16 Å, and for a π-junction (dashed line) in the range 16 Å < xNi < 40 Å.

FIG. 4. (Color online) Superconducting transitions for several pentalayer samples. Tc is determined as a temperature at which resistance attains 1/2 of its normal state value.
contacts (high-transmission channels). This is consistent with our observation of regions with short-cuts in the samples used for the \(T_c\) measurements, which prevented us from extending our \(I_c\) measurements to \(d_{\text{Ni}} < 30\) Å.

**SUMMARY**

In summary we have demonstrated that both the critical Josephson current and the critical temperature of Nb-Ni multilayers vary with the Ni thickness with approximately the same period, 16±1 Å. We deduce from the period a magnetic length \(L_m = 10\) Å, corresponding to an exchange energy of \(E_{\text{ex}} = 200\) meV. By measuring \(T_c\) in Ni-Nb pentalayers, we have observed a \(0 \rightarrow \pi\) transition at a thickness for the central Ni layer consistent with the theoretical prediction using Usadel theory. For higher thicknesses, we see further \(\pi \rightarrow 0\) and \(0 \rightarrow \pi\) transitions in the critical Josephson current, consistent with the period in our \(T_c\) measurements and with the predictions of theory. Our results demonstrate the feasibility of using strong ferromagnetic materials in the design of Josephson devices for future applications.

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15. The observed periodicity of \(I_c\) oscillations in Fig. 2 is about half the periodicity that was deduced from the previously published data alone (Ref. 13), where the variation of the Ni thickness was made in steps of 10 Å. With the current data we are able to resolve all minima in the \(I_c\) curves as function of layer thickness.
26. The details will be published elsewhere.