Observation of Periodic π-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 π phase shifts in these devices.

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Devices of superconducting materials as for example Josephson junctions [1], 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 [2], however, opens up the spin as an additional degree of freedom [3, 4]. 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 [5].

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 π between the two superconductors for certain parameter ranges of the middle ferromagnetic material [6]. The π 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_{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 non-monotonic, 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 π over the junction. This additional degree of freedom leads to a series of $0 \to \pi$ and $\pi \to 0$ transitions, that can be observed as zero crossings of I_c and as kinks and minima of T_c with varying ferromagnetic layer thickness [7].

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 [8, 9]. 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 [10, 11] or the I_c variations [12, 13, 14] were considered as function of 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 Letter we report studies of both the critical current and the critical temperature variations as 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 $L_M = \sqrt{\hbar D_F/E_{ex}}$ 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 $E_{ex} = 200\,\text{meV}$ in Ni.

The samples for the I_c measurements had a $10\times10\,\mu\text{m}^2$ 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 Å to 75 Å in steps of 5 Å (set I) and 4 Å (set II). 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

The Nb films were sputtered using a magnetron gun and were covered in situ with a Cu (set I) or a Au (set II) layer by thermal evaporation to prevent Nb oxidation. The ferromagnet layers of Ni were e-gun evaporated in a separate vacuum chamber at a pressure of $2{\cdot}10^{-7}$ torr and

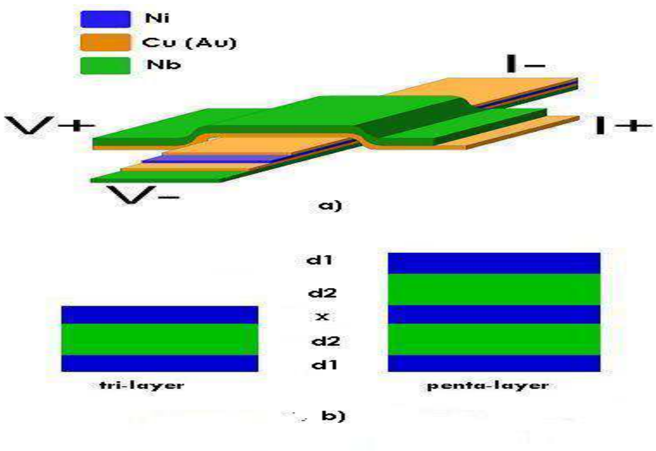

FIG. 1: Schematic layout: a) of the junctions for I_c experiments, b) of planar samples for T_c experiments.

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 ^{4}He cryostat in the range from 4.2 K down to 1.5 K.

The samples for the T_c studies were prepared by an insitu evaporation of Ni and Nb layers without photolithography, see Fig. 1b. 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(30Å)-Nb(430Å)-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(30Å)-Nb(430Å)-Ni(x)-Nb(430Å)-Ni(30Å), 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. 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.

The results of the critical current measurements are shown in Fig. 2. We have also included the set published earlier in Ref. [12]. 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 $\mathcal{T}$ of the superconductor-ferromagnet interface, which enters the calculations of I_c as a prefactor $\mathcal{T}^2$ [16]. Therefore, we have normalized the values of I_c by $\mathcal{T}^2$ varying between the sets, as indicated in the legend of Fig. 2. We use the Landauer formula to estimate a lower limit for $\mathcal{T}$, 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 $\mu\Omega$ for our junctions we get $\mathcal{T}_{min} \sim 0.012$. The values of $\mathcal{T}$ used for all of our sets are indeed larger than the estimated lower limit. 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 from 0 to π (or π to 0) [15]. This implies that we should expect the first pronounced minimum in the variation of T_c at a Ni layer thickness of $d_{\mathsf{min}} \approx 15$ Å.

The theoretical curve for the critical current at T=4.2 K normalized by $\mathcal{T}^2$ (solid line in Fig. 2) was

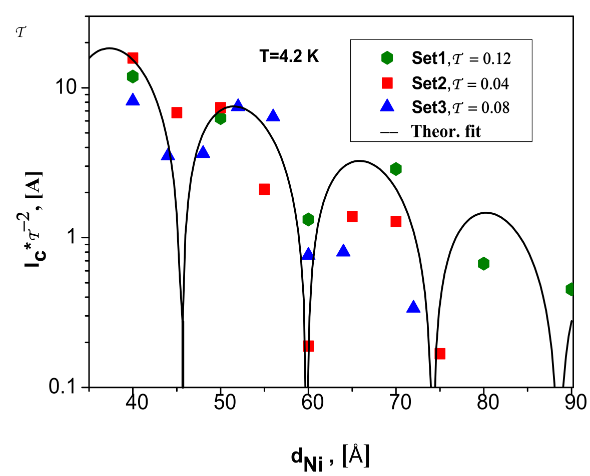

FIG. 2: The critical Josephson current I_c as function of the Ni-layer thickness d_{Ni}. The solid line shows the theoretical curve with $E_{ex} = 200$ meV used as a fitting parameter. The dots are the experimental data.

calculated with the theory of Ref. [16] using the following parameters: the Fermi velocity $v_F = 2.8 \cdot 10^7$ cm/s [17], a critical temperature of Nb $T_c = 8.5$ K, the exchange energy $E_{ex} = 200$ meV, and the mean free path $\ell = 28$ Å. The magnetic length in terms of these parameters can be determined to be $L_M = 10$ Å. Our result for the exchange energy is somewhat higher than the results obtained from spin-resolved photoemission spectroscopy where the splitting between spin-up and spin-down bands at the Fermi energy ranged between $2E_{ex} = 200$ meV and 350 meV [18]. The theoretical predictions for $2E_{ex}$ are typically higher and range from 600 - 850 meV [19]. 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 bi-layers.

The theoretical prediction of the minima in Fig. 2 were obtained with Eq. (19) in Ref. [16]. 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 ℓ. Equal spacing of the minima only takes place in the regime $d > \ell$. The theoretical prediction for the first minimum using the above fit is $d_{\mathsf{min}} \approx 17$ Å.

It was noted recently that the critical current of junctions with a given thickness of the ferromagnet Py, scatter considerably [14]. 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 shown 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 Φ_0 [20]. By using a magnetization for Ni of $M_s = 500$ Oe, we estimate for a 50 Å × 1μm 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.

Fig. 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 expected $0 \rightarrow \pi$ transition at $d_{\mathsf{min}} \approx$15-17 Å implied by the critical current measurements above. In order to ensure that the observed minimum arises from a $0 \rightarrow \pi$ transition, the variation of T_c in a Ni-Nb-Ni(x) structure was measured for the same range of Ni(x) thicknesses, see Fig. 3. For symmetry reasons, T_c should vary twice as fast for the trilayer compared to the 0-phase of the pentalayer. The absence of a pronounced local minimum in the trilayer with a single Nb layer therefore undoubtedly indicates that the minimum observed for the pentalayer containing two Nb layers must arise from a $0 \rightarrow \pi$ transition. The kink-like change of T_c at 17 Å also suggest that there is a crossing point of two phases, namely the 0− and π-phases.

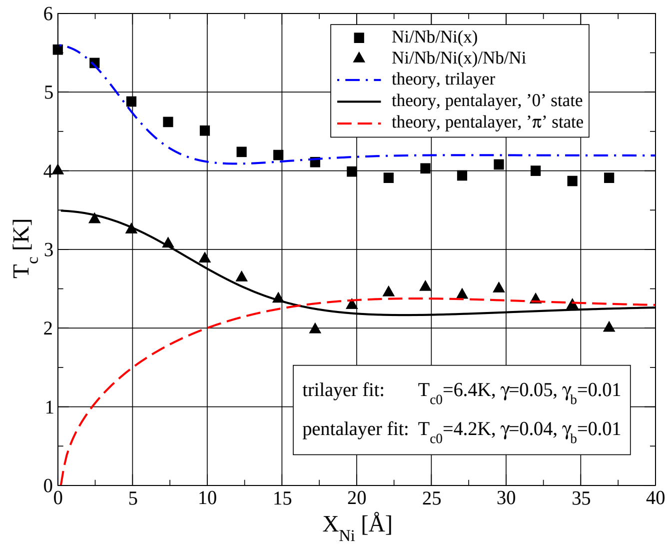

FIG. 3: Critical temperature T_c as a function of Ni-layer thickness x_{Ni} for the trilayer and pentalayer structures. The experimental data are shown as symbols, while the curves are fits to the data with the theory discussed in the text. For the pentalayer, the highest T_c is obtained for a zero junction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

In order to compare our experimental results for T_c with theory we solve the gap equation and compute T_c of our systems with the quasiclassical Green's function technique in the diffusive approximation [21]. Near T_c, the order parameter $\Delta \ll T_c$ and the Usadel equation can be linearized. We have generalized the results for symmetric trilayers by Fominov *et al.* in Ref. [22] to asymmetric F_1-S-F_2 trilayers and symmetric F_2-S-F_1-S-F_2 pentalayers. Instead of discretizing the spatial coordinate we Fourier-series expand the order parameter and find T_c by studying the resulting eigenvalues of the gap equation. With this technique [23], the accuracy as well as the speed of the numerics are improved immensely compared with previously used methods [22, 24].

Several material parameters serve as input to the model: the exchange field E_{ex} of Ni, the critical temperature T_{c0} of Nb in the absence of the Ni layers, and the diffusion constants of Ni (D_F) and Nb (D_S). The boundary conditions [25] at the Ni-Nb interface are expressed in terms of a normalized boundary resistance γ_b 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 T_c 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 $E_{ex} = 200$ meV and the diffusion constants $D_F = 2.8$ cm^2/s and $D_s = 3.9$ cm^2/s (with $D = \frac{1}{3} v_f \ell$)

obtained from the fit of I_c in Fig. 2. The fit parameters are the bulk Nb transition temperature T_{c0}, the interface resistance γ_b, and the materials' conductivity mismatch γ. The fits indicate that T_{c0} 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 T_{c0} to this fact.

We have calculated T_c as a function of x_{Ni} 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 $0 \to \pi$ transition takes place where the two T_c curves cross. We note, that the fit parameter T_{c0} is determined by the small thickness data points, and the remaining fit parameters γ and γ_b are determined by the fitting of the zero-phase curve. Having no additional fit parameter, the $0 \to \pi$ crossing at $x_{\mathsf{Ni}} = 16$ Å is in remarkable agreement with the experimental data for T_c, and with the prediction of the I_c 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 I_c data, where such corrections were taken into account, we see that $\ell \sim L_M$, while Usadel theory works well for $\ell \ll L_M$. Nevertheless, we obtain a remarkably good fit for T_c as function of x_{Ni}. This is probably due to the fact that the Ni-layers for the T_c 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 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 $d_{\mathsf{Ni}} < 30$ Å.

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_{ex}=200 meV. By measuring T_c in Ni-Nb pentalayers, we have observed a $0 \to \pi$ transition at a thickness for the central Ni layer consistent with the theoretical prediction using Usadel theory. For higher thicknesses, we see further $\pi \to 0$ and $0 \to \pi$ 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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