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Resistive transitions in Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers

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The superconducting phase transition in Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers, with superconducting (S) Nb and ferromagnetic (F) $Cu_{0.41}Ni_{0.59}$, has been experimentally studied as function of the F-layer thickness by measuring the temperature dependence of the electrical resistance $R(T)$. It is shown that the shape and the width of the $R(T)$ curves depends on the $Cu_{0.41}Ni_{0.59}$ thickness, in particular in the regime where $\pi-$coupling between the S-layers can be expected. To explain the data we developed a qualitative model which makes the interconnection between the superconducting phase transition and the 0 to π transition in SFS structures more evident.

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In the last few years the problem of interplay between superconducting (S) and ferromagnetic (F) orderings has attracted considerable attention [1 – 3]. This interest was stimulated by two closely connected experimental observations: the non-monotonic dependence of the critical temperature T_c on the F layer thickness, d_F, in SFS [4, 5] and FSFSF [6 – 8] heterostructures; and the existence of Josephson π junctions [9 – 13], structures showing a ground state with a phase difference π between the two S layers.

In this paper we studied the behavior of the dependence of the resistance R on the temperature T in the superconducting transition, of SFS trilayers with superconducting Nb and ferromagnetic $Cu_{0.41}Ni_{0.59}$, for a range of F-layer thicknesses d_F. $R(T)$ shows an unusual non-monotonic shape and broadening for the range of d_F values where π-coupling can be expected. To explain this, we developed a qualitative model which makes a natural connection between the superconducting phase transition measured in-plane and the 0-π transition in the critical current of SFS structures measured in perpendicular geometries.

Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers were deposited on Si(100) substrates in a UHV dc diode magnetron sputtering system with a base pressure less than 10^{-9} mbar and sputtering Argon pressure of $4 \cdot 10^{-3}$ mbar. The Nb and the $Cu_{0.41}Ni_{0.59}$ layers were deposited at typical rates of 0.1 nm/s and 0.04 nm/s, respectively, measured by a quartz crystal monitor calibrated by low-angle reflectivity measurements. $Cu_{1-x}Ni_x$ is a weak ferromagnetic alloy, whose magnetic strength is controlled through the Ni content, which in our films was checked by Rutherford-backscattering analysis. The Curie temperature, $T_{\rm Curie}$, and the magnetic moment per atom, μ_{at}, for this Ni concentration in $Cu_{0.41}Ni_{0.59}$ thin films were estimated to be $T_{\rm Curie} \approx 220$ K and $\mu_{at} \approx 0.12\,\mu_B$/at, respectively [14].

In order to study the dependence of the superconducting critical temperature as a function of the ferromagnetic layer thickness, $T_c(d_F)$, samples were grown with a constant Nb thickness of 14 nm and varying $Cu_{0.41}Ni_{0.59}$ thickness ($d_{\rm CuNi} = 1{-}15$ nm). The studied 22 samples were deposited in four different series, as summarized in the Table. To prevent Nb oxidation a 1 nm thick Al capping layer was deposited on top of the structures. It fully oxidizes after contact with atmosphere and does not influence the superconducting properties of the upper electrode.

The critical temperatures were resistively measured in a ^{4}He cryostat using a standard dc four-probe technique on unstructured samples. The contacts were arranged in line on the top of the samples. The distance between the current pads was about 1 cm, and the distance between the voltage pads was about 1 mm. The critical temperature T_c was defined as the point where $R = 0.1\,R_n$, with R_n the resistance value at 10 K.

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Samples deposited for each series. The thickness of the Nb layers is fixed at $d_{\rm Nb} = 14\,$nm

Series	$d_{\rm CuNi}$ (nm)
1	1.0, 2.0, 3.0, 4.0, 5.0, 6.0
2	0.5, 1.5, 2.5, 3.5, 4.5, 5.5
3	2.2, 3.2, 3.8, 4.2
4	7.0, 8.0, 9.0, 10.0, 12.0, 15.0

The transition temperature of the single Nb film with $d_{\rm Nb} = 28\,$nm was $T_{cS} = 8.55\,$K. The dependence of the superconducting transition temperature on $d_{\rm CuNi}$ is shown in Fig. 1. With increasing $d_{\rm CuNi}$, T_c first exhibits a rapid drop with an overall minimum for $d_{\rm CuNi} \approx 5\,$nm. T_c then increases slightly with $d_{\rm CuNi}$, saturating above 8 nm. This overall $T_c(d_F)$ behavior is a signature of the so-called $0 - \pi$ phase shift in S/F hybrids [2].

Apart from this standard behavior of $T_c(d_{\rm CuNi})$ we note that some data scattering is present in the thickness range $2.5\,\text{nm} < d_{\rm CuNi} < 8\ nm$. Moreover, as shown in the inset to Fig.1, in this thickness range the width of the transition curves, ΔT_c, defined as $\Delta T_c = T(R = 0.9R_n) - T(R = 0.1R_n)$, increases strongly, reaching a value of 0.6 K, while outside this range the transition curves are sharp ($\Delta T_c \approx 0.1\,$K).

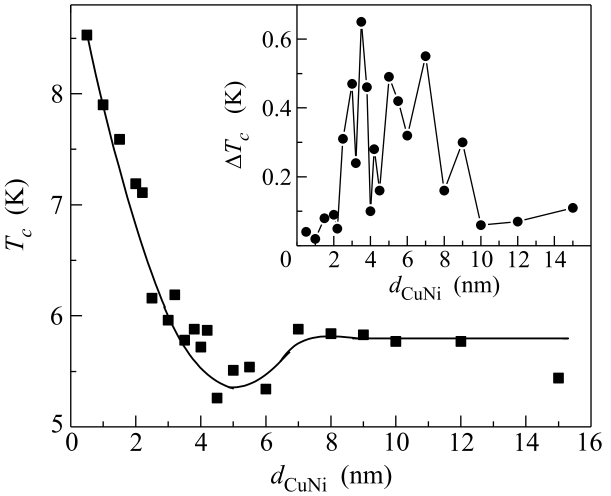

Fig.1. Critical temperature T_c of Nb/Cu$_{0.41}$Ni$_{0.59}$/Nb trilayers as a function of the ferromagnetic interlayer thickness, $d_{\rm CuNi}$. Inset: The width of the resistive transition, ΔT_c, as a function of $d_{\rm CuNi}$. The solid lines are guides to the eye

From theory [15, 16] we know that the appearance of a minimum in the $T_c(d_F)$ curve reveals the transition from the 0– to the π-phase. Similarly, a subsequent minimum would stem from a π– to 0–phase transition. In our experimental data we therefore assume that the π–phase sets in at $d_{\rm CuNi} \approx 5\,$nm and that it remains at least up to $d_{CuNi} = 15\,$nm. The minimum appears at a larger d_F value than observed in the Nb/Cu$_{0.41}$Ni$_{0.59}$ bilayers ($d_{\rm CuNi} \approx 3.5\,$nm) [14] as would be expected from the difference between bilayers and trilayers.

In the trilayer case, for $d_{\rm CuNi} \le 2\,$nm and $d_{\rm CuNi} \ge 8\,$nm, and therefore in the 0–phase and π–phase, respectively, the $R(T)$ curves show sharp transitions from normal to superconducting state as shown in Fig.2. In strong contrast, the $R(T)$ curves in the thickness interval $2.5\,\text{nm} \le d_{\rm CuNi} < 8\,$nm have a completely different form, as shown in Fig.3.

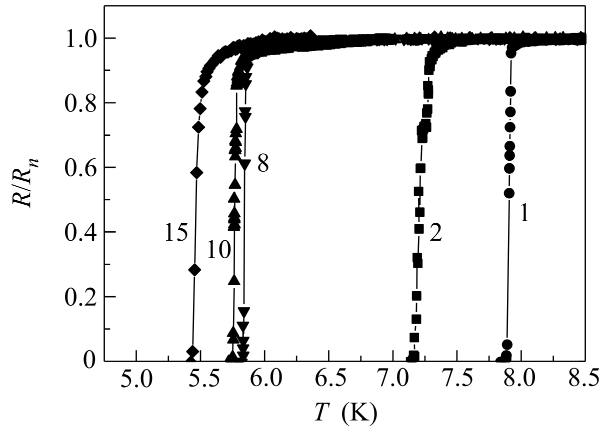

Fig.2. Typical shape of the normalized $R(T)$ dependencies for Nb/Cu$_{0.41}$Ni$_{0.59}$/Nb trilayers for $d_{\rm CuNi} \le 2\,$nm and $d_{\rm CuNi} \ge 8\,$nm. R_n is the resistance value at $T = 10\,$K. Numbers in the figure, close to each curve, indicate the thickness of the ferromagnetic layer in nm

Instead of sharp transitions with decreasing temperature the resistance first slowly decreases, followed by a sharp transition to the zero-resistance state. Note that in this Figure the results for samples from all series are shown. The $R(T)$ curve for the trilayer with $d_{\rm CuNi} = 8\,$nm is also given, to better show the difference in the $R(T)$ shapes. Note also that the crossover to broadened curves is quite sharp: between 2 nm and 3 nm in series 1, and between 1.5 nm and 2.5 nm in series 2. Indications of such broadenings have already been observed in S/F multilayers [5 – 7] but to our knowledge it was never investigated in detail. Note that for Nb/Cu$_{0.41}$Ni$_{0.59}$ bilayers from Ref.[14], in which a π–phase cannot occur, the $R(T)$ transitions are always sharp, less than 0.2 K wide for all Cu$_{0.41}$Ni$_{0.59}$ thicknesses [17]. Similarly, in other experiments on nominally Nb/Cu$_{0.41}$Ni$_{0.59}$ bilayers [18], no variations in the transition widths were detected in the region of the minimum in T_c.

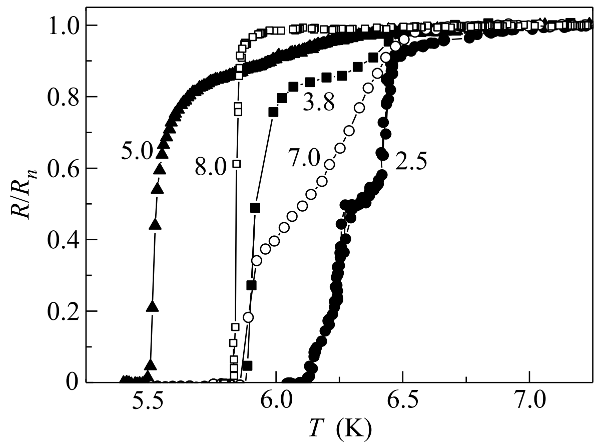

Fig.3. Typical shape of the normalized $R(T)$ dependencies for Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayers in the thickness interval $2.5\,\text{nm} \le d_{\text{CuNi}} < 8\,\text{nm}$. The $R(T)$ curve for the sample with $d_{\text{CuNi}} = 8\,\text{nm}$ is shown for comparison. R_n is the resistance value at $T = 10\,\text{K}$. Numbers in the figure, close to each curve, indicate the thickness of the ferromagnetic layer in nm

The broadening of the $R(T)$ curves disappears in a parallel magnetic field. Fig.4 shows the transition curves for the trilayer with $d_{\text{CuNi}} = 2.5\,\text{nm}$ for different values of the magnetic field applied parallel to the sample surface. It is evident from the inset that with increasing magnetic field the $R(T)$ curves become sharper.

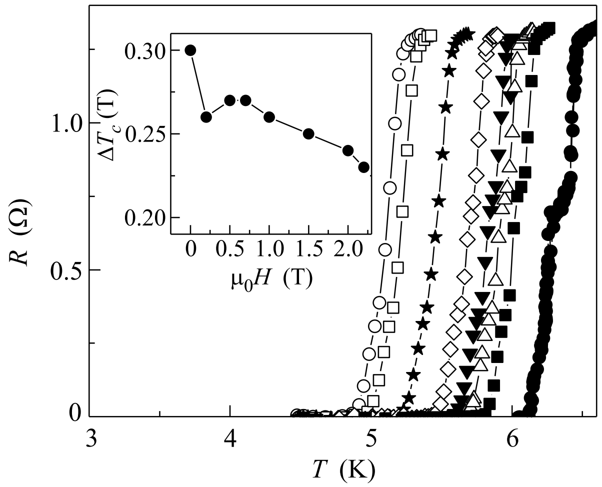

Fig.4. $R(T)$ transition curves for the Nb/$Cu_{0.41}Ni_{0.59}$/Nb trilayer with $d_{\text{CuNi}} = 2.5\,\text{nm}$ for different values of the parallel external magnetic field, H. From right to left $H = 0$, 0.2, 0.5, 0.7, 1.0, 1.5, 2.0, 2.2 T. Inset: The width of the resistive transition, ΔT_c, versus H

To explain the effect of ΔT_c broadening we consider our trilayers as a network of both SFS and SNS Josephson junctions (here N stands for normal metal). The unusual $R(T)$ shapes are a consequence of the interactions between local 0 and π junctions in the network, which may be caused by fluctuations of the samples parameters. Possible physical reasons for these variations are the roughness of the S/F interfaces (in both lateral dimensions) and local fluctuations of Cu and Ni content in the $Cu_{0.41}Ni_{0.59}$ alloy. As a rule, the decay length of superconductivity into a ferromagnetic layer and the characteristic lengths of this variation have the same scale, several nanometers, therefore the role of these interactions is much more important for thicknesses at which the entire sample goes from the 0 to the π phase, since in this regime small fluctuations in the F layer thickness or in the magnetic strength of the alloy can be crucial. A schematic representation of an SFS trilayer in this thickness interval is shown in Fig.5.

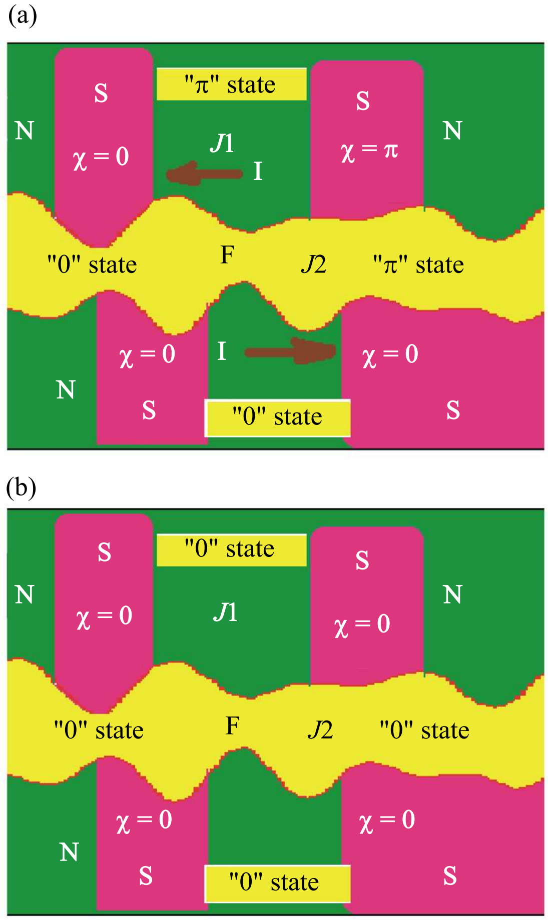

Fig.5. Sketch of the unit cell of a SNS+SFS network, representing our SFS trilayers. Junction $J1$ ($J2$) corresponds to an SNS (SFS) junction. The parameter χ represent the phase of the order parameter in each of the superconducting islands. I is the circulating Josephson current. The presence of interface roughness is taken into account. For other details see the text

At the beginning of the transition, superconductivity nucleates only in certain regions inside each S layer. Each Nb layer can be considered as to consist of S islands separated by N domains. For this reason they can be seen as a net of SNS junctions. In addition, the S layers are coupled perpendicularly via the F layer forming, in turn, a net of SFS junctions. So the entire trilayer can be seen as an SNS+SFS network. There are two types of Josephson junctions in the net, $J1$ and $J2$. The first type of coupling, $J1$, takes place across the areas in the S films which are still in the normal state at a certain value of the temperature. The ground state in these junctions corresponds to a phase difference of the order parameter between neighboring islands $\varphi = 0$, while the case $\varphi = \pi$ is a non-equilibrium state. The second type of junction, $J2$, which takes place across the ferromagnetic layer is a SFS one. It is reasonable to suppose that, due to the roughness of S/F interfaces and due to the variation of the in-plane material parameters, one of the SFS contacts can be in the $0-$state and another is in the $\pi-$ state, as shown in Fig.5a. The question is: what value of the phase difference occurs in the entire system, which contains four junctions? The problem of the ground state of this unit cell is very similar to what was already discussed for alternating array of 0 and π junctions [19].

At $T \approx T_c$, when the Josephson energy, E_N, associated to $J1$ is lower than E_F, the one related to $J2$, two junctions should be in the zero state and two in the π state. It is worth reminding that $J1$ is in the nonequilibrium state. This situation is schematically shown in Fig.5a. When T decreases the average distance between S islands in superconducting films becomes shorter, resulting in an increase of E_N while E_F remains constant, and leading to the opposite case, $E_N > E_F$. As a result all the junctions are in the 0-phase, with $J2$ in a nonequilibrium state, as shown in Fig.5b. On the other hand if the average thickness of the F layer is far from the point of 0 to π transition in the SFS structure, in the local Josephson junctions well defined correlations will dominate.

Now, to explain the unusual shape of the $R(T)$ curves it is necessary to consider also the effect of the bias current on the previous picture. At $T \approx T_c$ the application of a small measuring current generates Josephson superconducting currents having opposite directions in the top and bottom films, since at least one of the SNS junction in the cell is at nonequilibrium state and behaves as a π contact. Decreasing the temperature the volume of the S domains increases, so the average distance between them is reduced. This results in an increase of E_N and, consequently, of the circulating Josephson currents which, in turn, makes the increase of the S islands slower. These two competing mechanisms make the transition broader. If the temperature is further decreased, E_N becomes larger than E_F, the phase difference between the superconducting domains in the S layers goes to zero and the spontaneous circulating currents are switched off. In the absence of this restraining mechanism the volume of superconducting islands rapidly increases and a sharp transition into superconducting state takes place in the overall system.

The fact that the $R(T)$ curves become sharper for higher values of the magnetic field supports our model and rules out the possibility that the broad $R(T)$ curves observed in zero field were due to sample inhomogeneity, such as, for example, different T_c values between different superconducting regions. What rather happens is that, at large magnetic fields when the Josephson coupling between the Nb layers is completely suppressed, the SNS + SFS network breaks up resulting in sharp transition curves.

In conclusion, we have observed unusual broadening in the superconducting transition of SFS trilayers, which is confined to the thickness regime where the $0-\pi$ transition takes place, and which is in general absent in bilayers. A model describing the origin of this effect has been proposed. The model is based on the occurrence of SNS+SFS network in the system. An increase of the number of the SF layers in the structure should result in a further enlargement of anomalies in the transition curves. An intensive study of these $\mathrm{F[SFS]}_n\mathrm{F}$ structures with $n > 1$ is in progress.

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