NONUNIFORM SUPERCONDUCTING STATE IN SUPERCONDUCTOR/FERROMAGNET NANOSCALE LAYERED SYSTEM

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Superconducting Nb/Ni bilayers, prepared by magnetron sputtering are investigated in the range of temperatures 1.3-10K. For specimens with constant Nb layer thickness a non-monotonic behavior of the critical temperature upon increasing of the Ni layer thickness with distinct oscillations of $T_c(d_{Ni})$ of the Ni layer is found. The results are interpreted in terms of Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) inhomogeneous superconducting state in the ferromagnetic Ni layer.

Introduction

The coexistence of two antagonistic orderings, superconductivity (SC) and ferromagnetism (FM), seems to be very doubtful, because SC requires Cooper-pairing between electrons with antiparallel spin, whereas FM favors a parallel alignment of the electron spins. Nevertheless, the coexistence of both phenomena was theoretically considered by A. Larkin, Yu. Ovchinnikov, and P. Fulde, R. Ferrell [1,2]. Under strong restricted conditions for the exchange field,

$$0.71\Delta_0 < E_{exc} < 0.76\Delta_0 \qquad (1)$$

where Δ_0 is the superconducting gap, they predicted an nonuniform superconducting state in the ferromagnet with a spatially modulated pair amplitude and nonzero pairing momentum, δp. This state, called „3D-LOFF state“, is very difficult to realize experimentally because the value of the exchange field for most ferromagnets is much higher than Δ_0 (E_{exc}~ 1000 K, Δ_0 ~ 10K) .

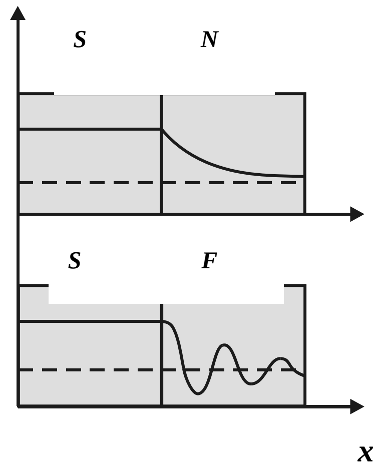

Figure 1. Proximity effect in a superconductor/normal metal (S/N) bilayer with an exponential decay of pairing function, $F_F \sim e^{-x/\xi_N}$ in the N-layer, and in a proximity coupled superconductor/ferromagnet (S/F) bilayer with oscillating $F_F \sim \cos(x/\xi_F)$ within the F-layer.

A more realistic system for observation of LOFF-like state was proposed by A. Buzdin and Z. Radovich [3,4] - a planar geometry, i.e. S/F layered system, where a quasy-1D LOFF-like state can appear. They proposed to separate superconductor and ferromagnet, then the strict limitation (1) for the exchange field will be not important. Such “proximity-coupled” S/F system is shown in Fig.1. For an S/N- proximity coupled system a monotonous decrease of the paring function, ***F***, within the N-layer [5] leads to a monotonous decrease of the critical temperature of the S/N bilayers with the N-layer thickness increasing, as was confirmed in a lot of experimental works. In contrast to S/N systems, for the S/F coupled bilayers a very unusual phenomenon was predicted by A.Buzdin: an oscillating dependence of T_c as a function of the magnetic layer thickness, d_F. Due to the proximity effect, Cooper pairs diffuse from S-layer into the ferromagnetic layer where are subjected to the exchange field. A maximum of $T_c(d_F)$ occurs when the period of the Usadel function describing the pair amplitude, ***F***, matches the thickness of the ferromagnetic layer. Further theoretical investigations [6] predict different behavior of $T_c(d_F)$: from a monotonic suppression of T_c and its saturation at some d_F up to re-entrant superconductivity depending on the interface transparency. In spite of the well developed theory of S/F layered systems, there exist only a few experimental investigations of $T_c(d_F)$ with reliable oscillations of $T_c(d_F)$ observed (see [7] and the References therein). Crucial difficulties arose in these experiments. We consider in the present work the experimental conditions necessary for reproducible observation of the LOFF-like 1D superconducting state in S/F layered system with distinct T_c-oscillations.

Sample preparation and characterization

Among different possible S/F–couples for sample preparation we have chosen Nb and Ni. The reason of the choice was a very small mutual solubility of these two metals, less than 4 at.% at ambient conditions [8], what leads to obtaining of a well resolved clean S/F-interface. On the other hand, we did not take absolute immiscible metals, like Nb and Gd, which could give an island-like growth of the F-layer and as a result, a poor quality rough S/F interface. The Nb/Ni samples have been prepared by DC magnetron sputtering on flame-polished glass substrates kept at room temperature. Pure Nb (99,99%, ') and Ni (99,99%) were used as sputtering targets.

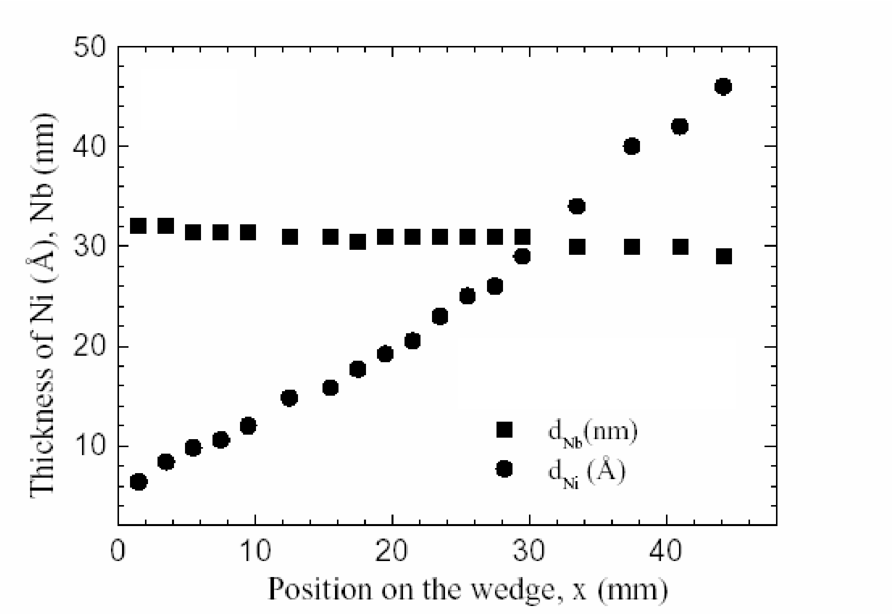

Figure 2. The results of RBS measurements for the thickness of Nb and Ni layers (sample set from Nb/Ni-wedge with d_{Nb}=const=31nm, d_{Ni}=variable).

The glass substrate of 10x75 mm^2 was put into the chamber symmetrically with respect to the centrum of the Nb target. Then, a wedge-shaped Ni layer was deposited by shifting the substrate away from the symmetry axis of the Ni target. The resulting wedge sample was cut across the wedge to 1.5 mm wide strips using a diamond cutter. Following this routine we obtained a batch of Nb/Ni samples with constant Nb layer thickness and variable Ni layer thickness. Platinum 50-μm wires were attached by silver paste for four-contact resistance measurements.

The X-ray small angle reflectivity scans ($2\theta < 10^0$, CuK_α radiation) were used to determine the layer roughness. Well resolved weakly damped oscillations of the reflectivity as a function of the angle with more than 15 periods of oscillations were detected, giving the evidence of high quality of the layers with smooth surfaces and interfaces. For a quantitative analysis of the X-ray data we used a simulation program, based on the Parratt formalism [9]. Fitting of the X-ray data gives the value of the layer thickness and the roughness σ_{rms} as the mean square deviation of the layer thickness from an "ideally" smooth layer, the value of σ_{rms} for the investigated samples was less than 0.3 nm.

For the precise thickness measurements we used Rutherford backscattering spectrometry, giving the possibility to determine the absolute thickness of Ni layers at the level of 1 nm with an accuracy of 0.03 nm. Details of the RBS measurements are described in [7]. The results of thickness determination by RBS for one of investigated Nb/Ni-wedge samples are presented in Fig.2.

Results and discussion

Resistive transitions for one batch of the Nb/Ni specimens are presented in Fig.3. The width of transition ($0.1R_N$-$0.9R_N$ criteria) for all investigated samples was less than 0.1 K, what allows determination of the critical temperature from the midpoint of the resistive transitions with a necessary accuracy. Figure 4 shows the dependence of the superconducting critical temperature T_c on the thickness of the ferromagnetic Ni layer for the samples with the thickness of Nb layer fixed at 31 nm. The critical temperature drops sharply when increasing d_{Ni} up to 1.2 nm, then it passes through a minimum and increases till the thickness d_{Ni} approaches ~ 2.5 nm.

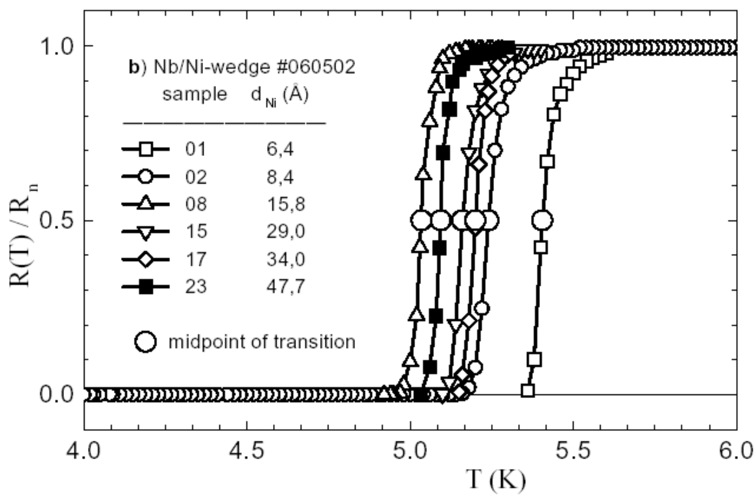

Figure 3. The resistive transitions R(T) normalized to R_n=R(T=10 K) for some of the measured samples from Nb/Ni-wedge with d_{Nb}=const=31 nm, d_{Ni}=variable.

T_c (d_{Ni}) demonstrates an oscillatory behavior as a function of the Ni layer thickness. This oscillatory behavior is the evidence of the 1D-LOFF-like superconducting state in the investigated Nb/Ni samples.

As it was mentioned above, the proximity effect in S/F system has a crucial difference from that of the superconductor/normal metal (S/N) system. In a usual S/N system the pairing function exponentially relaxes deep into the N-layer on the scale of the coherence length $\xi_N = (\hbar D_N /2\pi k_B T)^{1/2}$, here D_N is the diffusion constant of the normal metal, k_B is the Bolzman constant. In S/F layered system, in contrast to S/N proximity coupled layers, the pairing function in a pure ferromagnet ($l_F > \xi_F$) oscillates on a distance of the magnetic coherence length, $\xi_F = (\hbar v_F /E_{ex})$, and relaxes deep into the F-layer on the scale of electron mean free path, l_F. In strong ferromagnets like Ni, Fe the oscillation period of the pairing function can be a few times shorter than the decay length l_F. If the thickness of the F-layer is smaller than or comparable to the l_F, the weakly damped pairing function wave incident on the S/F interface, will interfere with the wave reflected from the opposite surface of the F-layer. Using an analogy with light, the F-layer works like a Fabry-Perot interferometer, which can be highly reflective or almost transparent depending on the relation between the wavelength and the thickness of the interferometer. Thus, due to the interference, the pairing function flux will be modulated as a function of ferromagnet layer thickness d_F. As a result, the coupling between S and F layers is modulated, and the

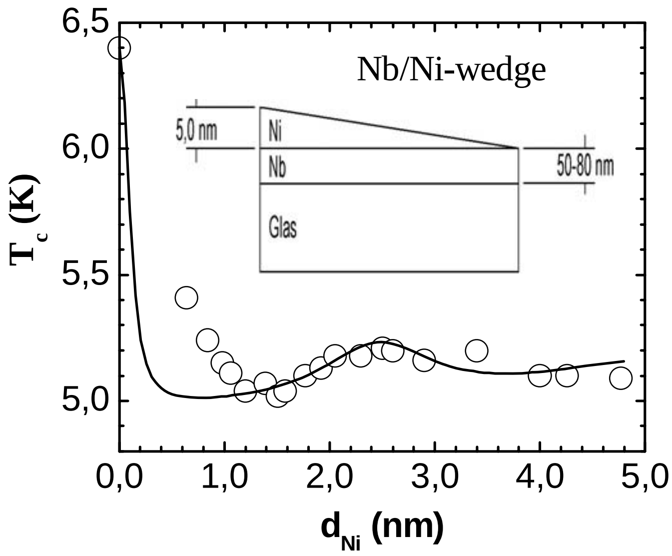

Figure 4. The dependence of the superconducting transition temperature T_c on the Ni layer thickness d_{Ni} for the sample set from the Nb/Ni-wedge with d_{Nb} =31 nm. The solid line demonstrates the best fit by the theory [6], the solid line is calculated using the parameters: T_{c0} =6.4 K, ξ_{BCS}(Nb)=42 nm, ξ_S =6.2 nm, $(N_F v_F)/(N_S v_S)$ =0.51, ξ_F/l_F =0.5, T_F =2.0, ξ_F =0.88 nm.

superconducting T_c oscillates as a function of d_F. We have observed this type of oscillations in investigated Nb/Ni samples. For the quantitative comparison with the S/F-proximity theory we used the model developed by L. Tagirov [6] for bilayers with a finite S/F interface transparency T_F. In frame of the theory [6] superconducting transition temperature of the S/F-sandwich can be determined as a solution of equation

$$\ln t_c + Re\psi(1/2 + \rho/t_c) - \psi(1/2) \ , \qquad (2)$$

where $Re\psi$ is the real part of the digamma function, $t_c = T_c/T_{c0}$ is the reduced transition temperature, T_{c0} is the transition temperature of single superconducting layer . Here the complex-valued pair-breaking parameter ρ is defined as $\rho = (k_S\xi_S)^2/2$, and k_S is the wave number of the pairing function propagation in the S-layer, which satisfy to the solution of equation:

$$(k_Sd_S)tan(k_Sd_S) =$$
$$= (N_Fv_Fd_S\xi_{BCS})/N_Sv_S\xi_S^2)[tanh(k_Fd_F)/\{(1-i\xi_F/l_F)^{1/2} + (2/T_F)tanh(k_Fd_F)\}] \quad , \qquad (3)$$

where N_F , N_S are the density of states at the Fermi level in F and S layers, respectively, v_F ,v_S is the Fermy velocity, $\xi_{BCS} = (1.78\hbar v_S)/(\pi^2 k_B T_{c0})$, $\xi_S=(2/\pi)\xi_{GL}(0)$, Ginzburg-Landau coherence length $\xi_{GL}(0)$ is determined from the slope of the upper critical field, $\xi_{GL}(0)=[(-dB_{c2}(T)/dT)(2\pi T_c/\phi_0]^{-1/2}$, and T_F is the quantum mechanical interface transparency parameter, which equals infinity for the perfectly transparent S/F interface, or is equal to zero for the completely reflecting interface. It takes into account the reflection of electrons from the interface caused by a mismatch of the Fermi momentum of the contacting metals.

According to the S/F proximity theory [6] three main regimes for T_c behavior can be observed in S/F bilayers depending on the thickness of the superconducting layer: a) for a large thickness, $d_S >>\xi_S$ the superconducting T_c can oscillate as a function of the ferromagnetic layer thickness d_F [7]; b) for a thinner superconducting layer, when $T_c \sim (0.2\text{-}0.3)T_{c0}$ the regime of re-entrant superconductivity can be realized (preliminary attempt of such behavior reported in [10]); c) for thinner superconducting layers the superconducting T_c falls down to zero upon increasing d_F ,.

One can conclude, that our experimental data belong to the regime a), demonstrating distinct oscillations of T_c (d_F). Fitting of the experimental results to the S/F-proximity effect theory [6], shown by solid cure in Fig.4 (with mediate value of the adjust parameter of interface transparency T_F = 2.0), shows a rather good agreement in the relevant range of Ni layer thickness above 1nm, but there is a deviation in the range of small thickness below 1 nm. The possible reason responsible for this deviation may be the existence of the magnetically "dead" layer at the S/F interface. As demonstrated in [11] the Nb/Fe couple always has an interdiffused layer at the S/F interface, which is magnetically indifferent, i.e. the local magnetic moment disappears when increasing the number of Nb neighbors surrounding the Fe ion in the cluster. The same physical picture is realized also in V/Fe couples [12]. A magnetically “dead-layer” has also been observed at the V/Ni interface [13], where the vanishing of the Ni layer magnetic moment at d_{Ni} below 1 nm has been declared. Finally, from a detailed study of Nb/Ni multilayers prepared by magnetron sputtering on a sapphire substrate [14] the authors concluded that below a Ni layer thickness of 1.4 nm the magnetic momentum in the Ni layer disappears due to the interdiffusion of Nb - Ni and no ferromagnetic order is present. This thickness is close to the thickness of d_{Ni} ~ 1.0 nm in our study (see Fig.4), where the theory deviates from the experimental points. The suppression of superconductivity by a nonmagnetic dead-layer is weaker than by ferromagnetic Ni, and $T_c(d_{Ni})$ decreases slowly which meets our experimental observation.

Conclusion

The proximity effect in Nb/Ni bilayers prepared by DC magnetron sputtering on smooth glass substrates was investigated. We observed distinct oscillations of the superconducting critical temperature for specimens with constant Nb layer thickness upon increasing the thickness of the Ni layer as an evidence of 1D-LOFF- like inhomogeneous superconducting pairing in the ferromagnetic Ni layer. We can formulate the necessary conditions for experimental realization of the 1D-LOFF like state in S/F layered structures:

- The sputtering technique with a high deposition rate has advantages in comparison with MBE process to fabricate S/F samples showing oscillating behaviour of $T_c(d_F)$;
- The S/F couples should not consist of immiscible metals, but of metals with restricted solubility and narrow composition ranges of intermetallic compound formation for obtaining of a smooth S/F interface;
- The substrate quality and deposition rate should ensure the S/F interface roughness less than the F-layer thickness.

S/F layered system prepared under those conditions will demonstrate 1D-LOFF-like superconducting state.

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