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Quasi-One-Dimensional Fulde-Ferel-Larkin-Ovchinnikov-Like State in $Nb/Cu_{0.41}Ni_{0.59}$ Bilayers

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In a ferromagnet (F) being in contact with a superconductor (S) an unconventional finite-momentum pairing of electrons forming Cooper pairs occurs. As a consequence, interference effects of the pairing wave function, leading to an oscillation of the critical temperature for increasing F-layer thickness in S/F bilayers, including extinction and recovery of the superconducting state, were predicted by theory. We observed experimentally all types of this behavior, calculated theoretically, in $Nb/Cu_{1-x}Ni_x$ bilayers ($x = 0.59$) of nanometer film thickness, prepared by magnetron sputtering (utilizing a moving magnetron deposition technique to provide a superb homogeneity of the ultrathin Nb layers), including a double extinction of superconductivity, giving evidence for a multiple reentrant state.

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1. Introduction. Singlet superconductivity and ferromagnetism usually not co-exist in a homogeneous material. The reason is that the superconducting state is established by Cooper pairs which are pairs of electrons with opposite momenta and antiparallel spins. Contrary, the ferromagnetic state is built up by electrons with parallel aligned spins. Thus, singlet superconductivity and ferromagnetism are long-range orders which are expected to exclude each other.

Nevertheless, Fulde-Ferrell [1] and Larkin-Ovchinnikov [2] (FFLO) predicted superconducting pairing to occur in the presence of an exchange field, *i.e.* on a ferromagnetic background, but with a non-vanishing momentum of the pair and in an extremely narrow range of parameters [3]. For superconductor/ferromagnet (S/F) layers, Buzdin and Kupriyanov [4] predicted an FFLO-like state, as the consequence of a S/F proximity effect, *i.e.* a pair amplitude establishing in the F-material by a penetration of electron pairs through the S/F interface. Due to the non-vanishing pairing momentum, the Cooper-pair wave function oscillates in the ferromagnetic layer. Interference effects between the part of the incoming pair amplitude reflected at the S/F boundary and the pair amplitude reflected at the outer surface of the ferromagnetic layer, lead to an oscillating behavior of the critical temperature T_c of the layered system as a function of increasing F-layer thickness d_F.

The phenomenon was studied experimentally for different S/F layered systems [5 – 10]. It turns out that samples made by magnetron sputtering due to their high quality surface and boundary properties are most suitable to study this type of S/F proximity-effect physics [11].

Moreover, an extremely accurate method to measure the thickness of the layers, especially of the very thin F layer has to be applied. Rutherford backscattering spectrometry (RBS) is such a method. For the first time it was applied in proximity effect investigations in Ref. [11].

Not only magnetic elements, but also ferromagnetic alloys were used as F-layer material [12 – 15]. In this case, the exchange energy of the material can be adjusted by the alloy composition. For a diluted magnetic alloy, thicker F layers can be used, which are much easier to handle. Now, RBS is applied to measure the composition of the deposited alloy layer and its thickness [15].

Concerning the theory, Radovic et al. [16, 17] gave a first description. Advanced calculations by Aarts et al. [18], Tagirov [19] and Fominov et al. [20] consider the

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finite transparency of the S/F interface. Single-mode [18, 19] and also multimode [20] solutions for the pair amplitude in S/F layers were studied. Not only T_c oscillations were predicted by these theories. Most spectacular is the result that superconductivity may vanish in a certain range of the F-layer thicknesses, and re-appear for a further increase of the thickness of the F-layer. This is a reentrant behavior of superconductivity.

Recently, aside the most pronounced T_c oscillations ever measured in S/F proximity effect systems to date, such reentrant behavior could be observed by us for Nb/$Cu_{41}Ni_{59}$ bilayers [15]. These experiments provided the first convincing evidence of a reentrant behavior of the superconducting state in S/F layers. By detailed studies we were able to realize experimentally all types of non-monotonic and reentrant behavior of superconductivity predicted by the theory: from very moderate suppression with a shallow minimum in the $T_c(d_F)$ dependence, over expressed critical temperature oscillations, till reentrant behavior. Even an indication of a multiple reentrance was found [15, 21, 22]. For a brief review of our work, containing an introduction into the basic physical mechanisms of the quasi-one-dimensional FFLO-like state in S/F bilayers see Ref. [23].

2. Experimentals. Figure 1 shows our wedge technique [11] to fabricate S/F bilayers of different thickness in the same run, yielding the same surface and boundary properties. First, a superconducting layer of constant thickness (here Nb), then a film of steadily increasing thickness (the wedge) of the F material (here $Cu_{41}Ni_{59}$ alloy), are deposited. Cutting into stripes across the thickness gradient yields a series of up to about 40 samples with constant S but different F-layer thickness. These are separately measured to determine their critical temperature.

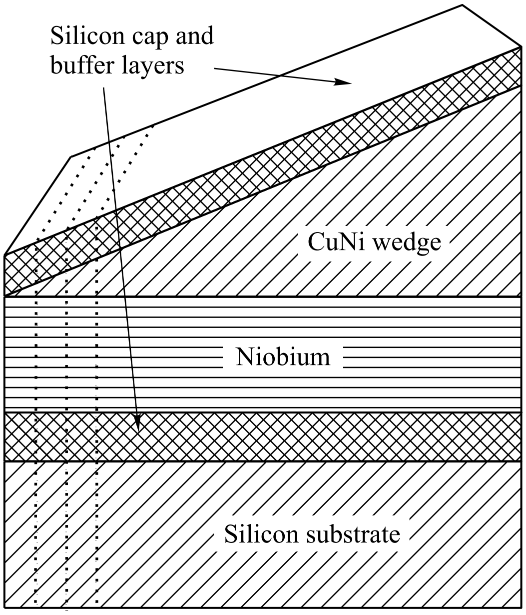

Fig. 1. Sketch of a Nb/CuNi-wedge sample with Nb layer of constant thickness, covered by a CuNi wedge. The sample is cut into stripes along dotted lines to produce a series of up to 40 specimens with variable ferromagnetic CuNi-alloy layer thickness

The thickness of the layers and their alloy composition were determined from RBS investigations (see Ref. [15] for details), demonstrating constant S and steadily increasing F-layer thickness d_F for the series of specimens investigated.

In more detail: The S and F films were prepared by magnetron sputtering on commercial (111) silicon substrates at room temperature. Pure argon was used as sputter gas. Three targets, Si, Nb and $Cu_{40}Ni_{60}$, were pre-sputtered for 10–15 minutes to remove contaminations. Moreover, Nb acts as a getter material, reducing the residual gas pressure in the chamber. Next, a silicon buffer layer was deposited by using a RF magnetron. This generates a clean interface for the Nb layer deposited subsequently. To get flat high-quality Nb layers (thickness 5–15 nm) by DC magnetron sputtering, we rotated the target around the symmetry axis of the vacuum chamber during deposition [15]. A dc-motor setup moved the full-power operating magnetron along the Si substrate of 80 × 7 mm size so that the surface was homogeneously sprayed with Nb. The average growth rate of the Nb film was about 1.3 nm/sec, whereas the deposition rate for a fixed, non-moving target would be about 4–5 nm/sec.

The wedge-shaped ferromagnetic layer was then deposited, utilizing the intrinsic spatial gradient of the deposition rate [11, 15]. The $Cu_{40}Ni_{60}$ target was RF sputtered with a rate of 3–4 nm/sec. Practically the same composition of the alloy was found in the film. A degradation of the resulting Nb/$Cu_{41}Ni_{59}$ bilayers at atmospheric conditions was prevented by a silicon cap of about 5–10 nm thickness.

Then samples of equal width (about 2.5 mm) were cut to obtain a batch of S/F-bilayer strips for $T_c(d_F)$ determination by four-probe resistance measurements. The critical temperature was determined from the midpoints of the resistive transitions.

The measurements were performed using conventional ^{4}He and ^{3}He cryostats as well as a ^{3}He/^{4}He dilution refrigerator down to 40 mK.

3. Results and Discussion. The superconducting $T_c(d_{\mathrm{CuNi}})$ measurements are shown in Fig.2. A clear non-monotonic behavior is observed when varying the ferromagnetic layer thickness. For fixed thickness of the

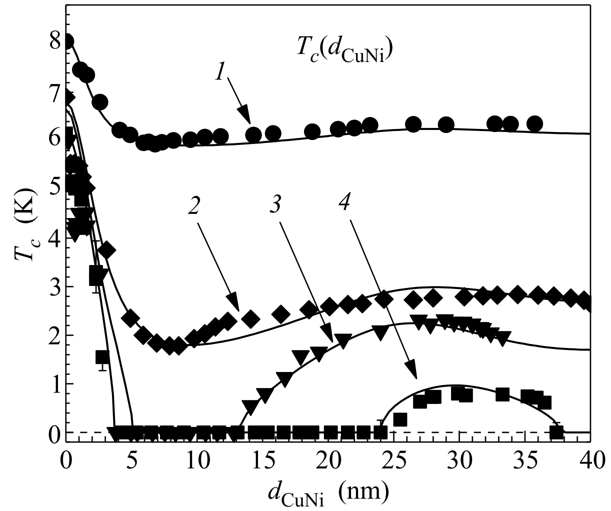

Fig.2. Dependence of the superconducting transition temperature T_c on the ferromagnetic layer thickness for sample series with different fixed thickness of the superconducting Nb layer: *1* – $d_{Nb} \simeq 14.1$ nm (S23), *2* – $d_{Nb} \simeq 7.8$ nm (S22), *3* – $d_{Nb} \simeq 7.3$ nm (S15), *4* – $d_{Nb} \simeq 6.2$ nm (S21). The solid lines are calculated within the theory [11, 19] (see values of parameters in the text)

Nb layer of $d_{\mathrm{Nb}} = 14.1$ nm (sample series S23) the T_c oscillation is flat with a shallow minimum. Reducing d_{Nb}, the transition temperature drops more sharply for increasing d_{CuNi} and the minimum becomes more expressed, as shown *e.g.* for series S22 with $d_{\mathrm{Nb}} = 7.8$ nm. Then, only a very small further reduction of d_{Nb} to 7.3 nm is enough to show the result of reentrant superconductivity in S/F bilayers mentioned above (first published by us in Ref. [15]). By reducing d_{Nb} to 6.2 nm, a double suppression of superconductivity is obtained, giving evidence for the multiple reentrant behavior predicted by the theory.

The theoretical curves were fitted following the strategy described in Refs. [11, 15, 22]. Throughout, the range for the superconducting coherence length ξ_S between 6.2 nm and 6.7 nm was applied (see the discussion in Ref. [22]) instead of $\xi_S \sim 10$ nm, as used in our previous publications on $Nb/Cu_{41}Ni_{59}$ bilayers. In detail, the fitting parameters are as follows for curves S15, S21, S22, and S23, with $T_{c0,\mathrm{Nb}}(d_{\mathrm{CuNi}} = 0\,\mathrm{nm}) = 6.67$, 6.2, 6.85, and 8.0 K, respectively (taken from [22]): $\xi_S = 6.3$, 6.1, 6.5, and 6.6 nm; $N_F v_F / N_S v_S = 0.22$ for all; $T_F = 0.67$, 0.65, 0.61, and 0.44; $l_F/\xi_{F0} = 1.3$, 1.1, 1.1, and 1.1; $\xi_{F0} = 9.5$, 11.2, 10.7, and 10.8 nm. Here, ξ_S, is the superconducting coherence length, $N_F v_F / N_S v_S$ is the ratio of the Sharvin conductances at the S/F interface, T_F is the interface transparency parameter, l_F, is the electron mean free path of conduction electrons in the ferromagnet, and ξ_{F0} is the magnetic coherence length [11].

The calculated curve for sample series S21 with a double extinction of superconductivity does not yield a further reentrance of superconductivity for higher values of d_{CuNi}. A slightly thicker Nb layer ($d_{\mathrm{Nb}} \approx 6.3$ nm), however, gives a prediction of a further island of superconductivity above $d_{\mathrm{CuNi}} \approx 51$ nm.

4. Conclusion. Our investigations clearly show the existence of a quasi-one-dimensional FFLO-like state in $Nb/Cu_{41}Ni_{59}$ bilayers. The non-monotonic behavior of the critical temperature predicted by theory, including the phenomenon of reentrant superconductivity with evidence for a multiple reentrant state, could be demonstrated experimentally.

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