Double re-entrance of superconductivity in superconductor/ferromagnet bilayers

A S Sidorenko[1], **V Zdravkov**[1], **J Kehrle**[2], **R Morari**[1], **G Obermeier**[2], **S Gsell**[2], **M Schreck**[2], **C Müller**[2], **V Ryazanov**[3], **S Horn**[2], **L R Tagirov**[4], **and R Tidecks**[2]
[1]Institute of Electronic Engineering and Industrial Technologies ASM, Kishinev, MD2028, Moldova
[2]Institut für Physik, Universität Augsburg, D-86159 Augsburg, Germany
[3]Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, 142432, Russia
[4]Solid State Physics Department, Kazan State University, Kazan, 420008, Russia

E-mail: `Anatoli.Sidorenko@int.fzk.de`

Abstract. We report on the first observation of a double suppression of superconductivity in a superconductor/ferromagnet layered system. The result was obtained using a superconductor/ferromagnetic-alloy bilayer of Nb/Cu$_{41}$Ni$_{59}$ with $d_{Nb} \simeq 6.2$ nm. As the thickness of the ferromagnetic alloy gradually increases, the superconducting transition temperature T_c drops sharply until a complete suppression of superconductivity is observed at $d_{CuNi} \simeq 2.5$ nm. At further increase of the Cu$_{41}$Ni$_{59}$ layer thickness, superconductivity restores at $d_{CuNi} \simeq 24$ nm. Then, with a subsequent increase of d_{CuNi}, superconductivity vanishes again at $d_{CuNi} \simeq 38$ nm. Our experiments give evidence for the realization of the quasi-one dimensional Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) like state in the ferromagnetic alloy layer.

1. Introduction

In superconductor-ferromagnet metal (S/F) contacts the superconducting pairing wave function not only exponentially decays into the F metal, as in the superconductor/normal metal (S/N) proximity effect, but simultaneously oscillates [1]. A variety of novel physical effects caused by these oscillations were predicted ([1] and references therein). Some of them have already been observed experimentally: non-monotonous and reentrant behavior of the superconducting critical temperature T_c as a function of the F metal layer thickness, Josephson junctions with intrinsic π-phase shift across the junction, inverted differential current-voltage characteristics. In this work we report on results of superconducting proximity effect experiments on Nb/Cu$_{1-x}$Ni$_x$ $x = 0.59$ bilayers. After a destruction by interference effects of the superconducting pairing wave function and a subsequent recovery, a second suppression of superconductivity is found, giving an impressive experimental evidence for a quasi-one dimensional Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) [2, 3] like state in the ferromagnetic layer.

The origin of FFLO state physics lies in the exchange splitting of the conduction band in a ferromagnetic metal. One of the singlet Cooper-pair electrons occupies the majority subband, for example spin-up, while the other one resides at the spin-down, minority subband. Although the pairing occurs with opposite directions of the wave number vectors of the electrons, their

© 2009 IOP Publishing Ltd

absolute values are not equal due to the exchange splitting. The resulting pairing state acquires a finite momentum of $\hbar Q_F = E_{ex}/v_F$, where $E_{ex} \ll E_F$ is the energy of the exchangesplitting of a free-electron-like, parabolic conduction band, E_F is the Fermi energy, and v_F is the Fermi velocity. Then, the pairing function of this state does not simply decay as it would be in a non-magnetic metal, but in addition oscillates on a wavelength scale given by $\lambda_{F0} = 2\pi\hbar v_F/E_{ex}$ for the case of a "clean" ferromagnet.

The oscillation of the pairing wave function in the F-metal is the reason for an oscillatory S/F proximity effect, yielding a non-monotonous, oscillating dependence of the superconducting critical temperature T_c on the ferromagnetic layer thickness d_F. The phenomenon can be qualitatively described using the analogy with the interference of light in a parallel-sided plate of glass with a mirror coated back side, at normal incidence [4]. As the interference conditions change periodically between constructive and destructive upon changing the thickness of the plate, the flux of light through the interface of incidence is modulated. In a layered S/F system, the pairing function flux crossing the S/F interface depends on the ferromagnetic layer thickness d_F because of the pairing function interference. As a result, the coupling between the S and F layers in a series of samples with increasing d_F is modulated, and the superconducting T_c oscillates as a function of d_F. The amplitude of T_c oscillation depends sensitively on the superconducting layer thickness.

The aim of our experiments was to realize the most spectacular evidence for the oscillatory proximity effect that would be the observation of the multiple reentrant behavior of the superconducting T_c predicted theoretically [5, 6, 7]. To achieve the goal we fabricated S/F samples using niobium as a material for the superconducting layer, and the $\mathrm{Cu_{41}Ni_{59}}$ alloy as a material for the ferromagnetic layer. The choice of alloy instead of a conventional elemental ferromagnet has the following advantages: the oscillation length λ_{F0} in strong ferromagnets, like iron, nickel or cobalt, is extremely short , because the exchange splitting energy E_{ex} is usually in the range 0.1-1.0 eV. Weak ferromagnets with an order of magnitude smaller exchange splitting of the conduction band allow the observation of the effect at much larger range of thicknesses d_F which can be easier controlled and characterized.

2. Experimentals

The S/F samples were prepared by magnetron sputtering on commercial (111) silicon substrates at room temperature. Three targets, Si, Nb and $\mathrm{Cu_{40}Ni_{60}}$, were pre-sputtered for 10-15 minutes to remove contaminations and reduce the residual gas pressure (by Nb as a getter material) in the chamber. Then, a silicon buffer layer was deposited using a RF magnetron, to generate a clean interface for the subsequently deposited niobium layer. To obtain flat, high quality Nb layers, the full-power operating magnetron was moved along the silicone substrate of 80×7 mm^2 size. Thus, the surface was uniformly sprayed with the material at a reduced deposition rate compared to a fixed , non-moving target. To prepare samples with variable thickness of the ferromagnetic layer we deposited a wedge-shaped ferromagnetic film utilizing the intrinsic spatial gradient of the deposition rate. The resulting Nb/$\mathrm{Cu_{41}Ni_{59}}$ bilayers were coated by a silicon cap of about 10 nm thickness to prevent degradation in an ambient atmosphere. Rutherford backscattering spectrometry (RBS) has been used to evaluate the thickness of Nb and $\mathrm{Cu_{1-x}Ni_x}$ layers as well as to check the composition of Cu and Ni in the deposited alloy layers (Fig. 1, a sketch of the layers stack is presented in the inset). After cutting the wedge into strips across the thickness gradient, we obtained 36-40 samples with variable $\mathrm{Cu_{41}Ni_{59}}$ layer thickness in the range $d_{CuNi} \approx 1-35$ nm, prepared at identical conditions in a single deposition run. Aluminum wires of 50 μm in diameter were then attached to the strips by an ultrasonic bonder for four-probe resistance measurements. For further sample preparation details see Ref. [8].

The superconducting critical temperature T_c was determined from the midpoints of resistive transitions curves R(T). The resistance measurements were performed by the DC four-probe

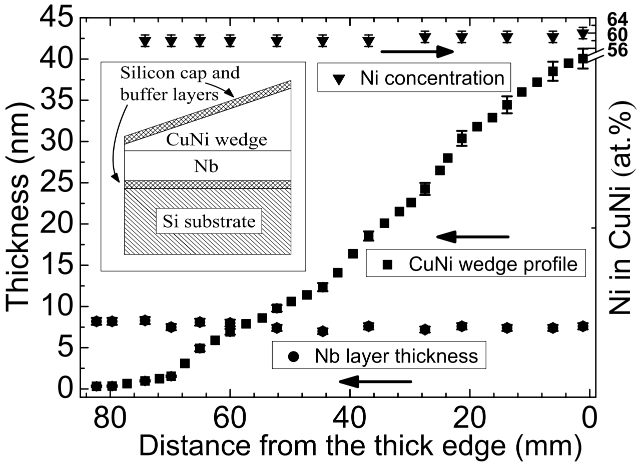

Figure 1. RBS profiles of the layers and sketch of the layers stack (inset) for the S22 series.

method using a 10 μA sensing current in the temperature range 0.4 K-10 K when measuring with an Oxford Instruments “Heliox” ^{3}He cryostat, and a 2 μA sensing current in the range 40 mK-1.0 K when measuring in an Oxford Instruments dilution refrigerator “Kelvinox”. The width of transition (0.1R$_N$-0.9R$_N$ criteria, where R$_N$ is the normal state resistance just above T_c) for most of the investigated samples was below 0.2 K, that allows to determine the T_c with a good accuracy.

3. Results and discussion

Figure 2 demonstrates the dependence of the superconducting transition temperature on the Cu$_{41}$Ni$_{59}$ layer thickness. The thickness of the Nb layer is fixed for each of the three series of samples presented. The transition temperature T_c for the specimens with $d_{Nb} \approx 14.1$ nm (S23) reveals a non-monotonous behavior with a shallow minimum at about $d_{CuNi} \approx 7.0$ nm. For the thinner niobium layer ($d_{Nb} \approx 7.8$ nm, S22) the transition temperature shows a pronounced minimum with subsequent increase of T_c to above 2.5 K. For the thinnest Nb layer ($d_{CuNi} \approx 6.2$ nm, S21), the superconducting T_c sharply drops upon increasing the ferromagnetic Cu$_{41}$Ni$_{59}$ layer thickness till a certain thickness $d_{CuNi} \approx 2.5$ nm. Then, in the range $d_{CuNi} \approx 2.5 - 24$ nm, the superconducting transition temperature vanishes (T_c is at least lower than the lowest temperature reached in our cryogenic setup, 40 mK). With a further increase of the Cu$_{41}$Ni$_{59}$ layer thickness, superconductivity restores again at $d_{CuNi} \approx 25.5$ nm, reaching a level of about 0.8K at $d_{CuNi} \approx 30$ nm, and then drops down again below 40 mK at $d_{CuNi} \approx 37.5$ nm. This phenomenon of a double suppression of superconductivity is the first experimental evidence for a multiple reentrant behavior of superconducting state in S/F layered systems.

The data simulation procedure follows the fitting strategy described in detail in our previous papers [8, 9]. The solid curves in Figure 2 show results of the fitting for the ”clean” case with parameters given in the figure caption. Calculations with the physical parameters of the S21 sample series, but for thicker Nb layer $d_{Nb} \approx 6.4$ nm, show that the next island of superconductivity is possible to observe in the range $d_{CuNi} \approx 53 - 70$ nm. We will search for the second reentrance of superconductivity in our further studies.

To conclude, we report on the first experimental observation of the double supression of superconductivity in S/F bilayers with constant Nb layer thickness ($\approx$ 6.2 nm) and variable Cu$_{1-x}$Ni$_x$ alloy ($x \approx 0.59$) layer thickness. The experimental realization of the reentrant

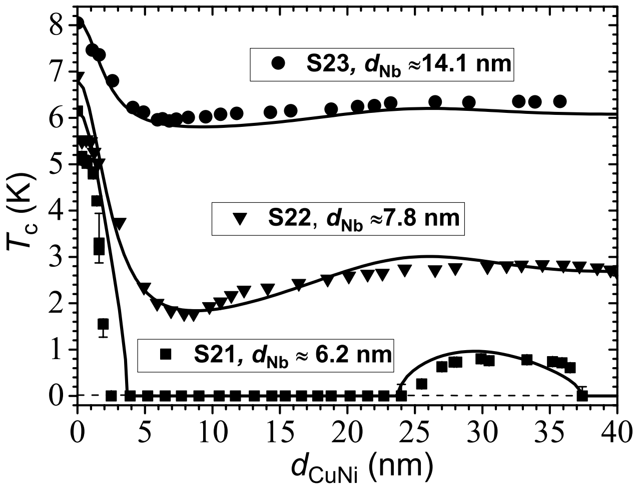

Figure 2. Non monotonous $T_c(d_F)$ dependence for the Nb/Cu$_{1-x}$Ni$_x$ bilayers ($x = 0.59$). Solid curves are calculated with values of parameters as follows: (S23) the superconducting coherence length $\xi_S = 10.0$ nm, the ratio of the Sharvin conductances $N_F v_F / N_S v_S = 0.22$, the S/F-interface transparency parameter $T_F = 0.43$, the ferromagnet coherence length $\xi_{F0} = 10.6$ nm, the ferromagnet mean free path to the coherence length ratio $l_F/\xi_{F0} = 1.1$; (S22) $\xi_S = 9.8$ nm, $N_F v_F / N_S v_S = 0.22$, $T_F = 0.55$, $\xi_{F0} = 10.6$ nm, $l_F/\xi_{F0} = 1.1$; (S21) $\xi_S = 9.6$ nm, $N_F v_F / N_S v_S$ $= 0.22$, $T_F = 0.59$, $\xi_{F0} = 11.0$ nm, $l_F/\xi_{F0} = 1.1$. The BCS coherence length for Nb was always taken $\xi_{BCS} = 42$ nm. The calculations give no further reentrance of superconductivity for the S21 series for $d_{CuNi} > 40$ nm.

superconductivity phenomenon is an essential progress towards the fabrication of a F$_1$/S/F$_2$ superconducting spin switch [10] for superconducting spintronics.

Acknowledgments

The authors are grateful to J Aarts, A I Buzdin, Z Radovic, M Yu Kupriyanov, A Golubov, C Attanasio and V Oboznov for stimulating discussions and cooperation. The work was partially supported by BMBF (project MDA01/007), RFBR (projects No 07-02-00963-a, 08-02-90105_Mol-a) and collaborative grant RFBR-Moldova “Investigation of nanostructures superconductor/ferromagnet – base element of superconducting spintronics”.

References

[1] Buzdin A I 2005 *Rev. Mod. Phys.* **77** 935
[2] Fulde P and Ferrell R 1964 *Phys. Rev.* **135** A550
[3] Larkin A I and Ovchinnikov Yu N 1964 *Zh. Eksp. Teor. Fiz.* **47** 1136 [1965 *Sov. Phys. JETP* **20** 762]
[4] Born M, Volf E 1968 *Principles of optics* (Pergamon Press 4th Edition) chapter 7
[5] Khusainov M G and Proshin Yu N 1997 *Phys. Rev.* B **56** 14283; Erratum: 2000 *Phys. Rev.* B **62** 6832
[6] Tagirov L R 1998 *Physica* C **307** 145
[7] Vodopyanov B P and Tagirov L R 2003 *Pis'ma v ZhETF* **78** 1043 [2003 *JETP Letters* **78** 555]
[8] Zdravkov V I, Sidorenko A S, Obermeier G, Gsell S, Schreck M, Müller C, Horn S, Tidecks R, Tagirov L R 2006 *Phys. Rev. Lett.* **97** 057004
[9] Sidorenko A S, Zdravkov V I, Prepelitsa A, Helbig C, Luo Y, Gsell S, Schreck M, Klimm S, Horn S, Tagirov L R, Tidecks R 2003 *Ann. Phys.* (Berlin) **12** 37
[10] Tagirov L R 1999 *Phys. Rev. Lett.* **83** 2058