



TESCO

nternational Atomic Energy Agency

SMR.1664 - 4

Conference on Single Molecule Magnets and Hybrid Magnetic Nanostructures

27 June - 1 July 2005

Josephson π -junctions on basis of superconductor-ferromagnet-superconductor sandwiches

> Valery RYAZANOV **Russian Academy of Sciences Institute of Solid State Physics Chernogolovka District** 142432 Moscow **RUSSIAN FEDERATION**

These are preliminary lecture notes, intended only for distribution to participants Strada Costiera 11, 34014 Trieste, Italy - Tel. +39 040 2240 111; Fax +39 040 224 163 - sci info@ictp.it, www.ictp.it

Josephson π -junctions on basis of superconductor-ferromagnet-superconductor sandwiches

V. V. Ryazanov,

V. A. Oboznov, A. S. Prokofiev, V. V. Bolginov, A. K. Feofanov

Institute of Solid State Physics, Chernogolovka, Russia, 142432

1. INTRODUCTION

It is well-known that the current-phase relation of Josephson junction is a 2π -periodical function. In case of tunnel or dirty metal barriers superconducting current through the junction is described by relation $I_s = I_c \sin \varphi$ and its energy is given by $E = E_J(1 - \cos \varphi)$, where φ is phase difference of macroscopic superconducting wave function (Ginzburg-Landau order parameter) on the junction banks. In 1977 Bulaevskii et al ¹ described supercurrent through the barrier with uncorrelated magnetic impurities and proved the possibility of sign change of the supercurrent and junction coupling energy:

$$I_s = -I_c \sin \varphi; \quad E = E_J (1 + \cos \varphi) \tag{1}$$

Such modification of the current-phase relation and the energy expression corresponds to π -shift of phase difference across the junction (see Fig. 1). Authors of Ref. 1 have named such junction as ' π -contact'. They have shown that the state with a spontaneous flux equal to half flux quantum, $\Phi_0/2$, has to exist in a superconducting loop containing the π -contact. Later a number of other mechanisms of the π -shift of phase difference were proposed.

Superconductor-ferromagnet-superconductor $(SFS) \pi$ -junction was predicted as early as 1982 by Buzdin et al ² but was detected only in 2000 ³. Inversion of the superconducting order parameter sign over the SFS junction occurs due to exchange-field-induced oscillations of the order parameter in the ferromagnetic layer, i.e. induced superconductivity in *F*-layer is spatially inhomogeneous and the superconducting order parameter contains nodes

where the phase changes by π . This state is a sort of Larkin-Ovchinnikov-Fulde-Ferrel (LOFF) state ^{4,5} that has to describe a hypothetical "magnetic superconductor" or superconductor in huge magnetic field.

This talk is devoted to detailed discussions of experiments on structures with SFS $\pi\text{-junctions}.$

2. PROXIMITY EFFECT IN THE SF SYSTEM

In recent years, considerable interest has been shown in artificial metallic multilayer systems with alternating magnetic and nonmagnetic layers. One of the aspect of this subject is the coexistence of superconductivity and ferromagnetism in SF-multilayered structures. The antagonism of these two phenomena differing in spin ordering is a cause of the strong suppression of superconductivity in the contact area of the S- and F- materials. However, the superconducting order parameter does not simply decay into the ferromagnet but also oscillates.



Figure 1. The current-phase relations and the energy vs. junction phase dependences of Josephson junctions in conventional (0-) and π -states.

As distinct from superconductor-normal metal system, where the coherence length ξ_N responsible for the decay of the order parameter in the N-layer is real, the coherence length ξ_F in a ferromagnet is complex, $\frac{1}{\xi_F} = \frac{1}{\xi_{F1}} + i\frac{1}{\xi_{F2}}$. This means that in the ferromagnet close to the SF-interface oscillations of the induced superconducting order parameter related to the imaginary part ξ_{F2} of the coherence length should arise together with the decay determined by the length ξ_{F1} . A physical origin of the order parameter oscillations in a ferromagnet is exchange splitting of the spin-up and spin-down electron subbands. Exchange splitting close to FS-interface results in a large increase of momentum difference Q between the incoming and reflected excitations, i.e. results in appearance of the Andreev bound state nonzero momentum. In the diffusive limit the order parameter oscillations are superimposed on the decay arising due to pair breaking by impurities in the presence of the exchange field. The order parameter (superconducting wave function) in the ferromagnetic half-space can be expressed in terms of the ξ_{F1} and ξ_{F2} as follows:

$$\Psi_F(x) = \Psi_{F0} \exp\left(-\frac{x}{\xi_{F1}}\right) \cos\left(\frac{x}{\xi_{F2}}\right).$$
(2)

Here Ψ_{F0} is the order parameter in the ferromagnet near the *SF*-interface and *x* is the coordinate in the direction perpendicular to the interface. The wavelength of the order parameter oscillations is

$$\lambda_{ex} = 2\pi \xi_{F2}.\tag{3}$$

For $E_{ex} \gg k_B T$, the values of ξ_{F1} and ξ_{F2} are equal and the complex coherence length can be written as

$$\xi_F = \sqrt{\frac{\hbar D}{2iE_{ex}}}.\tag{4}$$

In case of weak exchange energy $E_{ex} \geq k_B T$, that is valid for Nb-Cu/Ni multilayers discussed below, the thermal and exchange energies make comparable contribution to the pair decay process. The general expression for the complex coherence length is following ³:

$$\xi_F = \sqrt{\frac{\hbar D}{2(\pi k_B T + iE_{ex})}}.$$
(5)

Extracting from this expression the real and imaginary parts, (for this purpose it is convenient to write

$$\pi k_B T + i E_{ex} = \sqrt{(\pi k_B T)^2 + E_{ex}^2} exp(i \arctan(E_{ex}/\pi k_B T))),$$
 we obtain

$$\xi_{F1,2} = \sqrt{\frac{\hbar D}{((\pi k_B T)^2 + E_{ex}^2)^{1/2} \pm \pi k_B T}}.$$
(6)

Note that in case of $E_{ex} \geq k_B T$ the decay length ξ_{F1} increases with the temperature decrease, whereas the oscillation wavelength $2\pi\xi_{F2}$ decreases. This allows us to observe the transition of the SFS junction to the π -state by means of temperature change.

3. JOSEPHSON SFS JUNCTIONS AND π -STATE

It is easy to see from (2) that in case of SFS sandwich we should have a transition to the π -state at a F-layer thickness d_F close to $\pi\xi_{F2}$ and back transition to conventional '0-state' at d_F close to $2\pi\xi_{F2}$. The $0 - \pi$ transition by means of the F-layer thickness and temperature changes was observed for the first time in Ref. 3. More detailed reentrant dependence of the SFS junction critical currents vs. the F-layer thickness was measured in Ref. 6,7, however only the thickness range close to the 0- π -transition thickness was investigated in these works. Authors of Ref. 8 have obtained the following expression for the critical current of a dirty SFS structure in the limit $E_{ex} \gg k_BT$, used the formalism of quasiclassical Usadel equations:

$$I_c = I_{c0} y \frac{|\sinh y \cos y + \cosh y \sin y|}{\sinh^2 y \cos^2 y + \cosh^2 y \sin^2 y}, \ y = d_F / \xi_F^*, \tag{7}$$

where $\xi_F^* = \xi_{F1} = \xi_{F2}$. We will show below, that for the real SFS structures we have to use different ξ_{F1} and ξ_{F2} in (7), i.e. $y_1 = d_F/\xi_{F1}$ has to be used as an argument of sinh and cosh, while $y_2 = d_F/\xi_{F2}$ has to be used as an argument of sin and cos.

4. NONMONOTONIC T_c BEHAVIOR OF SF-BILAYERS

Anomalous, nonmonotonic dependence of the critical temperature, T_{c1} , of SF-multilayers on the F-layer thickness was first analyzed theoretically by Radovic et al ⁹ in 1991. It was shown later ^{15,11,12,13} that the nonmonotonic $T_{c1}(d_F)$ and $T_{c1}(E_{ex})$ characteristics inhere also in SF-bilayers and FSF-trilayers and are related to the nonmonotonic change of the transmission coefficient of the FS-interface. The $T_{c1}(d_F)$ minimum observed in experiments (see, for example Ref. 12,14) corresponds to largest FS-interface transparency and to strongest suppression of the superconducting layer by ferromagnetic one. A detailed theoretical analysis carried out in Ref. 16,17 has shown that the T_{c1} minimum has to occur at the thickness d_F close to a quarter of the period λ_{ex} of the order parameter spatial oscillations, i.e. at $(\pi/2)\xi_{F2}$. The simple sketchy consideration of the order parameter modulus behavior elucidated this inference is presented in Fig. 2. Since the



Figure 2. Sketchy picture of the superconducting order parameter distributions in SF-bilayers with different F-layer thicknesses. The middle panel corresponds to largest FS-interface transmission coefficient and minimal T_{c1} .

boundary condition at the free *F*-surface determines zero order parameter derivative $\Psi'_F(d_F) = 0$ the order parameter has node at *FS*-interface just at the *F*-layer thickness equal to a quarter of the spatial oscillations period and integral minimum of the bilayer order parameter is achieved.

To investigate the sign-reversal order parameter in ferromagnet close to SF-interface by means of studies on both SF-bilayers and Josephson SFS junctions, thin-film Nb - Cu/Ni structures and Nb - Cu/Ni - Nbsandwiches were fabricated. To compare results obtained on these different structures, the same ferromagnetic alloy $(Cu_{0.43}Ni_{0.57})$ with the Curie temperature $T_{Curie} \approx 150 \, K$ was used for F-layer deposition. The relatively weak ferromagnetism of the Cu/Ni alloy was important for preparing homogenous and continuous F-layers with the thickness comparable with the ξ_{F2} of the order of dozen nanometers. Conventional ferromagnetic metals (Co, Fe, Ni) can hardly be used for fabrication of the sufficiently uniform F-layers because the pair coherence length in this case is close to 1 nm. Moreover the application of the alloys with low T_{Curie} and E_{ex} allows us to observe the transition of the Josephson SFS junction to π -state and back by means of temperature change, since in case of $E_{ex} \geq k_B T$ temperature includes appreciable contribution to the coherence length (5),(6) and the period of spatial oscillations (3).

V. V. Ryazanov et al.



Figure 3. Critical temperature of a $Nb - Cu_{0.43}Ni_{0.57}$ bilayers vs. thickness of ferromagnetic layer. The inset shows the sample experimental geometry used in resistive experiments.

The inset of Fig. 3 shows the experimental geometry of the SF-bilayers used in our in-plane resistive experiments ¹⁴. The lower superconducting niobium layer with a thickness of 11 nm (close to the coherence length in the thin-film niobium $\xi_s \simeq 7 - 8 nm$ was deposited by dc-magnetron sputtering. The next copper-nickel alloy film was prepared in the same vacuum run by rf-sputtering. The weak ferromagnetism of the Cu/Ni alloy had allowed us to retain the superconductivity of the SF-bilayer with T_{c1} in range of 3 - 7 K when we changed the F-layer thickness between 20 and 1 nm as it is shown in Fig. 3. One can readily see that T_{c1} goes through the dip before reaching a saturation. A comparison of the experimental curve with the results of the detailed theoretical analysis has been carried out in Ref. 16,17 and has demonstrated reasonably good agreement. Plainly distinguishable dip centered at $d_F \simeq 5 nm$ has to correspond to a quarter of the period λ_{ex} .

5. THICKNESS DEPENDENCE OF SFS JUNCTION CRITICAL CURRENT

The experimental studies of the Josephson characteristics of the SFSjunctions ^{?,3} were carried out by us on thin-film sandwiches $Nb-Cu_{1-x}Ni_x-$ Nb, with x in the range 0.52 - 0.57 and the Curie temperature, T_{Curie} , of the copper-nickel layer in the range $30 - 150 K^3$. The weak ferromagnetism of the Cu/Ni alloy made possible flowing of supercurrents through the F-layers up to 30 nm in thickness, prepared with the roughness of 1 - 2 nm. Inset in the upper panel of Fig. 5 shows the schematic cross-section of a thin-film $Nb - Cu_{1-x}Ni_x - Nb$ sandwich. The bottom Nb electrode 100 μm wide and 110 nm thick was fabricated by dc-magnetron sputtering and subsequent photolithography and chemical etching. The deposition of the copper-nickel film was carried out by rf-sputtering after ion etching of the niobium surface (targets of various contents with x varying from 0.52 to 0.57 were used). Then the insulating layer with $50 \times 50 \ \mu m^2$ 'window' determining the junction area was prepared by lift-off photolithography. We used 170 nm thick SiO film as insulator, which was deposited by vacuum evaporation. The fabrication procedure was accomplished by dc-magnetron sputtering of the upper niobium electrode, $80 \,\mu m$ wide and $240 \,nm$ thick, after preliminary ion cleaning the surface of the copper-nickel layer. The upper electrode patterning was made by lift-off photolithography. The junction normal resistance R_n did not exceed $10^{-5} \Omega$, so the transport characteristics of the junctions were measured by the picovoltmeter based on the rf-SQUID with sensitivity better than $10^{-11} V$. The $I_c(H)$ patterns for 'fresh' or well demagnetized samples were described by the Fraunhopher relation with high accuracy, which indicates the fact of high uniformity of the thickness and the magnetic properties of the F-layer along the junction. The latter seems to be caused by averaging of the small-scale structure of magnetic domains in the F-layer resulting in highly uniform current flow through the ferromagnetic layer with zero net magnetization that was indicated by zero field position of the main peak of the Fraunhopher pattern 3 .

We have investigated the thickness dependence of the SFS junction critical current density in a wide thickness range. In the thickness interval 6-26 nm we have about 6 orders of the critical current density change with two vanishing: first $(0 - \pi)$ -transition takes place at thickness $d_{F,\pi 1} \simeq \pi \xi_{F2}$ about 11 nm and second (back) transition from π - to 0-state at thickness $d_{F,\pi 2} \simeq 2\pi \xi_{F2} = 22 - 23 nm$. Estimations of the ξ_{F2} value and a rate of the critical current decay proportional to ξ_{F1} for the scale used in the Figure show that $d_F \gg \xi_{F1}$. In this limit (7) can be modified to form reminiscent

$$j_c = j_{c0} \exp\left(-\frac{d_F}{\xi_{F1}}\right) \left|\cos\left(\frac{d_F}{\xi_{F2}}\right) + \sin\left(\frac{d_F}{\xi_{F2}}\right)\right|, d_F \gg \xi_{F1}$$
(8)

One can readily see that the second node of the critical current density at back $\pi - 0$ -transition occurs at the thickness $d_{F,\pi 2} = \frac{7}{4}\pi\xi_{F2} = \frac{7}{8}\lambda_{ex}$. A fitting to this formula is shown in Fig. 4 by solid lines. ξ_{F1} and ξ_{F2} differ near by a factor of three, that cannot be explained by the temperature contribution (see (6)), since one can see in Fig. 4 that double temperature increase only slightly changes ξ_{F1} and ξ_{F2} values. Additional contributions like spinorbit ^{18,19} and spin-flip scattering ^{7,20} have to be taken into account in (6), that increase the ξ_{F2} and decrease ξ_{F1} . Since spin-orbit scattering inheres in ferromagnets with large atomic number Z we guess that in our case of Cu/Ni alloy we deal with spin-flip scattering due to magnetic inhomogeneity that could be multidomain structure, domain walls, and above all Ni-rich clusters arising in $Cu_{1-x}Ni_x$ ferromagnet for x close to 0.5.

6. REENTRANT TEMPERATURE DEPENDENCES OF CRITICAL CURRENT IN SFS JUNCTIONS



Figure 4. Thickness dependences of the critical current density for $Nb - Cu_{0.47}Ni_{0.53} - Nb$ junctions at temperature 4.2 K.

(2):

TEMPERATURE $\pi - 0$ -TRANSITION

As it was discussed above the weak ferromagnetism of the Cu/Ni alloy made possible to induce transition to π -state not only as a function of the thickness, d_F , and magnetic moment, m_{at} , of *F*-layer but also by means of the *SFS* junction temperature change. We have investigated the second (π – 0) transition in detail. Fig. 5 shows experimentally measured dependences $j_c(T)$ at various Ni content (x = 0.52, 0.53 and 0.57) in the ferromagnetic



Figure 5. Anomalous temperature dependences of the SFS junction critical current density at various Ni content (x = 0.52, 0.53 and 0.57) in $Cu_{1-x}Ni_x$ interlayers of $Nb - Cu_{1-x}Ni_x - Nb$ sandwiches with thickness d_F close to value corresponding to the back transition from the π - to 0-state. The inset in the upper panel shows a schematic picture of the cross-section of the SFS junction. The inset in the middle panel is an example of Curie temperature detection by means of the anomalous Hall effect measurements of saturation magnetization ⁶.

 $Cu_{1-x}Ni_x$ interlayers of the $Nb - Cu_{1-x}Ni_x - Nb$ sandwiches with thickness d_F close to value corresponding to the second transition. The temperature decrease from 9 K down to 1 K is accompanied by the increase of 1-2 nm in the spatial oscillation period, λ_{ex} , that results in temperature transition from π - to 0-state (see (6) and discussion related to it). The reentrant temperature dependences are direct consequence of the transition at $T = T_{\pi 2}$ where the critical current vanishes ³. In the transition point the critical current $I_c(T)$ is formally equal to zero and then should change its sign. Since in real experiments we could measure only positive values of the critical current, the dependence $I_c(T)$ has a sharp cusp at $T = T_{\pi 2}$ that is the negative branch of the curve reflected to the positive region. In accordance with (4) ξ_{F2} is inverse proportional to $\sqrt{E_{ex}}$, therefore the thickness $d_{F,\pi 2}$ = $\frac{7}{4}\pi\xi_{F2}\propto\sqrt{E_{ex}}$. Inset in the middle panel of Fig.5 shows a procedure of Curie temperature finding from anomalous Hall effect measurements of the saturation magnetization ⁶ for $Cu_{0.47}Ni_{0.53}$ -film with thickness 22 nm. The overextended tail at temperature higher than T_{Curie} we refer to the Ni-rich clusters. A qualitative accordance with (4) is on hand: the larger magnetic content of the F-layer the smaller the period of spatial oscillations λ_{ex} and $d_{F,\pi 2}$. Moreover there is quite good quantitative agreement between values of λ_{ex} extracted from the SF-bilayer and SFS junction measurements for the $Cu_{0.43}Ni_{0.57}$ -ferromagnetic layers fabricated by the same rf-sputtering method. Comparison of data of Fig.3 and Fig.5, lower panel, shows that λ_{ex} obtained from formula for the bilayer T_c minimum $(d_{F,min} \simeq \lambda_{ex}/4)$ and one for the second transition give very close results. Thus, $\lambda_{ex} \simeq 20 nm$ and $\xi_{F2} \simeq 3.5 - 4 \, nm$ for the $Cu_{0.43} Ni_{0.57}$ -ferromagnetic film.

REFERENCES

- L. N. Bulaevskii, V. V. Kuzii, and A. A. Sobyanin, JETP Lett. 25, 290 (1977) [Pis'ma Zh. Eksp. Teor. Fiz. 25, 314 (1977)].
- A. I. Buzdin, L. N. Bulaevskij, S. V. Panyukov, *Pis'ma Zh. Eksp. Teor.Fiz.* 35 147 (1982) [*JETP Lett.* 35 178 (1982)]
- V. V. Ryazanov, V. A. Oboznov, A. Yu. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, *Phys. Rev. Lett.* 86, 2427 (2001).
- A. I. Larkin, and Yu. N. Ovchinnikov, Sov. Phys. JETP. 20, 762 (1965) [Zh. Eksp. Teor. Fiz. 47, 1136 (1964)].
- 5. P. Fulde, and R. A. Ferrel, Phys. Rev. 135, A550 (1964).
- T. Kontos, M. Aprili, J. Lesueur, F. Genet, B. Stephanidis, and R. Boursier, *Phys. Rev. Lett.* 89, 137007 (2002).
- H. Sellier, C. Baraduc, F. Lefloch, and R. Calemczuk, *Phys. Rev. B* 68, 054531 (2003).

- A. I. Buzdin, B. Vujichich, M. Yu. Kupriyanov, *Zh. Eksp. Teor. Fiz.* **101** 231 (1992) [Sov. Phys. JETP **74** 124 (1992)]
- Z. Radovic, L. Ledvij, L. Dobrosavljevic-Grujic, A. I. Buzdin, and J. R. Clem, *Phys. Rev. B.* 44, 759 (1991).
- J. S. Jiang, D. Davidovic, D. H. Reich and C. L. Chien, *Phys. Rev. Lett.* 74, 314 (1995).
- J. Aarts, J. M. E. Geers, E. Bruck, A. A. Golubov, and R. Coehoorn, *Phys. Rev. B* 56, 2779 (1997).
- L. Lazar, K. Westerholt, H. Zabel, L. R. Tagirov, Yu. V. Goryunov, G. G. Khaliullin, and I. A. Garifullin, *Phys. Rev. B* 61, 3711 (2000).
- 13. L. R. Tagirov, *Physica C* **307**, 145 (1998).
- V. V. Ryazanov, V. A. Oboznov, A. S. Prokof'ev, and S. V. Dubonos, *Pis'ma Zh. Eksp. Teor. Fiz.* **77**, 43 (2001) [*JETP Lett.* **77**, 39 (2003)].
- J. S. Jiang, D. Davidovic, D. H. Reich and C. L. Chien, *Phys. Rev. Lett.* 74, 314 (1995).
- Ya. V. Fominov, N. M. Chtchelkatchev, and A. A. Golubov, *Phys. Rev. B* 66, 014507 (2002).
- Ya. V. Fominov, N. M. Chtchelkatchev, and A. A. Golubov, *Pis'ma Zh. Eksp.* Teor. Fiz. 74, 101 (2001) [JETP Lett. 74, 96 (2001)].
- 18. E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B 55, 15174 (1997)
- 19. V. N. Krivoruchko and R. V. Petryuk, Phys. Rev. B 66, 134520 (2002).
- 20. A. I. Buzdin, Unpublished.