



Superconductivity and magnetism/Supraconductivité et magnétisme

Proximity effect in superconductor–ferromagnet heterostructures

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Abstract

We discuss the particularities of the proximity effect in superconductor–ferromagnet systems: the damped oscillatory behavior of the Cooper pair wave function, the oscillations of the critical temperature in S/F bilayers and multilayers and the conditions for the π -Josephson junctions formation. Also we outline the possibility of the formation of the novel type of the Josephson junction, intermediate between the 0 and π junctions. **To cite this article:** *A.I. Buzdin, V.V. Ryazanov, C. R. Physique 7 (2006).*

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Résumé

L'effet de proximité dans les hétérostructures supraconductrices–ferromagnétiques. Nous discutons les particularités de l'effet de proximité dans les systèmes supraconducteurs–ferromagnétiques : le comportement oscillatoire amorti de la fonction d'onde des paires de Cooper, les oscillations de la température critique dans les bicouches et multicouches S/F, et les conditions de formation de jonctions Josephson π . En outre, nous soulignons la possibilité de formation de jonctions Josephson intermédiaires entre les jonctions 0 et les jonctions π . **Pour citer cet article :** *A.I. Buzdin, V.V. Ryazanov, C. R. Physique 7 (2006).*

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1. Introduction

The coexistence of singlet superconductivity with ferromagnetism is very unlikely in bulk compounds, but it may be easily achieved in artificially fabricated layered ferromagnet/superconductors (F/S) systems. Due to the proximity effect, the Cooper pairs can penetrate into the F layer and induce superconductivity there. In such case we have the unique possibility to study the properties of superconducting electrons under the influence of a huge exchange field acting on the electron spins. In addition, it is possible to study the interplay between superconductivity and magnetism in a controlled manner, since by varying the layer thicknesses and magnetic content of F layers we change the relative strength of two competing orderings. The behavior of the superconducting condensate under these conditions is quite peculiar. Here we present the outlook of the particularities of the F/S systems, for more details see the recent reviews [1,2].

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Some years ago, Larkin and Ovchinnikov [3], and Fulde and Ferrell [4] demonstrated that in a pure ferromagnetic superconductor, at low temperature, the superconductivity may be non-uniform (this is the so-called FFLO or LOFF state). Due to the incompatibility of ferromagnetism and superconductivity it is not easy to verify this prediction by experiment. Moreover, the electron scattering on the impurities destroys the FFLO state very quickly and its observation is possible only in the clean limit [5]. It occurs that in a ferromagnet in contact with a superconductor the Cooper pair wave function has a damped oscillatory behavior [6–8] which may be considered, in some sense, as analogous to the decaying non-uniform FFLO superconducting state. This phenomenon, however, is quite general, and must be present in both clean and dirty limits. It results in many interesting effects: the spatial oscillations of the electron's density of states, the non-monotonous dependence of the critical temperature of S/F multilayers and bilayers on the ferromagnet layer thickness, the realization of the Josephson ' π '-junctions in S/F/S systems.

Note that practically all interesting effects related with the interplay between the superconductivity and the magnetism in S/F structures occur at the nanoscopic range of layer thicknesses. The observation of these effects became possible only recently due to the great progress in the preparation of high-quality hybrid F/S systems.

2. Generalized Ginzburg–Landau functional

Qualitatively, the phenomenon of the FFLO phase formation and the particularities of the proximity effect in S/F systems may be described if we generalize the standard Ginzburg–Landau expansion (see, for example, [9]):

$$F = a|\psi|^2 + \gamma|\vec{\nabla}\psi|^2 + \frac{b}{2}|\psi|^4 \quad (1)$$

where ψ is the superconducting order parameter, and the coefficient a vanishes at the transition temperature T_c . At $T < T_c$, the coefficient a is negative and the minimum of F in Eq. (1) is achieved for a uniform superconducting state with $|\psi|^2 = -\frac{a}{b}$. If we consider also the paramagnetic effect of the magnetic field, all the coefficients in Eq. (1) will depend on the energy of the Zeeman splitting $\mu_B H$, or an exchange field h in the ferromagnet. Usually, the orbital effect is much more important for the superconductivity destruction than the paramagnetic one. It explains why in the standard Ginzburg–Landau theory there is no need to take into account the field and temperature dependence of the coefficients γ and b . However, when the paramagnetic effect becomes predominant, this approximation fails. The qualitatively new physics emerges due to the fact that the coefficient γ changes its sign for relatively large h/T ratio. The negative sign of γ means that the minimum of the functional no longer corresponds to an uniform state. To describe such a situation it is necessary to add a higher order derivative term in the expansion (1), and the generalized Ginzburg–Landau expansion will be:

$$F_G = a(H, T)|\psi|^2 + \gamma(H, T)|\vec{\nabla}\psi|^2 + \frac{\eta(H, T)}{2}|\vec{\nabla}^2\psi|^2 + \frac{b(H, T)}{2}|\psi|^4 \quad (2)$$

The critical temperature of the second order phase transition into a superconducting state may be found from the solution of the linear equation for the superconducting order parameter

$$a\psi - \gamma\Delta\psi + \frac{\eta}{2}\Delta^2\psi = 0 \quad (3)$$

If we seek a non-uniform solution $\psi = \psi_0 \exp(i\mathbf{q}\mathbf{r})$, the corresponding critical temperature depends on the wave-vector \mathbf{q} and is given by the expression $a = -\gamma q^2 - \frac{\eta}{2}q^4$. Note that the coefficient a may be written as $a = \alpha(T - T_{cu}(H))$, where $T_{cu}(H)$ is the critical temperature of the transition into the uniform superconducting state. In a standard situation, the gradient term in the Ginzburg–Landau functional is positive, $\gamma > 0$, and the highest transition temperature coincides with $T_{cu}(H)$; it occurs for the uniform state with $q = 0$. However, in the case $\gamma < 0$, the maximum critical temperature corresponds to the finite value of the modulation vector $q_0^2 = -\gamma/\eta$ and the corresponding transition temperature into the non-uniform FFLO state $T_{ci}(H)$ is given by $a = \alpha(T_{ci} - T_{cu}) = \gamma^2/2\eta$. It is higher than the critical temperature T_{cu} of the uniform state. Therefore, we see that the FFLO state appearance may simply be interpreted as a change of sign of the gradient term in the Ginzburg–Landau functional.

3. Damped oscillatory dependence of the Cooper pair wave function in ferromagnets

To get idea about the peculiarity of the proximity effect in S/F structures, let us start from the description based on the generalized Ginzburg–Landau functional Eq. (2). Such an approach is adequate for a small wave-vector modulation

case, otherwise the microscopical theory must be used. We address the question of the proximity effect for a weak ferromagnet described by the generalized Ginzburg–Landau functional Eq. (2). More precisely, we consider the decay of the order parameter in the normal phase ($x > 0$), i.e., at $T > T_{ci}$, assuming that our system is in contact with another superconductor ($x < 0$) with a higher critical temperature, and the x axis is chosen perpendicular to the interface.

The induced superconductivity is weak and to deal with it, we may use the linearized equation for the order parameter (3), which is written for our geometry as

$$a\psi - \gamma \frac{\partial^2 \psi}{\partial x^2} + \frac{\eta}{2} \frac{\partial^4 \psi}{\partial x^4} = 0 \quad (4)$$

The solutions of this equation in the normal phase are of the type $\psi = \psi_0 \exp(kx)$, with a *complex wave-vector* $k = k_1 + ik_2$, and

$$k_1^2 = \frac{|\gamma|}{2\eta} \left(\sqrt{1 + \frac{T - T_{ci}}{T_{ci} - T_{cu}}} - 1 \right) \quad (5)$$

$$k_2^2 = \frac{|\gamma|}{2\eta} \left(1 + \sqrt{1 + \frac{T - T_{ci}}{T_{ci} - T_{cu}}} \right) \quad (6)$$

If we choose the gauge with the real order parameter in the superconductor, then the solution for the decaying order parameter in the ferromagnet is also real

$$\psi(x) = \psi_1 \exp(-k_1 x) \cos(k_2 x) \quad (7)$$

where the choice of the root for k is the condition $k_1 > 0$. So the decay of the order parameter is accompanied by its oscillation, which is the characteristic feature of the proximity effect in the considered system. When we approach the critical temperature T_{ci} the decaying wave-vector vanishes, $k_1 \rightarrow 0$, while the oscillating wave-vector k_2 goes to the FFLO wave-vector, $k_2 \rightarrow \sqrt{|\gamma|/\eta}$, so a FFLO phase emerges. Let us compare this behavior with the standard proximity effect described by the linearized Ginzburg–Landau equation for the order parameter

$$a\psi - \gamma \frac{\partial^2 \psi}{\partial x^2} = 0 \quad (8)$$

with $\gamma > 0$. In such case T_c simply coincides with T_{cu} , and the decaying solution is $\psi = \psi_0 \exp(-x/\xi(T))$, where the coherence length $\xi(T) = \sqrt{\gamma/a}$. The different behavior of the superconducting order parameter in S/F and S/N systems is illustrated in Fig. 1. The presented simple analysis shows the appearance of the oscillations of the order parameter in the presence of an exchange field. This is a fundamental difference between the proximity effect in S/F and S/N systems, and it is at the origin of many peculiar characteristics of S/F heterostructures.

In real ferromagnets, the exchange field is very large compared with the superconducting temperature and energy scales, so the gradients of the superconducting order parameter variations are also large, and cannot be treated in the framework of the generalized Ginzburg–Landau functional. To describe the relevant experimental situation we need to use a microscopical approach.

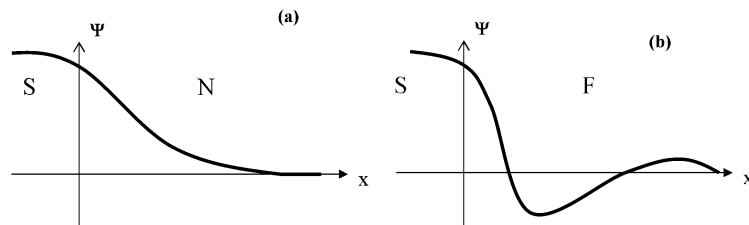


Fig. 1. Schematic behavior of the superconducting order parameter near the (a) superconductor-normal metal and (b) superconductor-ferromagnet interfaces.

If the electron scattering mean free path l is small (which is usually the case in S/F systems), the most natural approach is to use the Usadel equations [10] for the Green's functions averaged over the Fermi surface. The linearized Usadel equation for the anomalous Green function F_f in the ferromagnet is expressed

$$\left(|\omega| + i\hbar \operatorname{sgn}(\omega) + \frac{1}{\tau_s} \right) F_f - \frac{D_f}{2} \frac{\partial^2 F_f}{\partial x^2} = 0 \quad (9)$$

where $\omega = (2n + 1)\pi T$ are the Matsubara frequencies, and $D_f = \frac{1}{3}v_F l$ is the diffusion coefficient in the ferromagnet. The parameter τ_s^{-1} describes the magnetic scattering in the ferromagnetic alloys used as F layers. Note that this form of the Usadel equation in the ferromagnet implies strong magnetic uniaxial anisotropy when the magnetic scattering in the plane (xy) perpendicular to the anisotropy axis is negligible.

In the F region, we may neglect the Matsubara frequencies compared to the large exchange field ($h \gg T_c$). Also assuming first that the magnetic scattering is weak, we readily obtain the decaying solution for F_f

$$F_f(x, \omega > 0) = A \exp\left(-\frac{i+1}{\xi_f} x\right) \quad (10)$$

where $\xi_f = \sqrt{D_f/\hbar}$ is the characteristic length of the superconducting correlations decay (with oscillations) in F-layer. Due to the condition $h \gg T_c$, this length is much smaller than the superconducting coherence length $\xi_s = \sqrt{D_s/(2\pi T_c)}$, i.e., $\xi_f \ll \xi_s$. In a ferromagnet, the role of the Cooper pair wave function is played by Ψ that decays as

$$\Psi \sim \sum_{\omega} F(x, \omega) \sim \Delta \exp\left(-\frac{x}{\xi_f}\right) \cos\left(\frac{x}{\xi_f}\right) \quad (11)$$

We retrieve the damping oscillatory behavior of the order parameter Eq. (7). The important conclusion we obtain from the microscopic approach is that in the dirty limit in the absence of magnetic scattering, the scale for the oscillation and decay of the Cooper pair wave function in a ferromagnet is the same. If we take into account the magnetic scattering, then the decaying length ξ_{f1} becomes smaller than the oscillating length ξ_{f2} ; namely

$$\xi_{f1} = \xi_f / \sqrt{1 + \alpha_s^2 + \alpha_s} \quad \text{and} \quad \xi_{f2} = \xi_f / \sqrt{1 + \alpha_s^2 - \alpha_s}$$

where the parameter $\alpha_s = 1/(\tau_s h)$ characterizes the relative strength of the magnetic scattering.

The damped oscillatory behavior of the order parameter may lead to the electronic density of state (DOS) oscillations in a ferromagnet in contact with a superconductor [11]. This prediction has been confirmed by the experiment of [13], which up to now remains the only experimental observation of the DOS oscillations in F layer. Note that the magnetic scattering effect complicates this type of experiment, strongly reducing the amplitude of the oscillations.

4. Oscillatory superconducting transition temperature in S/F multilayers and bilayers

The damped oscillatory behavior of the superconducting order parameter in ferromagnets may produce the commensurability effects between the period of the order parameter oscillation (which is of the order of $2\pi\xi_{f2}$) and the thickness of a F layer. This results in the striking non-monotonous superconducting transition temperature dependence on the F layer thickness in S/F multilayers and bilayers. Indeed, for a F layer thickness smaller than ξ_{f2} , the pair wave function in the F layer changes a little and the superconducting order parameter in the adjacent S layers must be the same. The phase difference between the superconducting order parameters in the S layers is absent and we call this state the '0'-phase. On the other hand, if the F layer thickness becomes of the order of $\pi\xi_{f2}$, the pair wave function may go through zero at the center of the F layer providing the state with the opposite sign (or π shift of the phase) of the superconducting order parameter in the adjacent S layers, which we call the ' π '-phase. The increase of the thickness of the F layers may provoke the subsequent transitions from '0'- to ' π '-phases, which superpose on the commensurability effect and results in a very special dependence of the critical temperature on the F layer thickness [7,8]. The experimental observation [14] (see Fig. 2) of this unusual dependence in Nb/Gd was the first strong evidence in favour of the ' π '-phase appearance. For the S/F bilayers, the transitions between '0-' and ' π '-phases are impossible; nevertheless the commensurability effect between ξ_{f2} and F layer thickness also leads to the non-monotonous dependence of T_c on the F layer thickness. Processes of the normal quasiparticle reflection at the free F layer boundary

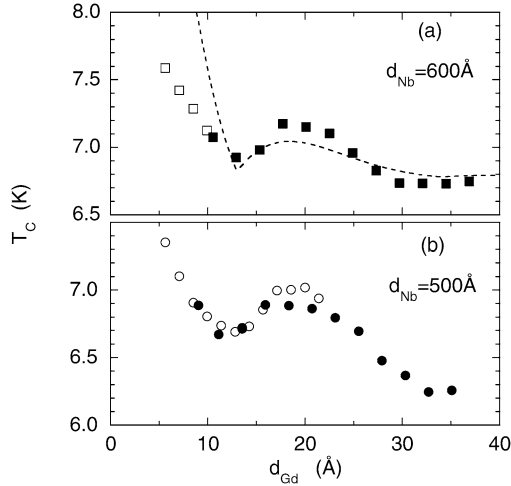


Fig. 2. Oscillatory-like dependence of the critical temperature of Nb/Gd multilayers versus thickness of Gd layer [14]. Dashed line in (a) is a fit using the theory [8].

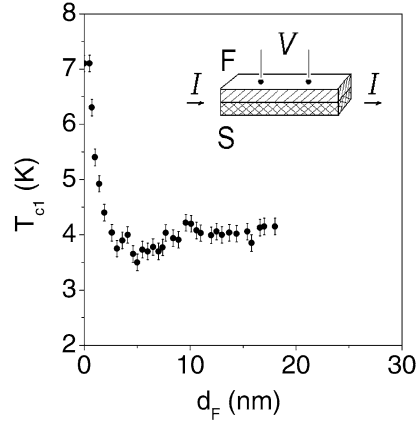


Fig. 3. Critical temperature of Nb/Cu_{0.43}Ni_{0.57} bilayer versus the thickness of the ferromagnetic layer [16].

and Andreev reflection at SF-interface interfere and this results in T_c minima reached when the F layer thickness is close to a quarter of the spatial oscillation period [15,7]. The dependence of the T_c of Nb/Cu_{0.43}Ni_{0.57} bilayer on the F layer thickness [16] is presented in Fig. 3. Recently a different oscillatory behavior of $T_c(d_f)$ in Nb/Co bilayers and multilayers has been observed experimentally in detail [17].

5. Superconductor–ferromagnet–superconductor ‘ π ’-junction

The experiments on the critical temperature of the S/F multilayers and bilayers attracted much interest to the proximity effect in S/F systems, but their interpretations were controversial (as a review see [18]) due to the very small value of the characteristic length ξ_{f2} (only several nanometers). The most direct proof of the ‘ π ’-phase existence would be the observation following the theoretical predictions [6,12] of the vanishing of the critical current at the ‘0’- to ‘ π ’-phase transition. The first experimental evidence of a 0– π transition in S/F/S (Nb–Cu_xNi_{1–x}–Nb) Josephson junction was obtained in [19] from the measurements of the temperature dependence of the critical current. The 0– π transition was signaled by the vanishing of the critical current with the temperature decrease. Such a behavior is observed for a F layer thickness d close to some critical value d_c . In fact, it simply means that the critical thickness d_c slightly depends on temperature. The temperature variation serves as a fine tuning and permits one to study this transition in detail. Later the damped oscillations of the critical current as a function of F layer thickness were observed in Nb/Al/Al₂O₃/PdNi/Nb [22] and Nb/Cu/Ni/Cu/Nb [23] junctions. The very recent experiments [21] have enabled us to observe the two-node thickness dependence of the critical current in Josephson SFS junctions with a ferromagnetic interlayer, i.e., both direct transition into π -state and reverse one from π - into 0-state (Fig. 4). This revealed that the ‘0’- to ‘ π ’-transition with the F layer thicknesses observed in [19] was the second one. The first transition occurs for an F layer thickness around 10 nm. The temperature dependences of the critical current near the first and the second ‘0’- to ‘ π ’-transitions are presented in Fig. 5.

The complete quantitative analysis of the S/F/S junctions is rather complicated, because the ferromagnetic layer may strongly modify superconductivity near the S/F interface. In addition, the boundary transparency and electron mean free path, as well as magnetic scattering, are important parameters affecting the critical current. In the case of small conductivity of the F layer or small interface transparency $\gamma_B : \sigma_f \xi_s / \sigma_s \xi_f \ll \max(1, \gamma_B)$ we may use the ‘rigid boundary’ conditions [24] with $F_s(-d_f/2) = \Delta e^{-i\varphi/2} / \sqrt{\omega^2 + \Delta^2}$ and $F_s(d_f/2) = \Delta e^{i\varphi/2} / \sqrt{\omega^2 + \Delta^2}$, where φ is the phase difference on the junction. We consider the junction with F layer thickness d_f and the x axis is chosen perpendicular to the F layer.

The solution of Eq. (9) in a ferromagnet satisfying the corresponding boundary conditions is written as

$$F(x) = \frac{\Delta}{\sqrt{\omega^2 + \Delta^2}} \left[\frac{\cos(\varphi/2) \cosh(kx)}{(\cosh(kd_f/2) + k\gamma_B \xi_n \sinh(kd_f/2))} + \frac{i \sin(\varphi/2) \sinh(kx)}{(\sinh(kd_f/2) + k\gamma_B \xi_n \cosh(kd_f/2))} \right] \quad (12)$$

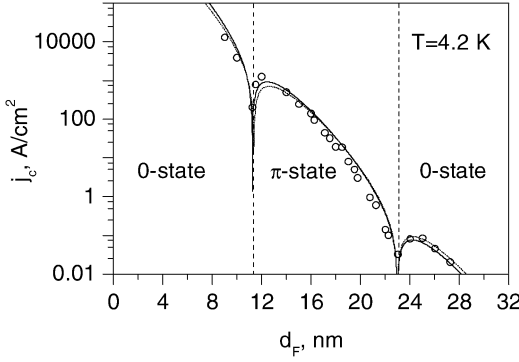


Fig. 4. The double-reversal F-layer thickness dependence of the critical current density for Nb/Cu_{0.47}Ni_{0.53}/Nb junctions at temperature 4.2 K [21]. The open circles present experimental results, the dashed line shows the fit using Eq. (14).

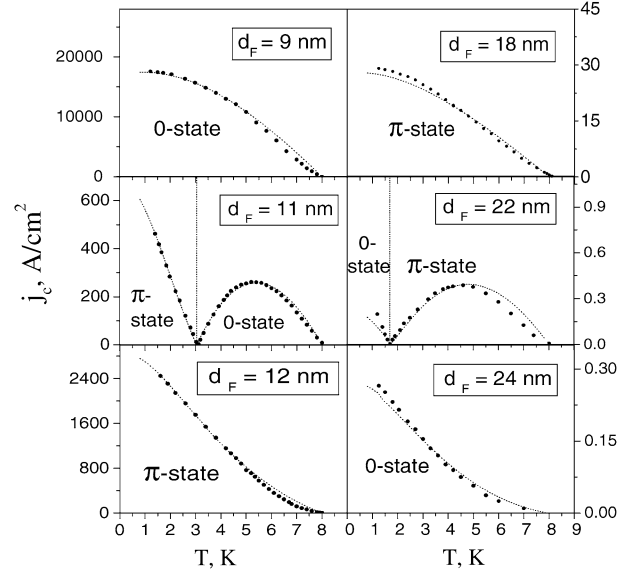


Fig. 5. Temperature dependence of the Nb/Cu_{0.47}Ni_{0.53}/Nb junctions critical current density at several F-layer thicknesses close to the critical ones [21]. The dashed lines show the calculation results based on the solution of the nonlinear Usadel equation [21].

where the complex wave-vector $k = \xi_{f1}^{-1} + i\xi_{f2}^{-1}$. This solution describes the $F(x)$ behavior near the critical temperature and gives the sinusoidal current-phase dependence $I_s(\varphi) = I_c \sin(\varphi)$ with the critical current

$$I_c = eSN(0)D_f\pi T \sum_{-\infty}^{\infty} \frac{\Delta^2}{\omega^2} \frac{2k/\cosh(kd_f)}{\tanh(kd_f)(1 + \Gamma_\omega^2 k^2) + 2k\Gamma_\omega} \quad (13)$$

where $\Gamma_\omega = \gamma_B \xi_n / |G_s|$. This expression may be generalized to take into account the different interface transparencies $\gamma_{B1}, \gamma_{B2} \gg 1$, it is enough to substitute in Eq. (13) $\Gamma_\omega^2 \rightarrow \gamma_{B1}\gamma_{B2}(\xi_n/|G_s|)^2$ and $2\Gamma_\omega \rightarrow (\gamma_{B1} + \gamma_{B2})\xi_n/|G_s|$.

In the most interesting limit from the practical point of view, when the F layer thickness $d_f > \xi_{f1}$ we obtain the universal expression for the $I_c(d_f)$ dependence

$$I_c \sim \exp\left(-\frac{d_f}{\xi_{f1}}\right) \left[\sin\left(\frac{d_f}{\xi_{f2}} + \Psi\right) \right] \quad (14)$$

where the angle $\pi/4 < \Psi < \pi/2$ depends on the magnetic scattering amplitude and the boundary transparencies. This expression gives a good description of the available experimental data (Fig. 4) permitting one to find the lengths ξ_{f1} and ξ_{f2} and then proves the important influence of the magnetic scattering on the properties of the S/F/S junctions. Formula (14) is strictly applicable near T_c but gives also a good approximation for the $I_c(d_f)$ dependence at low temperature everywhere except close to the critical thicknesses d_c corresponding to ‘0’–‘ π ’ transitions. The temperature dependence of the critical current is very peculiar for $d_f \approx d_{fc}$ and the corresponding experimental data are presented in Fig. 5. The theoretical description of these $I_c(T)$ dependences requires the solution of the nonlinear Usadel equation (see [21] and for more details [1]).

Bulaevskii et al. [25] pointed out that a ‘ π ’-junction incorporated into a superconducting ring would generate a spontaneous current and the corresponding magnetic flux would be half a flux quantum $\Phi_0 = h/2e$. The appearance of the spontaneous current is related to the fact that the ground state of the ‘ π ’-junction corresponds to the phase difference π and so this phase difference will generate a supercurrent in the ring which short circuits the junction. Naturally the spontaneous current is generated if there is an odd number of ‘ π ’-junctions in the ring. This circumstance has been exploited in a elegant way [20] to provide unambiguous proof of the ‘ π ’-phase transition. The observed half-period shift of the external magnetic field dependence of the transport critical current in triangular S/F/S arrays is presented in Fig. 6. The thickness of F layers of the S/F/S junctions was chosen in such a way that the junction nature changed from ‘0’ to ‘ π ’ with temperature variation.

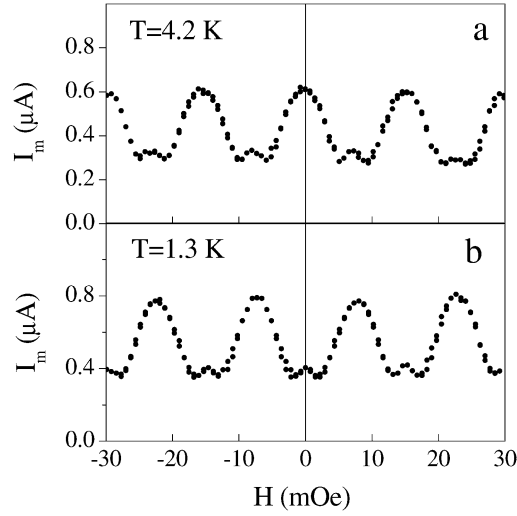


Fig. 6. Magnetic field dependence of the critical transport current for the network of five Nb/Cu_{0.46}Ni_{0.54}/Nb sandwiches at temperature (a) above and (b) below the transition to the π -state [20].

6. How the transition from ‘0’- to ‘ π ’-states occurs?

The current-phase relation for a Josephson junction is sinusoidal only near the critical temperature T_c [9]

$$j(\varphi) = I_1 \sin \varphi \quad (15)$$

At low temperature the higher harmonic terms appear. However, in the diffusive limit at $d_f > \xi_1$ they are very small and in the usual junctions their presence is to be hardly observed. In S/F/S junctions in general, in the dirty limit $I_1 \sim \exp(-d_f/\xi_1)$ and the second harmonic contribution happens to be very small and positive $\sim \exp(-2d_f/\xi_1)$ [26]. The peculiarity of the situation with the $0-\pi$ transition is that in the transition region the first harmonic term changes its sign passing through zero and then the role of the second harmonic contribution becomes predominant! To study the scenario of the $0-\pi$ transition we address to the general current-phase relation

$$j(\varphi) = I_1 \sin \varphi + I_2 \sin 2\varphi \quad (16)$$

which corresponds to the following phase dependent contribution to energy of the Josephson junction

$$E_J(\varphi) = \frac{\Phi_0}{2\pi c} \left[-I_1 \cos \varphi - \frac{I_2}{2} \cos 2\varphi \right] \quad (17)$$

If we neglect the second harmonic term, then the 0 state occurs for $I_1 > 0$. Near a $0-\pi$ transition $I_1 \rightarrow 0$ and the second harmonic term becomes important. The critical current at the transition $j_c = |I_2|$ and if $I_2 > 0$, the minimum energy always occurs at $\varphi = 0$ or $\varphi = \pi$, see Fig. 7.

In the opposite case ($I_2 < 0$) the transition from 0 to π state is continuous and there is region where the equilibrium phase difference takes any value $0 < \varphi < \pi$. The characteristics of such a ‘ φ -junction’ are very peculiar [27].

On experiment the second harmonic at $0-\pi$ transition was observed in Nb–Cu_{0.52}Ni_{0.48}–Nb junctions [30] for the F-layer thickness $d_f \approx 17$ nm (which corresponds to the first $0-\pi$ transition with thickness increase) at 1.1 K. For junctions with $x = 0.48$ [29] we may roughly estimate $\xi_1 \approx 4$ nm, $\xi_2 \approx 9$ nm, the magnetic scattering parameter $\alpha \approx 0.9$, $\xi_f \approx 6$ nm and $h \approx 100$ K and we have for the ratio $I_2/I_1 \sim 0.1 \exp(-d/\xi_1) \sim 10^{-3}$ which is somewhat smaller than the observed value 3×10^{-3} [30]. The measurements [30] could not provide information about the sign of the second harmonic. In the experiments [28] the second harmonic was also searched in the series of Nb–Cu_{0.47}Ni_{0.53}–Nb junctions near the second $0-\pi$ transition at $d_f \approx 22$ nm (the first transition occurs at $d_f \approx 11$ nm [21]). From the thickness dependence of the critical current in the series of [21] we may estimate $\xi_1 \approx 1.5$ nm and $\xi_2 \approx 3.5$ nm which gives $I_2/I_1 \sim 0.1 \exp(-d_f/\xi_1) \sim 10^{-8}$, which gives a very small value for $I_2 \lesssim 10^{-11}$ A, well below the experimental threshold.

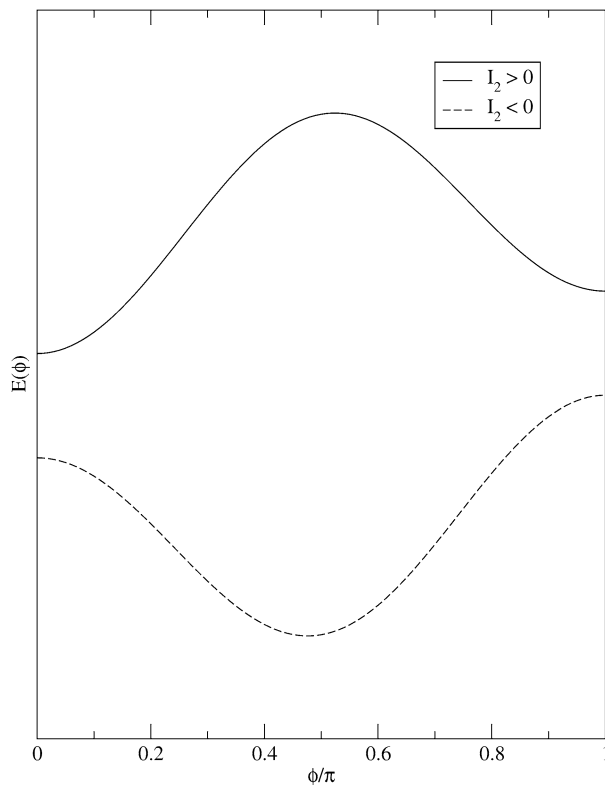


Fig. 7. Schematic plot of the phase-dependent Josephson junction's energy. The case $I_2 > 0$ corresponds to the discontinuous $0-\pi$ transition while at $I_2 < 0$ the minimum energy is reached at $0 < \varphi < \pi$.

However, the estimates of the positive intrinsic second harmonic term presented above presume that the junction is ideal. The thickness variation of the F layer switch on the another mechanism of the second order harmonic generation [27]. Indeed, the roughness of F layer in the real S/F/S junctions is of the order of 1 nm. This means that if the characteristic length δl of the thickness variation (along the contact surface) is larger than d_f , the critical current will vary locally as well. On the other hand if δl is much smaller than the Josephson length λ_J , which for the current density 10^6 A/m² [16] is of the order of the junction dimension (50×50 μm² in [16]), the measured characteristics of the junction will be effectively averaged. At the $0-\pi$ transition we deal with a system where the local current density is alternating $\pm I_1$ and $\bar{I}_1 = 0$. The resulting local phase variation leads to the appearance of the *negative* second harmonic in the averaged current-phase relation [31,27] $I_2 \sim -|I_1|(\delta l/\lambda_J)^2$, where λ_J is the Josephson length corresponding to the current density I_1 . The 1 nm roughness of the F layer in the experiments [30,29] permits one to estimate for $d_f \approx 17$ nm the value $|I_1| \sim 5 \times 10^6$ A/m² and $\lambda_J \sim (10-100)$ μm. At the present time there is no information about the characteristic length δl of thickness variation in the studied S/F/S junctions. Taking it as 1 μm for the 10×10 μm² junction [29,30] we have $I_2 \sim -5 \times (10^2-10^4)$ A/m² while the experimentally observed value is $\sim 3 \times 10^4$ A/m² and the sign of I_2 is unknown.

Apparently this mechanism was responsible for the very recent second harmonic observation in Nb–Cu_{0.47}Ni_{0.53}–Nb junctions with spatial variation in the barrier thickness [32]. Unfortunately the interesting question about the origin of the second harmonic generation at $0-\pi$ transition in S/F/S junctions is still open. The response on this question may shed light of the sign of the second harmonic and then on the fascinating possibility to have an experimental realization of the new type of the Josephson junction: ‘ φ -junction’.

7. Conclusion

During the last five years an enormous progress in the controllable fabrication of the superconductor–ferromagnet heterostructures has been achieved. The peculiar effects predicted earlier have been observed in experiments and we

have a general understanding of the mechanism of the superconductivity and ferromagnetism interplay in S/F systems. Now this domain of research enters into a period when the design of the new types of the devices becomes feasible and we may expect many interesting findings in the near future.

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References

- [1] A.I. Buzdin, *Rev. Mod. Phys.* 77 (2005) 935.
- [2] F.S. Bergeret, A.F. Volkov, K.B. Efetov, *Rev. Mod. Phys.* 77 (2005) 1321.
- [3] A.I. Larkin, Y.N. Ovchinnikov, *Zh. Eksp. Teor. Fiz.* 55 (1968) 2262, *Sov. Phys. JETP* 28 (1965) 1200.
- [4] P. Fulde, R.A. Ferrell, *Phys. Rev.* 135 (1964) 1550.
- [5] L.G. Aslamazov, *Zh. Eksp. Teor. Fiz.* 55 (1968) 1477, *Sov. Phys. JETP* 28 (1969) 773.
- [6] A.I. Buzdin, L.N. Bulaevskii, S.V. Panyukov, *Pis'ma Zh. Eksp. Teor. Phys.* 35 (1982) 147, *JETP Lett.* 35 (1982) 178.
- [7] A.I. Buzdin, M.Yu. Kuprianov, *Pis'ma Zh. Eksp. Teor. Phys.* 52 (1990) 1089, *JETP Lett.* 52 (1990) 487.
- [8] Z. Radovic, M. Ledvij, L. Dobrosaljevic-Grujic, A.I. Buzdin, J.R. Clem, *Phys. Rev. B* 44 (1991) 759.
- [9] P.G. de Gennes, *Superconductivity of Metals and Alloys*, Benjamin, New York, 1966.
- [10] L. Usadel, *Phys. Rev. Lett.* 95 (1970) 507.
- [11] A. Buzdin, *Phys. Rev. B* 62 (2000) 11377.
- [12] A.I. Buzdin, M.Yu. Kuprianov, *Pis'ma Zh. Eksp. Teor. Phys.* 53 (1991) 308, *JETP Lett.* 53 (1991) 321.
- [13] T.M. Kontos, M. Aprili, J. Lesueur, X. Grison, *Phys. Rev. Lett.* 86 (2001) 304.
- [14] J.S. Jiang, D. Davidović, D.H. Reich, C.L. Chien, *Phys. Rev. Lett.* 74 (1995) 314.
- [15] Ya.V. Fominov, N.M. Chitchev, A.A. Golubov, *Phys. Rev. B* 66 (2002) 014507.
- [16] V.V. Ryazanov, V.A. Oboznov, A.S. Prokof'ev, V.V. Bolginov, A.K. Feofanov, *J. Low Temp. Phys.* 136 (2004) 385.
- [17] Y. Obi, M. Ikebe, H. Fujishiro, *Phys. Rev. Lett.* 94 (2005) 057008.
- [18] A.S. Sidorenko, V.I. Zdravkov, A.A. Prepelitsa, C. Helbig, Y. Luo, S. Gsell, M. Schreck, S. Klimm, S. Horn, L.R. Tagirov, R. Tidecks, *Ann. Phys.* 12 (2003) 37.
- [19] V.V. Ryazanov, V.A. Oboznov, A.Yu. Rusanov, A.V. Veretennikov, A.A. Golubov, J. Aarts, *Phys. Rev. Lett.* 86 (2001) 2427.
- [20] V.V. Ryazanov, V.A. Oboznov, A.V. Veretennikov, A.Yu. Rusanov, *Phys. Rev. B* 65 (2001) R020501.
- [21] V.A. Oboznov, V.V. Bol'ginov, A.K. Feofanov, V.V. Ryazanov, A.I. Buzdin, *cond-mat/0508573*.
- [22] T.M. Kontos, M. Aprili, J. Lesueur, F. Genet, B. Stephanidis, R. Boursier, *Phys. Rev. Lett.* 89 (2002) 137007.
- [23] Y.A. Blum, A. Tsukernik, M. Karpovski, A. Palevski, *Phys. Rev. Lett.* 89 (2002) 187004.
- [24] A.A. Golubov, M.Yu. Kupriyanov, E. Il'ichev, *Rev. Mod. Phys.* 76 (2004) 411.
- [25] L.N. Bulaevskii, V.V. Kuzii, A.A. Sobyenin, *Pis'ma Zh. Eksp. Teor. Phys.* 25 (1977) 314, *JETP Lett.* 25 (1977) 290.
- [26] A. Buzdin, *Phys. Rev. B* 72 (2005) R100501.
- [27] A.I. Buzdin, A. Koshelev, *Phys. Rev. B* 67 (2003) 220504(R).
- [28] S.M. Frolov, D.J. Van Harlingen, V.A. Oboznov, V.V. Bol'ginov, V.V. Ryazanov, *Phys. Rev. B* 70 (2004) 144505.
- [29] H. Sellier, C. Baraduc, F. Lefloch, R. Calemczuk, *Phys. Rev. B* 68 (2003) 054531.
- [30] H. Sellier, C. Baraduc, F. Lefloch, R. Calemczuk, *Phys. Rev. Lett.* 92 (2004) 257005.
- [31] R.G. Mints, *Phys. Rev. B* 57 (2001) R3221.
- [32] S.M. Frolov, D.J. Van Harlingen, V.V. Bol'ginov, V.A. Oboznov, V.V. Ryazanov, *cond-mat/0506003*.