HAL
open science

First order 0-pi phase transitions in superconductor/ferromagnet/superconductor trilayers

A. V. Samokhvalov, Alexandre I. Buzdin

To cite this version:

A. V. Samokhvalov, Alexandre I. Buzdin. First order 0-pi phase transitions in superconductor/ferromagnet/superconductor trilayers. Physical Review B: Condensed Matter and Materials Physics (1998-2015), 2015, 92 (5), pp.054511 (1-11). 10.1103/PhysRevB.92.054511 . hal-01216388

HAL Id: hal-01216388

https://hal.science/hal-01216388v1

Submitted on 16 Oct 2015

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Distributed under a Creative Commons Attribution - ShareAlike 4.0 International License

arXiv:1509.02330v1 [cond-mat.supr-con] 8 Sep 2015

First order 0 - π phase transitions in superconductor/ferromagnet/superconductor trilayers

A. V. Samokhvalov[1,2] and A. I. Buzdin[3]

[1] *Institute for Physics of Microstructures, Russian Academy of Sciences, 603950 Nizhny Novgorod, GSP-105, Russia*

[2] *Lobachevsky State University of Nizhny Novgorod, Nizhny Novgorod 603950, Russia*

[3] *Institut Universitaire de France and University of Bordeaux, LOMA UMR-CNRS 5798, F-33405 Talence Cedex, France*

(Dated: September 9, 2015)

We study the thermodynamics of the diffusive SFS trilayer composed of thin superconductor (S) and ferromagnet (F) layers. On the basis the self-consistent solutions of nonlinear Usadel equations in the F and S layers we obtain the Ginzburg–Landau expansion and compute the condensation free energy and entropy of the 0 (even) and π (odd) order parameter configurations. The first order $0-\pi$ transition as a function of temperature T occurs, which is responsible for a jump of the averaged magnetic field penetration depth $\lambda(T)$ recently observed on experiments [N.Pompeo, et. al., Phys. Rev. B **90**, 064510 (2014)]. The generalized Ginzburg-Landau functional was proposed to describe SFS trilayer for arbitrary phase difference between the superconducting order parameters in the S layers. The temperature dependence of the SFS Josephson junction critical current demonstrates the strong anharmonicity of the corresponding current–phase relation in the vicinity of the $0-\pi$ transition. In rf SQUID, coexistence of stable and metastable 0 and π states provides integer and half–integer fluxoid configurations.

PACS numbers: 74.45.+c, 74.78.Na, 74.78.-w

I. INTRODUCTION

The ground state of the superconductor-ferromagnet-superconductor (SFS) trilayer at zero current can be 0 or π state, depending on the value of the phase difference between the superconducting order parameters in the two S electrodes. This phenomenon is related to the damped oscillatory behavior of the Cooper pair wave function in the ferromagnet due to the proximity effect[1,2] (for more references and reviews, see Refs. 3–5). Usually experiments directed towards the observation of the $0-\pi$ crossover in SFS trilayer were concentrated on the measurements of the critical Josephson current I_c of the SFS junction[6–9]. The $0-\pi$ transition manifests itself in the vanishing of I_c if higher–order harmonics of the current–phase relation are negligible[10–12].

Recently, an unusual electromagnetic response of SF systems was reported as a manifestation of the Cooper pair wavefunction oscillations inside the ferromagnet. Measurements of the London penetration depth in thin Nb/Ni bilayers[13] reveal a slightly nonmonotonic dependence of the penetration depth on the F layer thickness, which was in accordance with the theoretical analysis[14]. Anomalous Meissner effect in hybrid SF structures was the subject of several theoretical works[15–17] predicting an unusual paramagnetic response of such systems. Vanishing or inversion of the Meissner effect is believed to be attributed to spin-triplet superconducting correlations[18,19] generated in inhomogeneous F layer due to proximity effect and should result in the in-plane Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) instability[20–22]. Unusual drop of the screening with decrease of temperature was observed recently in[23] by microwave measurements of the London penetration depth λ in $Nb/Pd_{0.84}Ni_{0.16}/Nb$ trilayers.

The transition temperature of SF structures into the normal state has been examined both theoretically[24–27] and experimentally[28–31] (see Ref. 3,32, for reviews). However, the study of the thermodynamic properties of the phase transition between 0 and π states of SF hybrids is more sparse. A first-order $0-\pi$ transition was predicted for diffusive SFS junctions with a homogeneous F barrier, if the current–phase relation takes into account the second harmonic contribution[11]. Experimental evidence of a $0-\pi$ transition in SFS ($Nb/Cu_xNi_{1-x}/Nb$) Josephson junction was obtained from the measurements of the temperature dependence of the critical current in Refs. 8,9,33. Note that a small modulation of the thickness of the barrier may favor the continuous $0-\pi$ transition[11]. A first-order transitions between 0 and π states by temperature variation were demonstrated in both the clean[34,35] and dirty[27] limits using numerical self-consistent solutions of the microscopic Bogoliubov-de-Gennes[36,37] or Usadel[38] equations, respectively. The $0-\pi$ transition in ballistic SF systems has been shown to have a pronounced effect on the distribution of the Cooper pair wavefunction in the F region and the amplitude of order parameter $\Delta(T)$ in S–layers[34,35]. The theoretical model Proposed in Ref. 23 argued that the observed jump $\lambda(T)$ in $Nb/Pd_{0.84}Ni_{0.16}/Nb$ trilayers is related to the first order phase transition from 0 to π state on cooling.

In this work we develop a theoretical approach based on the nonlinear Usadel equations providing a general description of diffusive SFS junction with thin superconducting layers at the transition from 0 to π state. The

leakage of the Cooper pairs weakens the superconductivity near the interface with a F metal due to the proximity effect[3]. The magnitude of the superconducting order parameter suppression depends on the parameters characterizing the system such as the SF interface transparency, the thickness of the S and F layers, etc. For large interface transparency this effect seems to be especially strong and results in suppression of the superconducting order parameter and the transition temperature T_c of a thin superconducting layer in contact with a ferromagnet metal. If the thickness of a superconducting layer is smaller than a critical one, the proximity effect totally destroys a superconductivity. From the self-consistent solutions we obtain the Ginzburg–Landau expansion and compute the condensation free energy and entropy of the possible order parameter configurations as a function of temperature T. As T varies, we find that the first order phase transition between 0 and π states occurs, which is responsible for a jump of the averaged penetration depth $\lambda(T)$ observed in Ref. 23. We also calculate the current–phase relation $I(\varphi)$ of the SFS junction which reveals strong contribution of the higher harmonic terms. The $0-\pi$ states coexistence and switching leads to new modes of magnetic flux penetration in superconducting loop containing the SFS junction.

The paper is organized as follows. In Sec. II we briefly discuss the basic equations. We analyze the case of thin S layers and obtain the approximate solutions of the nonlinear Usadel equations in F layer near the superconducting critical temperature $T \lesssim T_c$. In Sec. III we find the temperature $T_c^{0,\pi}$ of the second–order superconducting phase transition to the normal state for 0 and π order-parameter configurations. The Sec. IV is devoted to the analysis of the temperature-driven transition between 0 and π states. We obtain the Ginzburg–Landau expansion and find the ground states of SFS junction near the critical temperatures $T_c^{0,\pi}$. In Sec. V we generalize the Ginzburg–Landau description for arbitrary phase difference φ between the superconducting order parameters of the S layers and find the strongly nonsinusoidal current–phase relation of the SFS junction in the vicinity of $0-\pi$ transition. In Sec. VI we show that the coexistence of 0 and π states leads to peculiarities of the magnetic flux penetration in superconducting loop with a single SFS junction. Sec. VII contains a brief summary and discussion.

II. MODEL AND BASIC EQUATIONS

Let us consider a SFS trilayers with a transparent SF interfaces and thin S layers of thickness $d_s \sim \xi_s$, where ξ_s is the superconducting coherence length. The considered geometry of the SFS structure is presented in Fig. 1. In the previous observations of 0-π transition in SFS junctions by temperature variation[6,9] the superconducting electrodes were rather thick to overcome the pair–breaking proximity effect of F layer and then the influence of the 0-π transition on the superconducting order parameter $\Delta(T)$ in the electrodes was negligible. From the theoretical point of view, weak depairing in S electrodes means that the pair potential at the SF interfaces is equal to its bulk value (the so-called rigid–boundary condition[5]). Here we study the SFS structure with relatively thin S layers and demonstrate that the 0-π transition leads to the jump of the amplitude of the superconducting order parameter, providing the anomalous temperature behavior of the effective penetration depth of the whole structure.

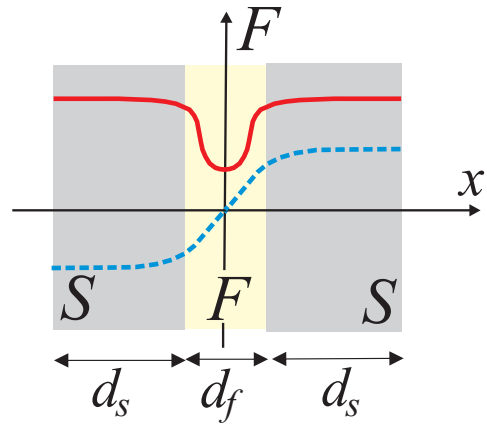

FIG. 1: (Color online) The schematic behavior of the pair wave function $F(x) = F_{s,f}$ inside the SFS trilayers. The red solid line represents approximately the behavior of the pair wave function in an even mode (0-state). Due to symmetry the derivative $\partial_x F_f$ is zero at the center of F layer. The pair wave function in the odd mode (blue dashed line) vanishes at the center of F layer, and $F(x)$ has a $\pi-$shift in diametrically opposite points ($\pi-$state).

To elucidate our results we start the qualitative discussion of the proximity effect on the properties of SFS sandwiches if the thickness of S layers is small enough. Due to the damped oscillations of the pair wave function F in the ferromagnetic layer, two different order–parameter configurations are possible inside the SFS trilayers (see Fig. 1). The first one corresponds to the case than the order parameter is an even function of the coordinate x, chosen perpendicular to the layers, and does not change its sign in F layer. It means that in the ground state the superconducting phase in both S layers must be the same ($0-$phase). For the second case the pair wave function is odd in x and cross zero at the center of the F layer which causes a $\pi-$shift in the superconducting phase of different S layers. This configuration corresponds to the $\pi-$phase of the SFS structure. Since pair–breaking proximity effect depends on the structure of the Copper pairs wavefunction in ferromagnetic layer, a suppression of superconductivity in thin S layers is expected to be different for 0 and π states. As a result the equilibrium superconducting gaps $\Delta_{0,\pi}$ for 0 and π states are different ($\Delta_0(T) \neq \Delta_\pi(T)$), and this leads to the different effective penetration depth $\lambda(T) \sim 1/\Delta(T)$ in 0 and π

states. So, 0-π transition in SFS trilayers has to be accompanied by a jump of λ. The coexistence of stable and metastable states in the vicinity of $0-\pi$ transition leads to a strong anharmonicity of the current-phase relation in the SFS junction under consideration. As a result, peculiarities of the magnetic flux penetration in a mesoscopic superconducting loop containing the SFS junction are expected.

Taking in mind that the F interlayer is dilute ferromagnetic alloys like Cu_xNi_{1-x} or Pd_xNi_{1-x} we use the Usadel equations[38] which are convenient in the diffusive limit (see[3] for details). Moreover it is important to take into account the magnetic disorder which is already present in the magnetic alloys and provides the main mechanism of the temperature induced 0-π transition[9,39]. We describe it by introducing the magnetic scattering time τ_s. Note that in practice the exchange field h acting on the electron's spins in the ferromagnet and magnetic scattering rate τ_s^{-1} are much larger than the superconducting critical temperature T_c: h, $\tau_s^{-1} \gg T_c$. The complete nonlinear Usadel equations for the normal $G(x,\omega,h)$ and anomalous $F(x,\omega,h)$ Green's functions in F layer are[9,39]:

$$-\frac{D_f}{2}\left[G(x,\omega,h)\,\partial_x^2 F(x,\omega,h) - F(x,\omega,h)\,\partial_x^2 G(x,\omega,h)\right] + \left[\omega + ih + \frac{G(x,\omega,h)}{\tau_s}\right] F(x,\omega,h) = 0\,, \qquad (1)$$

$$G^2(x,\omega,h) + F(x,\omega,h)F^+(x,\omega,h) = 1\,. \qquad (2)$$

Here D_f is the diffusion constant in the ferromagnet, $\omega = 2\pi T(n+1/2)$ is a Matsubara frequency at the temperature T. The equation for the function $F^+(x,\omega,h) = F^*(x,\omega,-h)$ coincides with Eqn. (1)[3]. For the $0-$state we should choose the even anomalous Green's functions F while for the $\pi-$state it should be the odd one. Using the usual parametrization of the normal and anomalous Green functions $G_s = \cos\theta_s$ and $F_s = \sin\theta_s$, the complete nonlinear Usadel equation in superconucting layers can be written for $\omega > 0$ as[3]

$$-\frac{D_s}{2}\partial_x^2\theta_s + \omega\sin\theta_s = \Delta\cos\theta_s\,, \qquad (3)$$

where D_s is the diffusion constant in the superconductor. Assuming the SF interfaces to be transparent we have at $x = \pm d_f/2$[40]:

$$F_s = F, \quad \sigma_s\,\partial_x F_s = \sigma_f\,\partial_x F\,, \qquad (4)$$

where σ_f and σ_s are the normal–state conductivities of the F and S metals, respectively. The boundary condition at the outer surfaces $x = \pm(d_s + d_f/2)$ is

$$\partial_x F_s = 0\,. \qquad (5)$$

For thin S-layers $d_s \lesssim \xi_s = \sqrt{D_s/2\pi T_{c0}}$, the inverse proximity effect is substantial, and the Usadel equation (3) for the S layers have to be completed by the self-consistency equation for the superconducting order parameter $\Delta(x)$:

$$\Delta(x) = \pi T\rho\sum_\omega F_s(x,\omega), \qquad (6)$$

where ρ is BCS coupling constant and T_{c0} is the critical temperature of a bulk sample of the material S.

For 0 and π states of SFS trilayers we have $F^+(x,\omega,h) = F^*(x,\omega,-h) = F(x,\omega,h)$, and one can replace $F^+(x,\omega,h)$ by $F(x,\omega,h)$ in Eqn. (2). Just below the critical temperature T_c anomalous Green's functions are small and the condition (2) can be rewritten as

$$G(x,\omega,h) \simeq 1 - F^2(x,\omega,h)/2\,. \qquad (7)$$

For $d_s \ll \xi_s$, where $\xi_s = \sqrt{D_s/2\pi T_{c0}}$ is the superconducting coherence length, the variations of the functions $\theta_s(x)$ and $\Delta(x)$ in the superconducting layers are small: $\theta_s(x) \simeq \theta_s$, $\Delta(x) \simeq \Delta$. Therefore, we can average Eq. (3) over the thickness of the S layers, using the boundary condition (5). Finally, we obtain the following boundary condition:

$$\left.\frac{\partial\theta_s}{\partial s}\right|_{s_f} = \frac{d_s\xi_f}{\xi_s^2}\left(\frac{\Delta\cos\theta_s - \omega\sin\theta_s}{\pi T_{c0}}\right), \qquad (8)$$

$$\left.\frac{\partial\theta_s}{\partial s}\right|_{-s_f} = \mp\left.\frac{\partial\theta_s}{\partial s}\right|_{s_f}, \qquad (9)$$

where $s = x/\xi_f$ and $s_f = d_f/2\xi_f$. The top (bottom) sign in (9) corresponds to the $0-$phase ($\pi-$phase), respectively.

Applying the method[11] we can find the approximate solution of the non-linear Usadel equation (1) in F layer near the superconducting critical temperature $T \lesssim T_c$. For the $0-$state we should choose the even anomalous Green's functions F while for the $\pi-$state it should be the odd one:

$$F(s,\omega) \simeq \begin{cases} a\cosh(qs) \\ b\sinh(qs) \end{cases} - \qquad (10)$$
$$\frac{1}{8k^2}\left(\alpha + \frac{3i}{4}\right)\begin{cases} a^3\cosh(3ks), & 0-\text{phase} \\ b^3\sinh(3ks), & \pi-\text{phase} \end{cases},$$

where $\xi_f^2 = D_f/h$, $k^2 = 2(\omega/h + i + \alpha\,\mathrm{sgn}(\omega))$ and $\alpha = 1/\tau_s h$ is the dimensionless magnetic scattering rate. The complex wave vector q is determined by the relations:

$$q^2 = k^2 \mp a^2\,(\alpha + i/4) \qquad (11)$$

for 0 and π phases, respectively. If $T_c < \tau_s^{-1}$, h, we may neglect the Matsubara frequencies in the Eq. (1) assuming that $k^2 = (k_1 + ik_2)^2 = 2(\alpha + i)$ for $\omega > 0$:

$$k_1 = \sqrt{\sqrt{1+\alpha^2} + \alpha}\,, \quad k_2 = \sqrt{\sqrt{1+\alpha^2} - \alpha}\,, \qquad (12)$$

Then the decay characteristic length ξ_{f1} and the oscillation period ξ_{f2}, may be written as

$$\xi_{f1} = \xi_f/k_1\,, \quad \xi_{f2} = \xi_f/k_2\,. \tag{13}$$

The ratio of the characteristic lengths $\xi_{f1}/\xi_{f2} < 1$ clearly shows that magnetic scattering decreases the decay length and increases the oscillation period[39].

A. Even mode (0−phase)

In the limit $|a| \ll 1$ we obtain from the even solution (10,11) the expansion of F in powers of the amplitude a:

$$\begin{aligned} F(s) &\simeq f\cosh(ks) - f^3 g_0(s) + o(f^4)\,, \\ g_0(s) &= \frac{1}{8k^2}\left[4ks(\alpha+i/4)\sinh(ks)\right. \\ &\quad \left.+(\alpha+3i/4)\cosh(3ks)\right]\,. \end{aligned} \tag{14}$$

Using the first boundary condition (4) at $s = s_f$ we get the relation between the amplitude a and Green's function $F_s = \sin\theta_s$ in superconductor:

$$f = f_0 + f_0^3\frac{g_0(s_f)}{\cosh(ks_f)}\,, \quad f_0 = \frac{F_s}{\cosh(ks_f)}\,. \tag{15}$$

Substitution of the solution (14) to the relations (8,9) results in the following equation with respect to the amplitude F_s in the S layers:

$$(\omega + 1/\tau_0)\,F_s = \Delta - \frac{1}{2}\left(\Delta F_s^2 + \varepsilon\Lambda_0 F_s^3\right) \tag{16}$$

where

$$\tau_0^{-1} = \varepsilon\pi T_{c0} k\,\tanh(ks_f) \tag{17}$$

is the depairing parameter of even mode and

$$\begin{aligned} \Lambda_0 &= \pi T_{c0}\left[\frac{i}{2k}\tanh(ks_f) - \right. \\ &\quad \left.\frac{\alpha+i/4}{k}\frac{\tanh(ks_f)}{\cosh^2(ks_f)} - \frac{s_f(\alpha+i/4)}{\cosh^4(ks_f)}\right]\,. \end{aligned} \tag{18}$$

Here the key parameter

$$\varepsilon = \frac{\sigma_f}{\sigma_s}\frac{\xi_s^2}{d_s\xi_f}$$

determines the influence of the proximity effect on the S layers.

B. Odd mode (π−phase)

In the limit $|b| \ll 1$ we obtain from the odd solution (10,11) the following expansion of F in powers of the amplitude b:

$$\begin{aligned} F(s) &\simeq f\sinh(ks) + f^3 g_\pi(s) + o(f^4)\,, \\ g_\pi(s) &= \frac{1}{8k^2}\left[4ks(\alpha+i/4)\cosh(ks)\right. \\ &\quad \left.-(\alpha+3i/4)\sinh(3ks)\right]\,. \end{aligned} \tag{19}$$

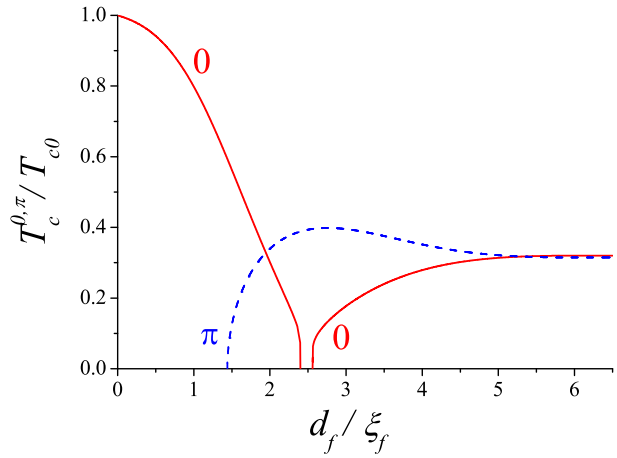

FIG. 2: (Color online) The typical dependence of the critical temperature $T_c^{0,\pi}$ on the thickness of F layer d_f for even mode(0−phase) (solid red line) and for odd mode(π−phase) (dashed blue line). Here we choose: $d_s = 2\xi_s$; $\sigma_f/\sigma_s = 0.12$; $\xi_s/\xi_f = 3$ ($\varepsilon = 0.18$); $h\tau_s = 7$.

Using the first boundary condition (4) at $s = s_f$ we get the relation between the amplitude b and Green's function $F_s = \sin\theta_s$ in superconductor:

$$f = f_\pi - f_\pi^3\frac{g_\pi(s_f)}{\sinh(ks_f)}\,, \quad f_\pi = \frac{F_s}{\sinh(ks_f)}\,. \tag{20}$$

Substitution of the solution (19) to the relations (8,9) results in:

$$(\omega + 1/\tau_\pi)\,F_s = \Delta - \frac{1}{2}\left(\Delta F_s^2 + \varepsilon\Lambda_\pi F_s^3\right)\,, \tag{21}$$

where

$$\tau_\pi^{-1} = \varepsilon\pi T_{c0} k\,\coth(ks_f) \tag{22}$$

is the depairing parameter of odd mode and

$$\begin{aligned} \Lambda_\pi &= \pi T_{c0}\left[\frac{i}{2k}\coth(ks_f) + \right. \\ &\quad \left.\frac{\alpha+i/4}{k}\frac{\coth(ks_f)}{\sinh^2(ks_f)} - \frac{s_f(\alpha+i/4)}{\sinh^4(ks_f)}\right]\,. \end{aligned} \tag{23}$$

III. THE CRITICAL TEMPERATURE OF SFS TRILAYER

To find the temperature $T_c^{0,\pi}$ of the second–order superconducting phase transition, the equations (16), (21) should be linearized with respect to the $F_s \ll 1$

$$F_s \simeq F_{s0} = \frac{\Delta}{\omega + 1/\tau_{0,\pi}}\,. \tag{24}$$

Substituting Eq. (24) into the self-consistency equation (11) one obtains the equation for the critical temperature $T_c^{0,\pi}$:

$$\ln\left(\frac{T_c^{0,\pi}}{T_{c0}}\right) = \Psi\left(\frac{1}{2}\right) - \mathrm{Re}\left[\Psi\left(\frac{1}{2}+\Omega_{0,\pi}\right)\right], \qquad (25)$$

where Ψ is the digamma function. The depairing parameter $\Omega_{0,\pi}(T) = 1/2\pi T\tau_{0,\pi}$

$$\Omega_{0,\pi} = \frac{\varepsilon}{2}\frac{T_{c0}}{T_c^{0,\pi}}\begin{cases} k\,\tanh(ks_f), & 0-\text{phase} \\ k\,\coth(ks_f), & \pi-\text{phase}\end{cases} \qquad (26)$$

is responsible for the superconductivity destruction in the S layers due to the proximity effect. Figure 2 shows a typical dependency of the critical temperature $T_c^{0,\pi}$ on the thickness of F layer d_f, obtained from Eqs. (25),(26). The crossing of the curves $T_c^0(d_f)$ and $T_c^\pi(d_f)$ occurs at d_f^*, and for $d_f > d_f^*$ the critical temperature of the $\pi-$phase becomes higher than the critical temperature of the $0-$phase.

IV. PHASE TRANSITIONS IN SFS TRILAYERS

To obtain the Ginzburg–Landau (GL) expansion for the $0-$ and $\pi-$states near the critical temperature $T_c^{0,\pi}$ we can use the equations (16),(21). Assuming $|F_s| \ll 1$ and using F_{s0} (24) as a zero-order approximation we find the solution of Eqs. (16),(21) within the first-order perturbation theory:

$$F_s = \sin\theta_s \simeq F_{s0} - \frac{F_{s0}^3}{2}\left[1 + \frac{\varepsilon\Lambda_{0,\pi}}{\omega + 1/\tau_{0,\pi}}\right]. \qquad (27)$$

Substitution of (27) into the self–consistency equation (6) one obtains a dependence of the superconducting gap Δ on the temperature $T = T_c^{0,\pi} - \delta T$

$$-a^{0,\pi}\frac{\delta T}{T_c^{0,\pi}} + b^{0,\pi}\Delta^2 = 0\,, \qquad (28)$$

where the coefficients $a^{0,\pi}$ and $b^{0,\pi}$ are determined by the following expressions:

$$a^{0,\pi} = 1 - \mathrm{Re}\left[\Omega_{0,\pi}\Psi^{(1)}(1/2+\Omega_{0,\pi})\right], \qquad (29)$$

$$b^{0,\pi} = \frac{-1}{(4\pi T_c^{0,\pi})^2}\mathrm{Re}\left[\Psi^{(2)}(1/2+\Omega_{0,\pi}) - \frac{\varepsilon\Lambda_{0,\pi}}{6\pi T_c^{0,\pi}}\Psi^{(3)}(1/2+\Omega_{0,\pi})\right], \qquad (30)$$

where $\Psi^{(n)}(z) = d^n\Psi(z)/dz^n$. Naturally all parameters $a^{0,\pi}, b^{0,\pi}$ and $T_c^{0,\pi}$ are different for 0 or π states. Figure 3 shows a typical dependence of the coefficients of the Ginzburg–Landau expansion $a^{0,\pi}$, $b^{0,\pi}$ on the thickness of F layer d_f, obtained from Eqs. (29),(30).

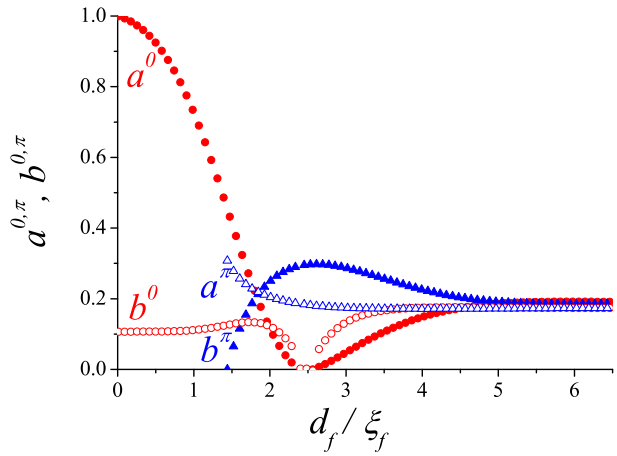

FIG. 3: (Color online) The typical dependence of the coefficients of the Ginzburg–Landau expansion $a^{0,\pi}$, $b^{0,\pi}$ on the thickness of F layer d_f: a^0 (b^0) – closed (open) red circles; a^π (b^π) – closed (open) blue triangles. Here we choose the parameters for Fig. 2.

The equilibrium value of superconducting gap

$$\Delta_{0,\pi}^2 = \frac{a^{0,\pi}}{b^{0,\pi}}\frac{T_c^{0,\pi} - T}{T_c^{0,\pi}} \qquad (31)$$

corresponds to the extremum of the standard Ginzburg-Landau functional

$$F_{GL}^{0,\pi}(T) = E_0\left[a^{0,\pi}\frac{T - T_c^{0,\pi}}{T_c^{0,\pi}}\Delta^2 + \frac{b^{0,\pi}}{2}\Delta^4\right], \qquad (32)$$

where the characteristic energy $E_0 = N(0)S_J d\,T_{c0}^2$ is determined by the total electron density of states $N(0)$, the cross section area S_J of the junction, the total thickness of the trilayer $d = 2d_s + d_f$ and the critical temperature T_{c0}. The value of the superconducting order parameter Δ, the temperatures T, $T_c^{0,\pi}$ are assumed to be measured in the units of T_{c0}. The functional (32) provides us the complete description of the ground states of SFS junction near the critical temperature $T_c^{0,\pi}$. The equilibrium energy of the system $E^{0,\pi}(T) = F_{GL}^{0,\pi}(\Delta_{0,\pi})$ is

$$E^{0,\pi}(T) = -E_0\frac{\left[a^{0,\pi}\left(T_c^{0,\pi} - T\right)/T_c^{0,\pi}\right]^2}{2b^{0,\pi}}. \qquad (33)$$

The condition of the first order transition between 0 and π states is $F^0(\Delta_0) = F^\pi(\Delta_\pi)$, and then the temperature of this transition T_0 is determined by:

$$\frac{T_c^0 - T_0}{T_c^\pi - T_0} = \frac{a^\pi}{a^0}\sqrt{\frac{b^0}{b^\pi}}. \qquad (34)$$

At $d_f \lesssim d_f^*$, first the transition from normal state to the superconducting 0 state occurs, but further decrease of the temperature provokes the transition from 0 to π

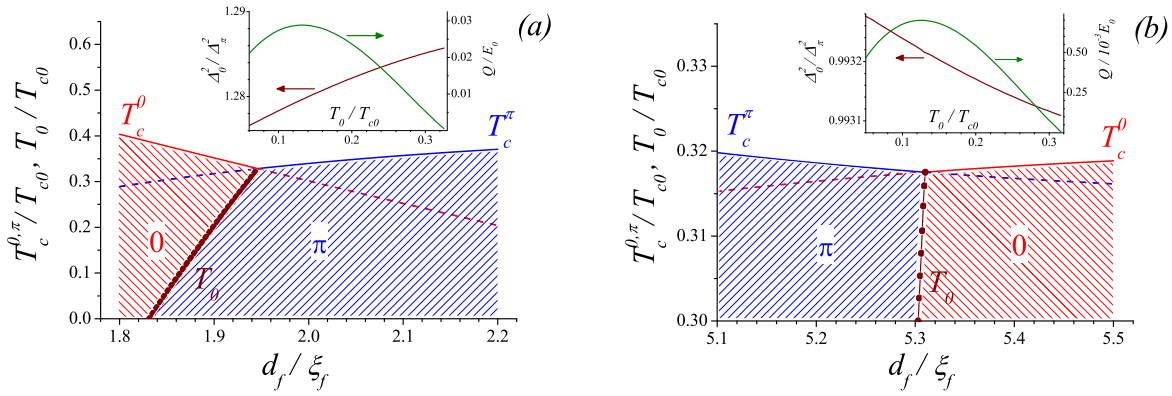

FIG. 4: The $(T,\, d_f)$ phase diagram for the SFS trilayers in the vicinity of crossing ($d_f \approx d_f^*$) of the curves $T_c^0(d_f)$ and $T_c^\pi(d_f)$: (a) $d_f^* \simeq 1.95\xi_f$; (b) $d_f^* \simeq 5.31\xi_f$. At temperature T_0 the first-order transition between the $0-$ and $\pi-$ states takes place. Here we choose the parameters for Fig. 2.The insert gives the dependence of the value of the superconducting gap jump $\Delta_0^2(T_0)/\Delta_\pi^2(T_0)$ (37) and the latent heat Q (36) on the transition temperature T_0.

state. The correspondent $(T,\, d_f)$ phase diagram for the SFS trilayers is shown in Fig. 4.

The $0-\pi$ transition is accompanied by a discontinuity in the entropy $S^{0,\pi}(T) = -\left[\partial E^{0,\pi}(T)/\partial T\right]$ at the temperature T_0:

$$\frac{S^\pi(T_0)}{S^0(T_0)} = \frac{T_c^0 - T_0}{T_c^\pi - T_0}\,. \tag{35}$$

Then the latent heat at the first order $0-\pi$ transition is

$$Q = \pm T_0\left[S^\pi(T_0) - S^0(T_0)\right] > 0 \tag{36}$$

for $0 \rightleftarrows \pi$ transitions, respectively. Simultaneously, at the transition temperature T_0 the superconducting order parameter jumps from Δ_0 to Δ_π or vice versa. The ratio of values Δ_0 and Δ_π is given by the expression:

$$\Delta_\pi^2(T_0) = \frac{a^\pi}{b^\pi}\left(\frac{T_c^\pi - T_0}{T_c^0}\right) = \Delta_0^2(T_0)\sqrt{\frac{b^0}{b^\pi}}\,. \tag{37}$$

The inserts in Fig. 4 show the dependence of the ratio $\Delta_0^2(T_0)/\Delta_\pi^2(T_0)$ (37) and the latent heat $Q(T_0)$ (36) on the transition temperature T_0. Certainly, the jump of the superconducting gap provokes the jump of the London penetration depth $1/\lambda \sim \Delta$. Figure 5 shows schematically the temperature dependence of the equilibrium gap $\Delta_{0,\pi}^2$ (31) and the Ginzburg–Landau energy $E^{0,\pi}(T)$ (33) in the vicinity of the $0 \to \pi$ transition for the case $T_c^\pi < T_c^0$ and $b^\pi > b^0$. We readily see that the superconducting order parameter decrease results in the positive jump of the London penetration depth. At all reasonable parameters h, τ_s^{-1}, ξ_f we obtain namely this scenario, which is indeed realized on the experiment[23].

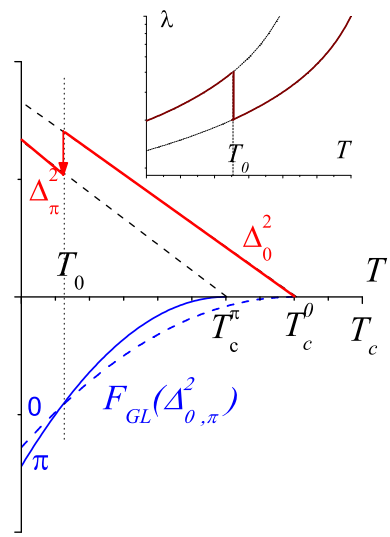

FIG. 5: (Color online) Schematic dependence of the gap $\Delta_{0,\pi}^2$ (31) and the Ginzburg–Landau energy $F_{GL}(\Delta_{0,\pi})$ (33) on the temperature T: $T_c^\pi < T_c^0$ and $(a^2/b)^\pi > (a^2/b)^0$. The inset gives the schematic temperature dependence of the penetration depth $\lambda \sim 1/\Delta_{0,\pi}$.

V. THE CURRENT–PHASE RELATION

Let us now discuss the peculiarities of the Josephson effect in the SFS trilayers at the first–order transition from 0 to π state. The theory describing how the 0 state is transformed into the π state for diffusive SFS junctions was developed in Ref. 11 for rigid boundary conditions[5] at SF interface. It was shown that the critical current does not vanish at the transition and is determined by the second–harmonic term in the current–phase relation

$$I(\varphi) = I_1 \sin\varphi + I_2 \sin 2\varphi\,, \tag{38}$$

which corresponds to the following phase–dependent contribution to the free energy of the junction:

$$E_J(\varphi) = \frac{\Phi_0}{2\pi c}\left[-I_1\left(1-\cos\varphi\right) - \frac{I_2}{2}\left(1-\cos 2\varphi\right)\right], \tag{39}$$

where $\Phi_0 = \pi\hbar c/e$ is the flux quantum. Since the amplitudes of the harmonics I_1 and I_2 depend strongly on the superconducting order parameter Δ in S layers (see Appendix A for details), the current–phase relation of the junction $I(\varphi)$ seems to be very sensitive both to a suppression of superconductivity in thin S layers and to jumps of Δ during the transitions between 0 and π states of the SFS trilayers.

To consider the Josephson coupling in the SFS trilayers in the presence of first–order $0-\pi$ transition, the Ginzburg–Landau functional (32) should be generalized for arbitrary phase difference φ between the superconducting order parameters in the S layers. For two coupled S layers with the order parameter $\Delta_{1,2} = \Delta\exp(\pm i\varphi/2)$ the Ginzburg–Landau expansion F^{φ}_{GL} of the free energy includes the mixing quadratic term $\Delta_1\Delta_2^* + \Delta_1^*\Delta_2 \sim \Delta^2\cos\varphi$. The mixing terms of fourth order have the form $(|\Delta_1|^2 + |\Delta_2|^2)(\Delta_1\Delta_2^* + \Delta_1^*\Delta_2) \sim \Delta^4\cos\varphi$ and $\Delta_1^2(\Delta_2^*)^2 + (\Delta_1^*)^2\Delta_2^2 \sim \Delta^4\cos 2\varphi$, and provide both φ and 2φ periodicity of the function F^{φ}_{GL}. Therefore, in general, the Ginzburg–Landau expansion can be written as

$$F^{\varphi}_{GL}(T,\varphi)/E_0 = -a_\varphi(T)\Delta^2 + \frac{b_\varphi}{2}\Delta^4\,, \tag{40}$$

which incorporates the phase–dependent contribution via the coefficients

$$a_\varphi(T) = \gamma_1\cos\varphi + \frac{T_c - T}{T_c^*}(1+\gamma_2\cos\varphi)\,,$$
$$b_\varphi = \beta_0 + \beta_1\cos\varphi + \beta_2\cos(2\varphi)\,.$$

For convenience the superconducting order parameter Δ, temperatures T, T_c, T_c^* and the exchange field h are measured in units of T_{c0}. To find the parameters T_c, T_c^*, $\gamma_{1,2}$, $\beta_{0,1,2}$ we take into account that the generalized functional (40) reduces to the expression (32) if $\varphi = 0$, π:

$$F^{\varphi}_{GL}(T,0) = F^0_{GL}(T)\,, \quad F^{\varphi}_{GL}(T,\pi) = F^{\pi}_{GL}(T)\,. \tag{41}$$

As a result for $T \le \min(T_c^0, T_c^\pi)$ the coefficients T_c, T_c^*, $\gamma_{1,2}$, $\beta_{0,1,2}$ can be expressed as

$$T_c^* = \frac{2}{a^0/T_c^0 + a^\pi/T_c^\pi}\,, \quad T_c = \frac{T_c^*}{2}\left(a^0 + a^\pi\right)\,,$$
$$\gamma_1 = \frac{a^0 - a^\pi}{2} - \frac{T_c}{T_c^*}\gamma_2\,, \quad \gamma_2 = \frac{T_c^*}{2}\left(\frac{a^0}{T_c^0} - \frac{a^\pi}{T_c^\pi}\right)\,,$$
$$\beta_1 = \left(b^0 - b^\pi\right)/2\,, \quad \beta_0 + \beta_2 = \left(b^0 + b^\pi\right)/2\,.$$

The coefficient β_2 in expansion (40) can be determined from the current-phase relation for rigid boundary conditions (see Appendix A):

$$\beta_2 = -\frac{\pi}{192}\frac{h}{T_c^3}\frac{\xi_f}{d}\,\mathrm{Im}\left\{\frac{1}{k\sinh^2\delta}\times \left[\frac{\delta}{2} - i\frac{\alpha + i/4}{\sinh\delta}\left(\cosh\delta - \frac{\delta}{\sinh\delta}\right)\right]\right\}\,, \tag{42}$$

where $\delta = 2s_f k = d_f/\xi_{f1} + i d_f/\xi_{f2}$. Substitution of the equilibrium value of the order parameter

$$\Delta^2_\varphi(T) = a_\varphi(T)/b_\varphi\,. \tag{43}$$

into the expression (40) provides the temperature dependence of the free energy $E(T,\,\varphi)$ of the SFS trilayers for an arbitrary phase difference φ

$$E(T,\,\varphi)/E_0 = -a^2_\varphi(T)/2b_\varphi\,. \tag{44}$$

which results in the following current–phase relation $I(\varphi) = (2e/\hbar)\,\partial E/\partial\varphi$:

$$I(T,\,\varphi) = I_0\,\sin\varphi\,\frac{a_\varphi(T)}{b_\varphi}\times \left[\gamma_1 + \gamma_2\frac{T_c - T}{T_c^*} - \frac{a_\varphi(T)}{2b_\varphi}(\beta_1 + 4\beta_2\cos\varphi)\right]\,, \tag{45}$$

where $I_0 = 2\pi c\,E_0/\Phi_0$. The current–phase relation $I(\varphi)$ (45) is shown in Fig. 6a for fixed thickness of the barrier $d_f = 1.94\xi_f$ in the vicinity of the $0-\pi$ transition and several values of the temperature T. For chosen parameters of SFS trilayers (see Fig. 3) the first 0-π transition takes place at $d_f \lesssim 1.95\xi_f$. Corresponding value of the coefficient $\beta_2 \simeq -0.013$ gives a rather large value of the second–harmonic term ($|I_2| \sim |I_1|$), which dominates near the transition. Due to the strong contribution of higher harmonics the current–phase relation is rather anharmonic, and the maximal value of $|I(T,\,\varphi)|$ occurs at $\varphi \ne \pi/2$. Figure 6b shows the temperature dependence of the critical current $I_c(T) = \max_\varphi|I(T,\,\varphi)|$ near the temperature T_0. The characteristic multimode anharmonicity of the current-phase relation results in the disappearance of the typical nonmonotonic temperature dependence of the critical current in a vicinity of the $0-\pi$ transition. We have obtained the positive amplitude of second harmonic $I_2 > 0$, which means that it occurs discontinuously by a jump between 0 and π phase states at the transition point T_I, where the critical current $I_c(T)$ formally changes it sign. The shift of the temperature T_I with respect to T_0 depends on the higher harmonics contribution.

For comparison figure 7 shows the current–phase relations $I(\varphi)$ (45) and the temperature dependence of the critical current $I_c(T)$ in a vicinity of the second $\pi-0$ transition (see Fig. 3) which takes place at $d_f \lesssim 5.31\xi_f$. Corresponding value of the coefficient $\beta_2 \simeq -0.18\,10^{-4}$ gives a small value of the second–harmonic term ($|I_2| \ll |I_1|$), and the the current–phase relation is quite harmonic except the case $T \approx T_0 \simeq T_I$. The SFS junction reveals the typical nonmonotonic behavior of the critical current I_c

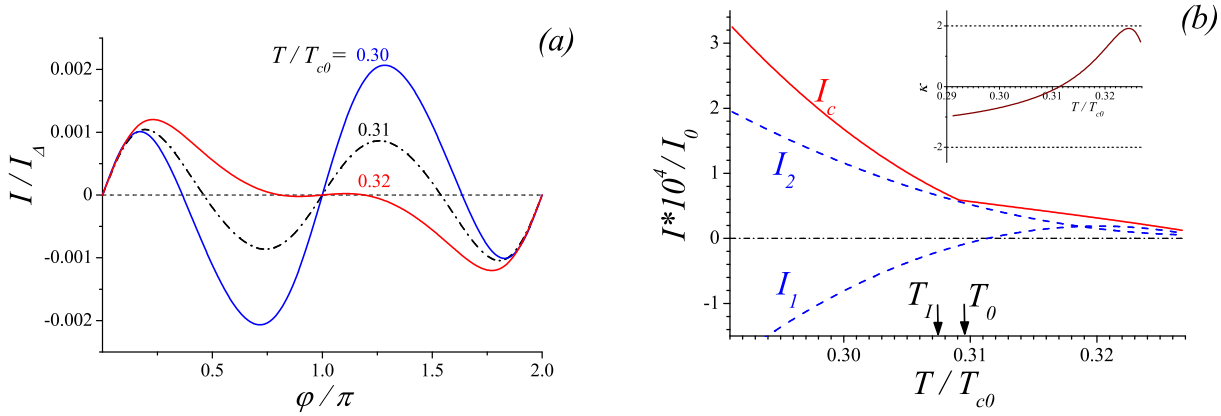

FIG. 6: (Color online) (a) Current–phase relation of SFS junction $I(\varphi)$ (45) for several values of the temperature $T/T_{c0} =$ 0.30, 0.31, 0.32 in the vicinity of the $0-\pi$ transition for $d_f = 1.94\xi_f$ ($T_c^0/T_{c0} \simeq 0.332$, $T_c^\pi/T_{c0} \simeq 0.327$, $T_c/T_{c0} \simeq 0.329$). The case $T = T_0 \simeq 0.311\,T_{c0}$ is shown by the dash-dotted curve ($I_\Delta = I_0\,\Delta_0^2(T)$, where $\Delta_0(T) = 1.764\,T_c\,\tanh(1.74\sqrt{T_c/T-1}$ is BCS superconducting gap for the temperature T). (b) Dependence of the critical current $I_c(T) = \max|I(T,\,\varphi)|$ (red solid line) and the amplitudes of harmonics I_1, I_2 (blue dashed lines) on temperature T. The insert gives the temperature dependence of a relative amplitude of the first harmonic $\kappa = I_1/I_2$. Here we choose the parameters of Fig. 3: $d_s = 2\xi_s$; $\sigma_f/\sigma_s = 0.12$; $\xi_s/\xi_f = 3$ ($d_f^* \simeq 1.946\xi_f$, $\varepsilon = 0.18$) and $h = 10T_{c0}$, $h\tau_s = 7$.

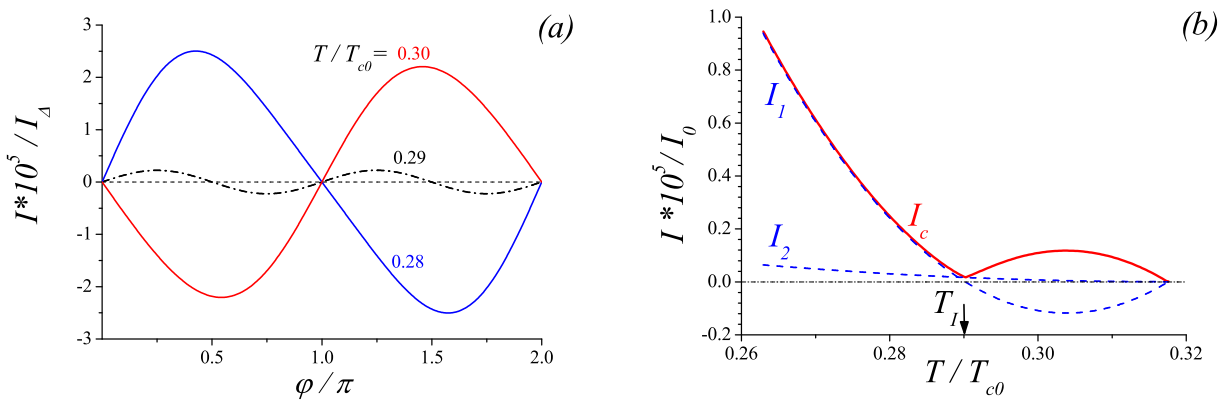

FIG. 7: (Color online) (a) Current–phase relation of SFS junction $I(\varphi)$ (45) for several values of the temperature $T/T_{c0} =$ 0.28, 0.29, 0.30 in the vicinity of the second $0-\pi$ transition for $d_f = 5.3\xi_f$ ($T_c^0/T_{c0} \simeq 0.317$, $T_c^\pi/T_{c0} \simeq 0.318$, $T_c/T_{c0} \simeq 0.3175$). The case $T = T_0 \simeq 0.29\,T_{c0}$ is shown by the dash-dotted curve ($I_\Delta = I_0\,\Delta_0^2(T)$, where $\Delta_0(T) = 1.764\,T_c\,\tanh(1.74\sqrt{T_c/T-1}$ is BCS superconducting gap for the temperature T). (b) Dependence of the critical current $I_c(T) = \max|I(T,\,\varphi)|$ (red solid line) and the amplitudes of harmonics I_1, I_2 (blue dashed lines) on temperature T. Here we choose the parameters of Fig. 3: $d_s = 2\xi_s$; $\sigma_f/\sigma_s = 0.12$; $\xi_s/\xi_f = 3$ ($d_f^* \simeq 5.31\xi_f$, $\varepsilon = 0.18$) and $h = 10T_{c0}$, $h\tau_s = 7$.

as a function of the temperature T, and the position of the cusp T_I naturally coincides with the temperature T_0.

If we restrict our consideration to the two harmonics approach (38), one will be able to determine the amplitude of both harmonics I_1 and I_2 via critical current I_c measurements, as it has been proposed in Ref. 41. Since a relative amplitude of the first harmonic $\kappa = I_1/I_2$ is small ($|\kappa| < 2$) in the vicinity of $0-\pi$ transition (see the insert in Fig. 6b), the system has two stable states $\varphi = 0$ and $\varphi = \pi$ at $I = 0$. To "depin" the Josephson phase from the low energy 0 (π) state or from the high energy π (0) for $I_1 > 0$ ($I_1 < 0$), respectively, the critical current

$$I_{c\pm}(\kappa) = \frac{I_2}{32}\left(\sqrt{\kappa^2+32} \pm 3|\kappa|\right)^{3/2} \times \left(\sqrt{\kappa^2+32} \mp |\kappa|\right)^{1/2} \qquad (46)$$

should be applied[41]. During switching from the voltage state to the zero resistance state at $T \approx T_0$, the phase may stick in the high energy state with a probability close

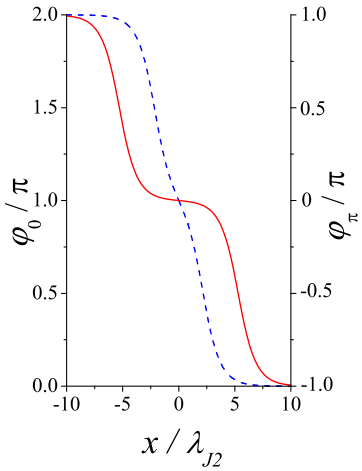

FIG. 8: (Color online) The shape of the Josephson vortex $\varphi_{0,\pi}(x)$ (48),(49) for two values of the relative amplitude of the first harmonic $\kappa = I_1/I_2$ in the vicinity of the $0-\pi$ transition: blue dashed line – $|\kappa| = 0.05$; red solid line – $|\kappa| = 10^{-4}$.

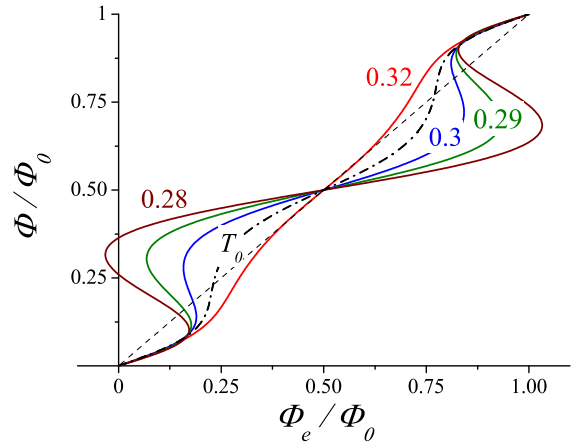

FIG. 9: (Color online) Magnetic flux through the single–junction loop Φ as a function of the external flux Φ_e for the normalized inductance $L_I = 250$ and several values of the temperature $T/T_{c0} = 0.28, 0.29, 0.3, 0.31, 0.32$ in the vicinity of the $0-\pi$ transition for $d_f = 1.94\xi_f$. The case for $T = T_0 \simeq 0.31\,T_{c0}$ is shown by the dash-dotted curve. Here we choose the parameters of Fig. 6 and $L_I = 500$.

to 50%, and the smaller current I_{c-} becomes measurable in the SFS structure with a low damping[42]. Then for $|\kappa| \ll 1$ near $0-\pi$ transition the amplitude of the harmonics $I_{1,2}$ is determined by the relations

$$|I_1| \simeq (I_{c+} - I_{c-})/\sqrt{2}\,, \qquad I_2 \simeq (I_{c+} + I_{c-})/2\,. \quad (47)$$

The structure of the Josephson vortex which may exist in a long Josephson junction with a large second harmonic ($|\kappa| < 2$) is rather peculiar and is described by the following expressions[11]

$$\varphi_0 = \begin{cases} 2\pi - \varphi_\kappa(x/\lambda_{J2}), & x < 0 \\ \varphi_\kappa(x/\lambda_{J2}), & x > 0 \end{cases}, \quad \varphi_\pi = \varphi_0 - \pi \quad (48)$$

for $I_1 > 0$ ($I_1 < 0$), respectively, where

$$\varphi_\kappa(\chi) = \arccos\left(1 - \frac{2(1+|\kappa|)}{1+|\kappa|\cosh^2(\chi\sqrt{1+|\kappa|})}\right) \quad (49)$$

and the Josephson length $\lambda_{J2}^{-2} = c\Phi_0 S/8\pi^2 I_2 t$ depends on the current density I_2/S and the effective junction thickness t. The change of the form of the Josephson vortex in the vicinity of the $0-\pi$ transition is shown in Fig. 8.

VI. SINGLE–JUNCTION LOOP

Let us consider a small superconducting loop with an inductance L interrupted by the single SFS junction (rf SQUID). We assume that the junction is described by the current–phase relation (45) and choose the parameters in a narrow region near the $0-\pi$ transition. The ground state of the circuit is determined by minimizing the SQUID free energy

$$W(T,\,\phi) = E(T,\,\phi) + \frac{\Phi_0^2}{8\pi^2 L}\,(\phi - \phi_e)^2\,, \qquad (50)$$

where $\phi_e = 2\pi\Phi_e/\Phi_0$ – is the normalized magnetic flux of an external field through the loop[43]. The total magnetic flux through the loop $\Phi = (\phi/2\pi)\,\Phi_0$ is related to the external flux Φ_e as

$$\phi_e = \phi + L_I I(T,\,\phi)/I_0\,, \qquad (51)$$

with the normalized inductance $L_I = 2\pi L\,I_0/c\Phi_0$. Figure 9 shows the dependence $\Phi(\Phi_e)$ determined by Eqs. (50) and (51) for several values of the temperature T near the transition temperature T_0. New features of $\Phi(\Phi_e)$ dependence appear for T close to T_0 due to the coexistence of stable and metastable 0 and π states. Strong anharmonicity of the current-phase relation for $T \approx T_0 \sim T_c$ results in the coexistence of integer and half–integer fluxoid configuration in SQUID's and generation of two flux jumps per one external flux quantum. This behavior is similar to the magnetic-flux penetration in superconducting loop with a clean SFS junction at low temperatures $T \ll T_c$[44].

VII. SUMMARY

We have studied the thermodynamics of diffusive SFS trilayer with relatively thin S layers through self-consistent solutions of nonlinear Usadel equations, in

the dirty limit. Our results may be viewed as generalization of those obtained in Ref. 11, when the superconducting electrodes are the rather thin, and the critical temperature T_c is affected by the ferromagnetic layer. We have shown that as the temperature T is varied a given SFS junction can flip from the 0 state to the π state. The resulting phase transition is first–order, in agreement with the experiments[8,9,33] and is responsible for a jump of the amplitude of the superconducting order parameter Δ, providing the anomalous temperature behavior of the effective penetration depth $\lambda(T)$ observed in Ref. 23. Taking the typical parameters $N(0) \sim 10^{33}\,\mathrm{erg}^{-1}\,\mathrm{sm}^{-3}$, $T_{c0} \sim 10\,\mathrm{K} \approx 10^{-15}\,\mathrm{erg}$ we may estimate for the $\mathrm{Nb/Pd_{0.84}Ni_{0.16}/Nb}$ trilayer[23,45] ($d \simeq 30\,\mathrm{nm}$, $S \sim 0.1\,\mathrm{sm}^2$) the latent heat $Q \sim 0.01 E_0 \approx 0.3\,\mathrm{pJ}$. Such picojoule latent heat can be readily observed via standard ac calorimetry techniques used to measure specific and latent heats in films: attojoule level results have been reported[46]. We see, therefore, that the latent heat Q (36) associated with the first–order $0-\pi$ transition in SFS trilayer is quite observable.

We have proposed the general form of the Ginzburg-Landau functional to describe SFS trilayer for arbitrary phase difference φ between the order parameters in the superconducting layers. Calculation of the current–phase relation $I(\varphi)$ shows that the ground state of the SFS junction is 0 or π, and the transition between the 0 and π states appears discontinuous. The current–phase relation strongly deviates from the simple sinusoidal one due to strong dependence of superconductivity in thin S layers on the structure of the pair wave function in the ferromagnetic, even at temperatures T near the critical value T_c. The characteristic anharmonicity of the current–phase relation results in disappearance of the typical non-monotonic temperature dependence of the critical current in a vicinity of the $0-\pi$ transition. Certainly, the anharmonicity of the current–phase relation becomes less pronounced for thick ferromagnetic layer, if $d_f \gg \xi_f$.

We show that coexisting stable and metastable 0 and π states appear in the vicinity of $0-\pi$ transition. As a consequence, integer and half-integer fluxoid configurations exist in the superconducting loop interrupted by the junction. The coexistence of 0 and π states is manifested as two jumps in the dependence of enclosed magnetic flux in the loop per period[44].

acknowledgmentsThe authors thank E. Silva, V.V. Ryazanov and A.S. Mel'nikov for stimulating discussions. This work was supported, in part, by the NanoSC-COST (Belgium), Action MP1201 , by ANR (France), Grant MASH and by the Russian Foundation for Basic Research. One of the authors (A.V.S.) is supported by the Russian Scientific Foundation Grant No 15-12-10020.

Appendix A: The current–phase relation for rigid boundary conditions

Let us briefly remind of approach developed in Ref. 11 to study the problem of the second–harmonic contribution to the the current–phase relation (38) in the framework of the rigid boundary conditions. The general expression for the supercurrent through a SFS junction is given by

$$I_s = -I_0(T/T_c)\sum_{\omega>0}\mathrm{Im}\left\{F^+F_s'\right\}\,, \tag{A1}$$

where $F^+(s,h) = F^*(s,-h)$, $I_0 = 4\pi T_c e S N(0) D_f/\xi_f$, S is the area of cross section of the junction, and $N(0)$ is the electron density of states per one-spin projection.

The solution of Eqns. (1), (7) for $T \lesssim T_c$ and an arbitrary phase difference φ is[11]

$$\begin{aligned} F(s) &\simeq a\,\cosh(qs) + b\,\sinh(ps) \\ &+\frac{b^2-a^2}{8k^2}\left(\alpha+\frac{3i}{4}\right)\left[a\cosh(3ks)+b\sinh(3ks)\right]\,, \end{aligned} \tag{A2}$$

where the complex wave vectors q and p are different from the wave vector k due to nonlinear effects. Just below T_c the nonlinear corrections seem to be small and wave vectors q and p are determined by the relations

$$q \simeq k - \frac{a^2}{2k}\left(\alpha+i/4\right) + \frac{b^2}{2k}\left(\alpha+5i/4\right)\,, \tag{A3}$$

$$p \simeq k - \frac{a^2}{2k}\left(\alpha+5i/4\right) - \frac{b^2}{2k}\left(\alpha+i/4\right)\,. \tag{A4}$$

The function $F^+(s)$ is obtained by replacing b in expression (A2) by $-b$. Neglecting the influence of the F layer on the S layers, one can find the amplitudes a and b from the rigid boundary conditions assuming that the anomalous Green's function for the Matsubara frequency ω at the boundary of left (right) S layer coincides with the bulk one: $F_s(\omega,\mp s_f) = \Delta_B\,\mathrm{e}^{\pm i\varphi/2}/\Omega$, where Δ_B is the temperature dependent BCS order parameter and $\Omega = \sqrt{\omega^2+|\Delta_B|^2}$. Using the boundary conditions

$$F(\pm s_f) = F_s(\omega,\pm s_f) \tag{A5}$$

we get from (A2),(A4),(A4) the following expressions for the amplitudes a and b:

$$a = a_0 + a_1\,, \quad b = b_0 + b_1\,, \tag{A6}$$

$$a_0 = \frac{\Delta_B}{\Omega}\frac{\cos(\varphi/2)}{\cosh(\delta/2)}\,, \quad b_0 = -i\frac{\Delta_B}{\Omega}\frac{\sin(\varphi/2)}{\sinh(\delta/2)}\,, \tag{A7}$$

$$a_1 = \frac{-a_0}{8k^2\cosh(\delta/2)}\left[\left(b_0^2-a_0^2\right)(\alpha+3i/4)\left(\cosh(3\delta/2)+2\delta\sinh(\delta/2)\right)+i\delta\left(a_0^2+b_0^2\right)\sinh(\delta/2)\right], \tag{A8}$$

$$b_1 = \frac{-b_0}{8k^2\sinh(\delta/2)}\left[\left(b_0^2-a_0^2\right)(\alpha+3i/4)\left(\sinh(3\delta/2)-2\delta\cosh(\delta/2)\right)+i\delta\left(a_0^2+b_0^2\right)\cosh(\delta/2)\right], \tag{A9}$$

where $\delta=\delta_1+i\delta_2$ and $\delta_{1,2}=d_f/\xi_{f1,f2}$. Substitution of the solutions (A2), (A6), (A7), (A8) to the general expression for the total Josephson current (A1) results in the following expression for the amplitude of the first harmonic $I1$ of the current–phase relation (38):

$$I_1 = \frac{I_0}{8}\left(\frac{\Delta_B}{T_c}\right)^2\left[\mathrm{Im}\left\{\frac{ik}{\sinh\delta}\right\}-\frac{1}{12}\left(\frac{\Delta_B}{T_c}\right)^2\mathrm{Im}\left\{\frac{i(\alpha+5i/4)}{k\sinh\delta}-\frac{i(\alpha+i/4)\left(1-\delta/\tanh\delta\right)}{k\sinh^3\delta}\right\}\right]. \tag{A10}$$

The second harmonic amplitude is much smaller and described by the following expression

$$I_2 = \frac{I_0}{192}\left(\frac{\Delta_B}{T_c}\right)^4\mathrm{Im}\left\{\frac{1}{k\sinh^2\delta}\left[\frac{\delta}{2}-i\frac{\alpha+i/4}{\sinh\delta}\left(\cosh\delta-\frac{\delta}{\sinh\delta}\right)\right]\right\}. \tag{A11}$$

Actually the Josephson energy (39) is the phase dependent contribution to the Ginzburg-Landau energy (40).

$$\frac{1}{2}E_0\left(\frac{\Delta}{T_{c0}}\right)^4\beta_2\cos 2\varphi \equiv -\frac{\Phi_0 I_2}{4\pi c}\cos 2\varphi\,. \tag{A12}$$

The last equality determines the coefficient β_2 (42) in the functional (40):

$$\beta_2 = -\frac{\pi}{192}\left(\frac{T_{c0}}{T_c}\right)^3\frac{h}{T_{c0}}\frac{\xi_f}{d}\,\mathrm{Im}\left\{\frac{1}{k\sinh^2\delta}\left[\frac{\delta}{2}-i\frac{\alpha+i/4}{\sinh\delta}\left(\cosh\delta-\frac{\delta}{\sinh\delta}\right)\right]\right\}, \tag{A13}$$

[1] A. I. Buzdin, L. N. Bulaevskii, and S. V. Panyukov, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 147 (1982) [JETP Lett. **35** 178 (1982)]

[2] A. I. Buzdin and M. Yu. Kuprianov, Pis'ma Zh. Eksp. Teor. Fiz. **53**, 308 (1991) [JETP Lett. **53**, 321 (1991)]

[3] A. I. Buzdin, Rev. Mod. Phys., **77**, 935 (2005).

[4] F. S. Bergeret, A. Volkov, and K. Efetov,Rev. Mod. Phys. **77**, 1321 (2005).

[5] A. A. Golubov, M. Yu. Kupriyanov, and E. Il'ichev, Rev. Mod. Phys., **76**, 411 (2004).

[6] V. V. Ryazanov, V. A. Oboznov, A. Yu. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. **86**, 2427 (2001).

[7] T. Kontos, M. Aprili, J. Lesueur, F. Genet, B. Stephanidis, and R. Boursier, Phys. Rev. Lett. **89**, 137007 (2002).

[8] S. M. Frolov, D. J. Van Harlingen, V. A. Oboznov, V. V. Bolginov, and V. V. Ryazanov, Phys. Rev. B **70**, 144505 (2004).

[9] V. A. Oboznov, V. V. Bolginov, A. K. Feofanov, V. V. Ryazanov, and A. I. Buzdin, Phys. Rev. Lett. **96** 197003 (2006)

[10] S. M. Frolov, D. J. Van Harlingen, V. V. Bolginov, V. A. Oboznov, V. V. Ryazanov, Phys. Rev. B **74**, 020503 (2006)

[11] A. I. Buzdin, Phys. Rev. B **72**, 100501 (2005).

[12] M. Houzet, V. Vinokur and F. Pistolesi Phys. Rev. B **72**, 220506 (2005)

[13] T. R. Lemberger, I. Hetel, A. J. Hauser, and F. Y. Yang, J. Appl. Phys. **103**, 07C701 (2008).

[14] M. Houzet and J. S. Meyer, Phys. Rev. B **80** 012505 (2009).

[15] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B **64**, 134506 (2001).

[16] Y. Asano, A. A. Golubov, Y.V. Fominov, and Y. Tanaka, Phys. Rev. Lett. **107**, 087001 (2011).

[17] T. Yokoyama, Y. Tanaka, and N. Nagaosa, Phys. Rev. Lett. **106**, 246601 (2011).

[18] F.S. Bergeret, A. F. Volkov and K. B. Efetov, Phys. Rev. Lett. **86**, 4096 (2001).

[19] A. Kadigrobov, R. I. Shekhter and M. Jonson, Europhys. Lett. **54**, 394 (2001).

[20] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964).

[21] A. I. Larkin and Y. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].

[22] S. Mironov, A. Melnikov, and A. Buzdin, Phys. Rev. Lett. **109**, 237002 (2012).

[23] N. Pompeo, K. Torokhtii, C. Cirillo, A. V. Samokhvalov, E. A. Ilyina, C. Attanasio, A. I. Buzdin, and E. Silva, Phys. Rev. B **90**, 064510 (2014).

[24] A. I. Buzdin and M. Y. Kuprianov, Pisma Zh. Eksp. Teor. Fiz. **52**, 1089 (1990) [JETP Lett. **52**, 487 (1990)]

[25] Z. Radovic, M. Ledvij, L. Dobrosavljevic-Grujic, A. I.

Buzdin, and John R. Clem, Phys. Rev. B **44**, 759 (1991).
[26] I. Baladie and A. Buzdin, Phys. Rev. B **67**, 014523 (2003).
[27] S. Tollis, Phys. Rev. B **69**, 104532 (2004).
[28] J. S. Jiang, D. Davidovic, Daniel H. Reich, and C. L. Chien, Phys. Rev. Lett. **74**, 314 (1995).
[29] Th. Muhge, N. N. Garifyanov, Yu. V. Goryunov, G. G. Khaliullin, L. R. Tagirov, K. Westerholt, I. A. Garifullin, and H. Zabel, Phys. Rev. Lett. **77**, 1857 (1996).
[30] V. Shelukhin, A. Tsukernik, M. Karpovski, Y. Blum, K. B. Efetov, A. F. Volkov, T. Champel, M. Eschrig, T. Lofwander, G. Schon, and A. Palevski, Phys. Rev. B **73**, 174506 (2006).
[31] I. A. Garifullin, D. A. Tikhonov, N. N. Garifyanov, L. Lazar, Yu. V. Goryunov, S. Ya. Khlebnikov, L. R. Tagirov, K. Westerholt, and H. Zabel, Phys. Rev. B **66**, 020505 (2002).
[32] A. S. Sidorenko, V. I. Zdravkov, A. A. Prepelitsa, C. Helbig, Y. Luo, S. Gsell, M. Schreck, S. Klimm, S. Horn, L. R. Tagirov, and R. Tidecks, 2003, Ann. Phys. **12**, 37 (2003).
[33] H. Sellier, C. Baraduc, F. Lefloch, and R. Calemczuk, Phys. Rev. Lett. **92**, 257005 (2004).
[34] P. H. Barsic, Oriol T. Valls, and K.Halterman, Phys. Rev. B **73**, 144514 (2006).
[35] P. H. Barsic, Oriol T. Valls, and K.Halterman, Phys. Rev. B **75**, 104502 (2007).
[36] N. N. Bogolubov, Zh. Eksp. Teor. Fiz. **34**, 58 (1958) [Sov. Phys. JETP **7**, 41 (1958)].
[37] P. G. deGennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).
[38] K. D. Usadel, Phys. Rev. Lett. **95**, 507 (1970).
[39] M. Faure, A. I. Buzdin, A. A. Golubov, and M. Yu. Kupriyanov, Phys. Rev. B **73**, 064505 (2006).
[40] M. Yu. Kuprianov and V. F. Lukichev, Zh. Eksp. Teor. Fiz. **94**, 139 (1988) [Sov. Phys. JETP **67**, 1163 (1988)].
[41] E. Goldobin, D. Koelle, R. Kleiner, and A. Buzdin, Phys. Rev. B **76**, 224523 (2007).
[42] H. Sickinger, A. Lipman, M. Weides, R.G. Mints, H. Kohlstedt, D. Koelle, R. Kleiner, and E. Goldobin, Phys. Rev. Lett. **109**, 107002 (2012).
[43] A. Barone and G. Paterno, *Physics and application of the Josephson Effect* (Wiley, New York, 1982).
[44] Z. Radovic, L. Dobrosavljevic-Grujic, and B. Vujicic, Phys. Rev. B **63**, 214512 (2001).
[45] K. Torokhtii, C. Attanasio, C. Cirillo, E.A. Ilyina, N. Pompeo, S. Sarti, E. Silva, Physica C **479**, 140 (2012).
[46] O. Bourgeois, S. E. Skipetrov, F. Ong, and J. Chaussy, Phys. Rev. Lett. **94**, 057007 (2005).