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

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(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 of 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 $\pi$ (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 \textbf{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 $\pi$ 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 $\pi$ 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\textsuperscript{1,2} (for more references and reviews, see Refs. \textsuperscript{3,4}). 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\textsuperscript{5,6}. The $0 - \pi$ transition manifests itself in the vanishing of $I_c$ if higher–order harmonics of the current–phase relation are negligible\textsuperscript{7,8}.

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 reveal a slightly nonmonotonic dependence of the penetration depth on the F layer thickness, which was in accordance with the theoretical analysis\textsuperscript{9,10}. Anomalous Meissner effect in hybrid SF structures was the subject of several theoretical works\textsuperscript{11,12} 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\textsuperscript{13,14} generated in inhomogeneous F layer due to proximity effect and should result in the in-plane Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) instability\textsuperscript{15,16}. Unusual drop of the screening with decrease of temperature was observed recently in\textsuperscript{17} by microwave measurements of the London penetration depth $\lambda$ in Nb/Pd\textsubscript{0.84}Ni\textsubscript{0.16}/Nb trilayers.

The transition temperature of SF structures into the normal state has been examined both theoretically\textsuperscript{18–21} and experimentally\textsuperscript{22–24} (see Ref. \textsuperscript{25} for reviews). However, the study of the thermodynamic properties of the phase transition between 0 and $\pi$ 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\textsuperscript{26}. Experimental evidence of a $0 - \pi$ transition in SFS (Nb/Cu\textsubscript{x}Ni\textsubscript{1–x}/Nb) Josephson junction was obtained from the measurements of the temperature dependence of the critical current in Refs. \textsuperscript{27,28}. Note that a small modulation of the thickness of the barrier may favor the continuous $0 - \pi$ transition\textsuperscript{29}. A first-order transitions between 0 and $\pi$ states by temperature variation were demonstrated in both the clean\textsuperscript{30,31} and dirty\textsuperscript{27} limits using numerical self-consistent solutions of the microscopic Bogoliubov-de-Gennes\textsuperscript{32,33} or Usadel\textsuperscript{34} 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\textsuperscript{35,36}. The theoretical model Proposed in Ref. \textsuperscript{23} argued that the observed jump $\lambda(T)$ in Nb/Pd\textsubscript{0.84}Ni\textsubscript{0.16}/Nb trilayers is related to the first order phase transition from 0 to $\pi$ 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 $\pi$ state. The
leakage of the Cooper pairs weakens the superconductivity near the interface with a F metal due to the proximity effect. 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 $\pi$ 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 $T^{0,\pi}_c$ of the second-order superconducting phase transition to the normal state for 0 and $\pi$ order-parameter configurations. The Sec. IV is devoted to the analysis of the temperature-driven transition between 0 and $\pi$ states. We obtain the Ginzburg–Landau expansion and find the ground states of SFS junction near the critical temperatures $T^{0,\pi}_c$. In Sec. V we generalize the Ginzburg–Landau description for arbitrary phase difference $\varphi$ 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 $\pi$ 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 $\xi_s$ is the superconducting coherence length. The considered geometry of the SFS structure is presented in Fig. 1. In the previous observations of 0–$\pi$ transition in SFS junctions by temperature variation the superconducting electrodes were rather thick to overcome the pair–breaking proximity effect of F layer and then the influence of the 0–$\pi$ 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). Here we study the SFS structure with relatively thin S layers and demonstrate that the 0–$\pi$ 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.

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 $F(x)$ in the ferromagnetic layer, two different order–parameter configurations are possible inside the SFS trilayers (see Fig. II). The first one corresponds to the case that 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 $\pi$ states. As a result the equilibrium superconducting gaps $\Delta_{0,\pi}$ for 0 and $\pi$ 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 $\pi$.
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-π 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 into mind that the F interlayer is dilute ferromagnetic alloys like Cu₈Ni₈-x or Pd₈Ni₈-x we use the Usadel equations[1] which are convenient in the diffusive limit (see[2] 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. We describe it by introducing the introducing the mesoscopic superconducting loop containing the SFS junction.

The usual parametrization of the normal and anomalous Green’s functions in F layer are[3]:

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

(1)

(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). For the 0-state we should choose the even anomalous Green’s functions \( F \) while for the π-state it should be the odd one. Using the usual parametrization of the normal and anomalous Green functions \( G_\alpha = \cos \theta_\alpha \) and \( F_\alpha = \sin \theta_\alpha \), the complete nonlinear Usadel equation in superconducting layers can be written for \( \omega > 0 \) as:

\[
\frac{-D_s}{2} \frac{\partial^2 \theta_\alpha}{\tau_s} + \omega \sin \theta_\alpha = \Delta \cos \theta_\alpha,
\]  

(3)

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

\[
F_s = F, \quad \sigma_f \partial_x F_s = \sigma_f \partial_x F,
\]  

(4)

where \( \sigma_f \) and \( \sigma_s \) 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

\[
\partial_x F_s = 0.
\]  

(5)

For thin S-layers \( d_s \ll \xi_s = \sqrt{D_s/2\pi T_{c0}} \), 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 \( \Delta(x) \):

\[
\Delta(x) = \pi T \rho \sum_\omega F_s(x, \omega),
\]  

(6)

where \( \rho \) 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 \( F^+(x, \omega, h) = F^+(x, \omega, h) \) and one can replace \( F^+(x, \omega, h) \) by \( F(x, \omega, h) \) in Eqn. (4). Just below the critical temperature \( T_c \) anomalous Green’s functions are small and the condition (2) can be rewritten as

\[
G(x, \omega, h) \approx 1 - F^2(x, \omega, h)/2.
\]  

(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:

\[
\Delta(x) \approx \theta_s(x), \quad \Delta(x) \approx \Delta_0.
\]  

Therefore, we can average Eq. (4) over the thickness of the S layers, using the boundary condition (5). Finally, we obtain the following boundary condition:

\[
\frac{\partial \theta_s}{\partial s} \bigg|_{s_f} = \frac{d_s \xi_f}{\xi_f} \left( \frac{\Delta \cos \theta_s - \omega \sin \theta_s}{\pi T_{c0}} \right),
\]  

(8)

\[
\frac{\partial \theta_s}{\partial s} \bigg|_{-s_f} = \mp \frac{\partial \theta_s}{\partial s} \bigg|_{s_f},
\]  

(9)

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

Applying the method[11] we can find the approximate solution of the nonlinear Usadel equation (4) 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 π-state it should be the odd one:

\[
F(s, \omega) \approx \begin{cases} 
\frac{a \cosh(qs)}{b \sinh(qs)} & -,
\end{cases}
\]  

(10)

\[
\frac{1}{8k^2} \left( \alpha + \frac{3i}{4} \right) \begin{cases} 
a^2 \cosh(3ks) & , \quad 0 \text{- phase} \\
b^3 \sinh(3ks) & , \quad \pi \text{- phase}
\end{cases}
\]  

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

\[
q^2 = k^2 \mp a^2 (\alpha + i/4)
\]  

(11)

for 0 and π phases, respectively. If \( T_c < \tau_s^{-1} h \), we may neglect the Matsubara frequencies in the Eq. (11) assuming that \( k^2 = (k_1 + ik_2)^2 = 2(\alpha + i) \) for \( \omega > 0 \):

\[
k_1 = \sqrt{1 + a^2 + \alpha}, \quad k_2 = \sqrt{1 + a^2 - \alpha},
\]  

(12)
Then the decay characteristic length $\xi_{f1}$ and the oscillation period $\xi_{f2}$, may be written as

$$\xi_{f1} = \frac{\xi_f}{k_1}, \quad \xi_{f2} = \frac{\xi_f}{k_2}. \quad (13)$$

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$^{10}$.

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 $a$:

$$F(s) \simeq f \cosh(k s) - f^3 g_0(s) + o(f^4), \quad (14)$$

$$g_0(s) = \frac{1 - i}{8k^2} [4ks(\alpha + i/4) \sinh(k s) + (\alpha + 3i/4) \cosh(3k s)] \cdot$$

Using the first boundary condition $^{11}$ at $s = s_f$ we get the relation between the amplitude $a$ and Green’s function $F_s = \sin \theta_s$ in superconductor:

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

Substitution of the solution $^{14}$ to the relations $^{2}$ results in the following equation with respect to the amplitude $F_s$ in the $S$ layers:

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

where

$$\tau_0^{-1} = \varepsilon \pi T_{c0} k \tanh(k s_f) \quad (17)$$

is the depairing parameter of even mode and

$$\Lambda_0 = \frac{\pi T_{c0}}{2k} \left[ i \frac{\tan(k s_f)}{\cosh^2(k s_f)} - \frac{\alpha + i/4}{k} \frac{\tanh(k s_f)}{\cosh^4(k s_f)} \right]. \quad (18)$$

Here the key parameter

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

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

B. **Odd mode ($\pi$–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 $b$:

$$F(s) \simeq f \sinh(k s) + f^3 g_\pi(s) + o(f^4), \quad (19)$$

$$g_\pi(s) = \frac{1 - i}{8k^2} [4ks(\alpha + i/4) \cosh(k s) -(\alpha + 3i/4) \sinh(3k s)] \cdot$$

Using the first boundary condition $^{11}$ at $s = s_f$ we get the relation between the amplitude $b$ and Green’s function $F_s = \sin \theta_s$ in superconductor:

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

Substitution of the solution $^{19}$ to the relations $^{21}$ results in:

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

where

$$\tau_\pi^{-1} = \varepsilon \pi T_{c0} k \coth(k s_f) \quad (22)$$

is the depairing parameter of odd mode and

$$\Lambda_\pi = \frac{\pi T_{c0}}{2k} \left[ i \frac{\coth(k s_f)}{\sinh^2(k s_f)} + \frac{\alpha + i/4}{k} \frac{\coth(k s_f)}{\sinh^4(k s_f)} \right]. \quad (23)$$

III. **THE CRITICAL TEMPERATURE OF SFS TRILAYER**

To find the temperature $T_{c0}^{0,\pi}$ of the second–order superconducting phase transition, the equations $^{16}$, $^{21}$ should be linearized with respect to the $F_s \ll 1$

$$F_s \simeq F_{c0} = \frac{\Delta}{\omega + 1/\tau_{0,\pi}} \cdot (24)$$
IV. PHASE TRANSITIONS IN SFS TRILAYERS

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

\[
F_s = \sin \theta_s \simeq F_{s0} - \frac{F_{s0}^3}{2} \left[ 1 + \frac{\varepsilon \Lambda_0,\pi}{\omega + 1/\tau_0,\pi} \right].
\]  

(27)

Substitution of \( 27 \) into the self-consistency equation \( 15 \) one obtains a dependence of the superconducting gap \( \Delta \) on the temperature \( T = T_{c,0}^{0,\pi} - \delta T \)

\[
- a^{0,\pi} \frac{\delta T}{T_{c,0}^{0,\pi}} + b^{0,\pi} \Delta^2 = 0,
\]  

(28)

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

\[
a^{0,\pi} = 1 - \text{Re} \left[ \Omega_0,\pi \Psi^{(1)}(1/2 + \Omega_0,\pi) \right],
\]  

\[
b^{0,\pi} = \frac{1}{4 \pi T_{c,0}^{0,\pi}^2} \text{Re} \left[ \Psi^{(2)}(1/2 + \Omega_0,\pi) \right] - \frac{\varepsilon \Lambda_0,\pi}{6 \pi T_{c,0}^{0,\pi}} \Psi^{(3)}(1/2 + \Omega_0,\pi),
\]  

where \( \Psi^{(n)}(z) = d^n \Psi(z) / dz^n \). Naturally all parameters \( a^{0,\pi}, b^{0,\pi} \) and \( T_{c,0}^{0,\pi} \) are different for 0 or π states. Figure 2 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. \( 29,30 \).

The equilibrium value of superconducting gap

\[
\Delta_0,\pi^2 = \frac{a^{0,\pi} T_{c,0}^{0,\pi} - T}{b^{0,\pi} T_{c,0}^{0,\pi}} \Delta^2 + \frac{b^{0,\pi} \Delta^4}{2},
\]  

(31)

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

\[
E^{0,\pi}(T) = -E_0 \frac{a^{0,\pi} T_{c,0}^{0,\pi}(T - T_{c,0}^{0,\pi})^2}{2 b^{0,\pi}},
\]  

(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:

\[
\frac{T_0^0 - T_0}{T_{c,0}^0 - T_0} = \frac{a^{0}}{a^{0}} \sqrt{\frac{b^{0}}{b^{0}}}
\]  

(34)

At \( d_f \ll d_f^* \), 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.
The 0–π transition is accompanied by a discontinuity in the entropy $S^{0,\pi}(T) = -[\partial E^{0,\pi}(T)/\partial T]$ at the temperature $T_0$:

$$\frac{S^{\pi}(T_0)}{S^{0}(T_0)} = \frac{T^0_0 - T_0}{T^\pi_0 - T_0}. \quad (35)$$

Then the latent heat at the first order 0–π transition is

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

for $0 \rightarrow \pi$ transitions, respectively. Simultaneously, at the transition temperature $T_0$ the superconducting order parameter jumps from $\Delta_0$ to $\Delta_\pi$ or vice versa. The ratio of values $\Delta_0$ and $\Delta_\pi$ is given by the expression:

$$\Delta^2_0(T_0) = \frac{a^\pi}{b^\pi} \left( \frac{T_0^\pi - T_0}{T_0^\pi} \right) = \Delta^2_0(T_0) \sqrt{\frac{b^\pi}{b^\pi}}. \quad (37)$$

The inserts in Fig. 4 show the dependence of the ratio $\Delta^2_0(T_0)/\Delta^2_0(T_0)$ and the latent heat $Q(T_0)$ 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 $\Delta^2_{0,\pi}$ and the Ginzburg–Landau energy $E^{0,\pi}(T)$ in the vicinity of the 0 – π transition for the case $T^\pi_0 < T^0_0$ 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 $h$, $\tau^{-1}_s$, $\xi_f$ we obtain namely this scenario, which is indeed realized on the experiment. 33.

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 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

$$I(\varphi) = I_1 \sin \varphi + I_2 \sin 2\varphi, \quad (38)$$
which corresponds to the following phase–dependence contribution to the free energy of the junction:

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

(39)

where $\Phi_0 = \pi hc/e$ is the flux quantum. Since the amplitudes of the harmonics $I_1$ and $I_2$ depend strongly on the superconducting order parameter $\Delta$ 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 $\Delta$ during the transitions between 0 and $\pi$ 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 $\varphi$ 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 $F^c_{\text{GL}}$ 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 $(|\Delta_1|^2 + |\Delta_2|^2)(\Delta_1 \Delta_{2}^* + \Delta_{1}^* \Delta_{2}) \sim \Delta^2 \cos \varphi$ and $(|\Delta_1|^2 + |\Delta_2|^2)^2 \sim \Delta^4 \cos 2\varphi$, and provide both $\varphi$ and $2\varphi$ periodicity of the function $F^c_{\text{GL}}$. Therefore, in general, the Ginzburg–Landau expansion can be written as

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

(40)

which incorporates the phase–dependence contribution via the coefficients

$$a_\varphi(T) = \gamma_1 \cos \varphi + \frac{T_c - T}{T_c^*} (1 + \gamma_2 \cos \varphi),$$

$$b_\varphi = \beta_0 + \gamma_1 \cos \varphi + \beta_2 \cos(2\varphi).$$

For convenience the superconducting order parameter $\Delta$, temperatures $T$, $T_c$, $T_c^*$ and the exchange field $h$ are measured in units of $T_c 0$. 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 $\varphi = 0, \pi$:

$$F^c_{\text{GL}}(T, 0) = F^0_{\text{GL}}(T), \quad F^c_{\text{GL}}(T, \pi) = F^c_{\text{GL}}(T).$$

(41)

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

$$T_c = \frac{2}{a_0 + a_0^* + a^* / T_c^*}, \quad T_c^* = \frac{T_c^*}{2} \left( a_0 + a_0^* \right),$$

$$\gamma_1 = \frac{a_0 - a_0^*}{2} - \frac{T_c}{T_c^*} \gamma_2, \quad \gamma_2 = \frac{T_c^*}{2} \left( \frac{a_0}{T_c^*} - \frac{a_0^*}{T_c^*} \right),$$

$$\beta_1 = \frac{b_0 - b_0^*}{2}, \quad \beta_0 + \beta_2 = \frac{b_0 + b_0^*}{2}.$$

The coefficient $\beta_2$ in expansion (40) can be determined from the current–phase relation for rigid boundary conditions (see Appendix A):

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

(42)

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

$$\Delta^2(T) = a_\varphi(T)/b_\varphi,$$

(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 $\varphi$

$$E(T, \varphi)/E_0 = -a_\varphi^2(T)/2b_\varphi,$$

(44)

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

$$I(T, \varphi) = I_0 \sin \varphi \left( \frac{a_\varphi(T)}{b_\varphi} \times \left[ \gamma_1 + \frac{T_c - T}{T_c^*} - \frac{a_\varphi(T)}{2b_\varphi} (\beta_1 + 3\beta_2 \cos \varphi) \right] \right),$$

(45)

where $I_0 = 2\pi e E_0 / \Phi_0$. The current–phase relation $I(\varphi)$ is shown in Fig. 6a, for fixed thickness of the barrier $d_f = 1.94\xi_{f1}$ in the vicinity of the 0–$\pi$ transition and several values of the temperature $T$. For chosen parameters of SFS trilayers (see Fig. B) the first 0–$\pi$ transition takes place at $d_f \approx 0.95\xi_f$. Corresponding value of the coefficient $\beta_2 \approx -0.013$ 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 $|I(\varphi)|$ occurs at $\varphi \neq \pi/2$. Figure 6a shows the temperature dependence of the critical current $I_c(T) = \max |I(\varphi)|$ 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 $\pi$ phase states at the transition point $T_1$, where the critical current $I_c(T)$ formally changes it sign. The shift of the temperature $T_1$ with respect to $T_0$ depends on the higher harmonics contribution.

For comparison figure 7 shows the current–phase relations $I(\varphi)$ and the temperature dependence of the critical current $I_c(T)$ in a vicinity of the second $\pi$–0 transition (see Fig. E) which takes place at $d_f \leq 5.31\xi_f$. Corresponding value of the coefficient $\beta_2 \approx -0.18 \times 10^{-4}$ gives a small value of the second–harmonic term $(|I_2| \ll |I_1|)$, and the current–phase relation is quite harmonic except the case $T \approx T_c \approx T_1$. The SFS junction reveals the typical nonmonotonic behavior of the critical current $I_c$. 


The cusp of a relative amplitude of the first harmonic \( \kappa \) approach (38), one will be able to determine the amplitudes of harmonics \( a_0 \) and \( \phi \) small \( (|\kappa| < 2) \) in the vicinity of the 0–π transition for \( d_f = 1.94 \xi_f \) \( (T_0^c/T_{c0} \approx 0.332, T_0^c/T_{c0} \approx 0.327, T_c/T_{c0} \approx 0.329) \). The case \( T = T_0 \approx 0.311 T_{c0} \) is shown by the dash-dotted curve \( (I_{\Delta} = I_0 \Delta_0^c(T), \text{ where } \Delta_0(T) = 1.764 T_c \tanh(1.74/\sqrt{T_c/T-1} \text{ is BCS superconducting gap for the temperature } T) \). (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 \( d_s = 2 \xi_s; \sigma_f/\sigma_s = 0.12; \xi_s/\xi_f = 3 \) \( (d_f^c \approx 1.946 \xi_f, \varepsilon = 0.18) \) and \( h = 10 T_{c0} \), \( \tau_s = 7 \).

as a function of the temperature \( T \), and the position of the cusp \( T_f \) 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 amplitudes 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–π transition (see the insert in Fig. 6), 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 \( \pi \) (0) for \( I_1 > 0 \) \( (I_1 < 0) \), respectively, the critical current

\[
I_{c\pm}(\kappa) = \frac{I_2}{32} \left( \sqrt{\kappa^2 + 32} \mp 3|\kappa| \right)^{3/2} \tag{46}
\]

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

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

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:

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

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 \( \lambda_{J2}^{-2} = e \Phi_0 S / 8 \pi^2 I_2 t \) 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 \(^{11}\) 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

\[
W(T, \phi) = E(T, \phi) + \frac{\Phi_0^2}{8 \pi^2 L} (\phi - \phi_c)^2, \quad (50)
\]

where \( \phi_c = 2\pi \Phi_e / \Phi_0 \) is the normalized magnetic flux of an external field through the loop. The total magnetic flux through the loop \( \Phi = (\phi / 2\pi) \Phi_0 \) is related to the external flux \( \Phi_e \) as

\[
\phi_c = \phi + L_1 I(T, \phi) / I_0, \quad (51)
\]

with the normalized inductance \( L_1 = 2\pi L / e \Phi_0 \). 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 \(\pi\) 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 \( T \ll T_c \).

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 \( \pi \) state. The resulting phase transition is first-order, in agreement with the experiments2,3,8 and is responsible for a jump of the amplitude of the superconducting order parameter \( \Delta \), providing the anomalous specific and latent heats in films: attojoule level results have been reported4. We see, therefore, that the latent heat \( Q \) is 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 \( \varphi \) 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 \( \pi \), and the transition between the 0 and \( \pi \) states appears in the vicinity of \( 0 \).\( \pi \) states is manifested strongly due to 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 nonmonotonic 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 \( \pi \) states exist in the superconducting loop interrupted by the junction. The coexistence of 0 and \( \pi \) states is manifested as two jumps in the dependence of enclosed magnetic flux in the loop per period2,3.

Acknowledgments The 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 contribu-
tion to the the current–phase relation in the framework of the rigid boundary conditions. The general expression for the supercurrent through a SFS junction is given by

\[
I_s = -I_0(T/T_c) \sum_{\omega > 0} \text{Im} \left\{ F^+ F^s_\omega \right\}, \quad (A1)
\]

where \( F^+ (s, h) = F^* (s, -h), I_0 = 4\pi T_c e SN(0) D_f / \xi_f, S \) 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 Eqs. (1), (7) for \( T \approx T_c \) and an arbitrary phase difference \( \varphi \) is

\[
F(s) \simeq a \cosh(gs) + b \sinh(ps) + \frac{b^2 - a^2}{8k^2} \left( \alpha + \frac{3i}{4} \right) \left[ a \cosh(3ks) + b \sinh(3ks) \right], \quad (A2)
\]

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

\[
q \simeq k - \frac{a^2}{2k} (\alpha + i/4) + \frac{b^2}{2k} (\alpha + 5i/4), \quad (A3)
\]

\[
p \simeq k - \frac{a^2}{2k} (\alpha + 5i/4) - \frac{b^2}{2k} (\alpha + i/4). \quad (A4)
\]

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 \( \omega \) at the boundary of left (right) S layer coincides with the bulk one: \( F_s(\omega, \pm s_f) = \Delta_B e^{\pm i \varphi/2}/\Omega, \) where \( \Delta_B \) is the temperature dependent BCS order parameter and \( \Omega = \sqrt{\omega^2 + |\Delta_B|^2}. \) Using the boundary conditions

\[
F(\pm s_f) = F_s(\omega, \pm s_f) \quad (A5)
\]

we get from (A2), (A4), (A1) the following expressions for the amplitudes \( a \) and \( b \):

\[
a = a_0 + a_1, \quad b = b_0 + b_1, \quad (A6)
\]

\[
a_0 = \frac{\Delta_B}{\Omega} \cos(\varphi/2) \cosh(\delta/2), \quad b_0 = -\frac{\Delta_B}{\Omega} \sin(\varphi/2) \sinh(\delta/2), \quad (A7)
\]
The last equality determines the coefficient $\beta$. I. A. Buzdin, Phys. Rev. B

Actually the Josephson energy (39) is the phase dependent contribution to the Ginzburg-Landau energy (40). M. Houzet, V. Vinokur and F. Pistolesi Phys. Rev. B 12

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

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

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

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

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

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

The last equality determines the coefficient $\beta_2$ (42) in the functional (10):

$$\beta_2 = -\frac{\pi}{192} \left( \frac{T_c}{\Delta_B} \right)^3 \frac{h}{\pi c} \text{Im} \left\{ \frac{1}{k \sinh^2 \delta} \left[ \delta - i \frac{\alpha + i/4}{\sinh \delta} \left( \cosh \delta - \frac{\delta}{\sinh \delta} \right) \right] \right\}. \quad (A13)$$
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