Time-reversal symmetry breaking and spontaneous currents in *s*-wave/normal-metal/*d*-wave superconductor sandwiches

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We study the physical properties of an *s*-wave–normal-metal–*d*-wave junction in terms of the Andreev bound-state solutions to the Bogoliubov–de Gennes equation in the normal-metal layer. The phase dependence of bound states with different orientations leads to superconducting states with broken time-reversal symmetry for generic orientations of the *d*-wave superconductor crystal. The occurrence of such a state and the associated spontaneous supercurrent along the junction and magnetic field is analyzed also in the framework of Ginzburg-Landau theory. [S0163-1829(97)00246-4]

I. INTRODUCTION

During the last few years the order-parameter symmetry has been one of the intensively debated issues in the field of high-temperature superconductivity. A growing number of experiments leaves little doubt that the basic symmetry of the Cooper pairs has $d_{x^2-y^2}$-wave character in many of the high-temperature superconductors.[1,2] The unconventional symmetry of the order parameter has important implications for the Josephson effect. For d-wave superconductors the Josephson coupling is subject to an additional phase dependence caused by the internal phase structure of the Cooper pair wave function. The phase properties of the Josephson effect have been discussed within the framework of the generalized Ginzburg-Landau[3] (GL) as well as the tunneling Hamiltonian approach.[4] It was found that the current-phase relation depends on the mutual orientation of the two coupled superconductors and their interface. This property is the basis of all the phase-sensitive experiments probing the order-parameter symmetry. In particular, it is possible to create multiply connected d-wave superconductors which generate half-integer flux quanta as observed in experiments.[5]

Various interesting phenomena occur in 45° interfaces of $d_{x^2-y^2}$-wave superconductors, where one of the nodes of the pair wave function lies parallel to the interface normal vector (Fig. 1). For an interface to a normal metal or an insulator a bound state appears at zero energy giving rise to a zero-bias anomaly in the I-V characteristics of quasiparticle tunneling[6] and a nontrivial temperature dependence of the critical current.[7,8] It was also shown that in such an interface to an s-wave superconductor the energy minimum corresponds to a Josephson phase different from 0 or π.[9] Based on Ginzburg-Landau theories it was suggested that this is connected with a local breakdown of time reversal symmetry $\mathcal{T}$.[10–12] The s-wave and d-wave order parameter can form a complex combination, a so-called $(s+id)$ state, close to this 45° junction. This leads to a phase difference of $+\pi/2$ or $-\pi/2$ across the interface, which corresponds to two degenerate states.[12,13] It can be seen from the GL formulation that under this condition a spontaneous current flows parallel to the interface which produces a local field distribution.[11]

In this paper we consider a 45° interface with a normal metal between the d-wave and the s-wave superconductor, a device which we call the SND junction.[14] Also for this configuration a $\mathcal{T}$-violating state appears and generates a supercurrent mainly in the region of the normal metal. It is our goal to demonstrate that this current has a simple and intuitive interpretation in terms of subgap Andreev bound states in the sandwiched normal-metal layer. Let us first outline the basic idea for the situation shown in Fig. 1, where $\alpha=\pi/4$ and the c axis is parallel to the interface. In terms of the phase difference $\varphi=\varphi_d-\varphi_s$, the Josephson current carried by a bound state with a specific orientation β can be ex-

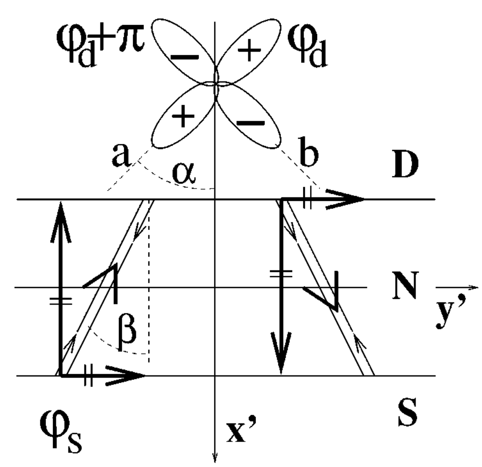

FIG. 1. Schematic view of the SND junction. The angle α denotes the orientation of the d-wave superconductor (crystal axis a and b) and β the momentum direction of the bound state. The currents generated by the bound states tend to cancel in the direction perpendicular to the interface, whereas they add parallel to the interface and generate a spontaneous current.

panded in harmonics of the phase difference φ as $I_\beta(\varphi)=I_1(\beta)\sin(\varphi)+I_2(\beta)\sin(2\varphi)+\cdots$. In the geometry considered, each bound state with orientation $0<\beta<\pi/4$ that sees the ''+'' lobe with phase φ_d, has a mirror bound state with orientation $-\beta$ that sees the ''−'' lobe of the $d_{x^2-y^2}$-pair wave function with phase $\varphi_d+\pi$. As a result, in the total current perpendicular to the interface, all odd harmonics cancel, and the Josephson coupling is reduced. The leading term is $I_\perp \sim \sin(2\varphi)$,[15–17] and the stable ground state with $I_\perp=0$ is at $\varphi=\pm\pi/2$ and, thus, breaks time-reversal symmetry. The Josephson current parallel to the interface, however, has contributions from the odd harmonics and to leading order $I_\parallel \sim \sin(\varphi)$. Remarkably, this parallel contribution is *nonzero* in the ground state and constitutes a spontaneous current — a direct manifestation of the broken time-reversal symmetry.

In the following two sections, we discuss these phenomena within the Ginzburg-Landau framework and in terms of the Andreev bound states in the normal-metal layer, respectively.

II. GINZBURG-LANDAU ANALYSIS

Let us first consider the properties of the SND junction on a phenomenological level by means of GL theory. We describe the superconducting state by two order parameters, η_s (s wave) and η_d (d wave), which correspond to the local pairing amplitudes. The corresponding GL free energy $\mathcal{F}$ has the general form

$$\frac{\mathcal{F}}{f_0}=\int d^3r\Bigg[\sum_{\mu=s,d}\left\{\left(\frac{T}{T_{c\mu}}-1\right)|\eta_\mu|^2+\beta_\mu|\eta_\mu|^4+\xi_\mu^2|\mathbf{\Pi}\eta_\mu|^2\right\}+\gamma_1|\eta_s|^2|\eta_d|^2+\frac{\gamma_2}{2}(\eta_s^{*2}\eta_d^2+\eta_s^2\eta_d^{*2})+\frac{(\nabla\times\mathbf{A})^2}{8\pi f_0}+\tilde{\xi}^2[(\Pi_x\eta_s)^*(\Pi_x\eta_d)-(\Pi_y\eta_s)^*(\Pi_y\eta_d)+\text{c.c.}]\Bigg], \quad (1)$$

where f_0 is a free-energy density, T_{cs} and T_{cd} are the transition temperatures of η_s and η_d, respectively, and $\beta_{s,d}$, $\gamma_{1,2}$, $\xi_{s,d}$, and $\tilde{\xi}$ are real coefficients ($\xi_{s,d}$ corresponds to the zero-temperature coherence length). These coefficients and the transition temperatures are in general different in the three regions of the SND junction. We have introduced the notation $\mathbf{\Pi}=\nabla-(2\pi i/\Phi_0)\mathbf{A}$, with vector potential $\mathbf{A}$ and flux quantum $\Phi_0=hc/2e$. To study the properties of the SND junction we minimize this free energy with respect to $\eta_{s,d}$ and $\mathbf{A}$. Assuming homogeneity along the interface the problem reduces to one spatial dimension which corresponds to the [1,1,0] direction in the coordinates used in $\mathcal{F}$ ($\hat{x}=\hat{a}$ and $\hat{y}=\hat{b}$). We call this direction x' and the perpendicular ones y' and z.

We solve the complete set of GL equations numerically, for the case in which the coefficients in $\mathcal{F}$ are identical for both order parameters and throughout the system. The transition temperatures are only different from zero in the corresponding superconducting regions. We assume the interfaces between the different layers to be completely transparent, i.e., the order parameters are continuous and have a continuous derivative. For our calculation we choose $\beta_s=\beta_d=1/2$,

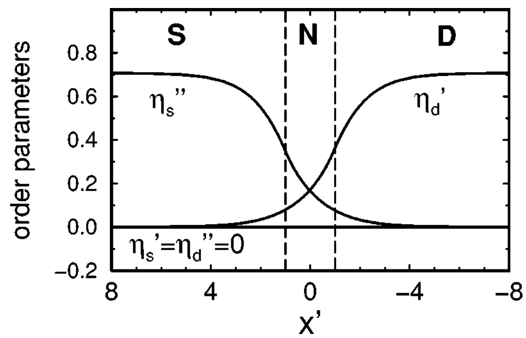

FIG. 2. Spatial dependence of the order parameter in the SND junction ($\alpha=\pi/4$) based on the GL theory. The parameters of GL free energy are given in the text. The temperature is $T=T_{cs,d}/2$ and the width L of the normal-metal layer is 2 in units of ξ_s.

$\xi_s=\xi_d=1$ (unit of length), $\gamma_1=4/5$, $\gamma_2=2/5$ and $\tilde{\xi}=1$. This leads to $f_0=H_c^2/8\pi$, where H_c is the thermodynamic critical field at $T=0$. We fix $\Phi_0/2\sqrt{2}\pi H_c\xi_s^2=4$ which corresponds to the London penetration depth λ at $T=0$ in units of ξ_s. The result is shown in Fig. 2 for the order parameters and in Fig. 3 for the magnetic field and the supercurrent along the y' direction.

Both order-parameter components penetrate the normal-metal layer (proximity effect) and coexist there in a combination, which for the case $\alpha=\pi/4$ is entirely determined by the mixing term $(\gamma_2/2)(\eta_s^{*2}\eta_d^2+\eta_s^2\eta_d^{*2})$. Within the weak-coupling approach which we assume to apply, at least, within the normal-metal layer, γ_2 is positive.[18] This term yields the basic $\cos(2\varphi)$ dependence of the SND-junction free energy. It fixes the phase difference between η_s and η_d to $\varphi=\varphi_d-\varphi_s=\pm\pi/2$ in accordance with the argument given above. The mixed state has the $\mathcal{T}$ violating $s\pm id$ character in the normal metal.

The supercurrent density follows from $\mathcal{F}$ as $\mathbf{J}=-2c\,\partial\mathcal{F}/\partial\mathbf{A}$. We find that the current component $J_{x'}=J_\perp$ vanishes in the junction state with lowest energy and that a spontaneous supercurrent flows parallel to the y' direction and generates a magnetic-field distribution B_z in and close to the metal layer (Fig. 3). Within the GL formulation the supercurrent $J_{y'}=J_\parallel$ is a result of the spatial variation of the two order-parameter components,

$$J_{y'}=\frac{\pi c\tilde{\xi}^2}{\Phi_0}\mathrm{Im}\{\eta_s\partial_{x'}\eta_d^*+\eta_d\partial_{x'}\eta_s^*\}, \quad (2)$$

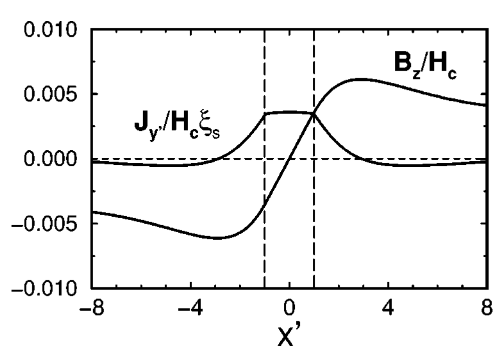

FIG. 3. Spatial dependence of the spontaneous supercurrent and the magnetic field in the SND junction based on the GL theory under the same conditions as in Fig. 2.

where we have omitted the diamagnetic part which contributes only to the Meissner screening effect. Note that this part of $J_{y'}$ has essentially the $\sin\varphi$ dependence anticipated above. Under symmetric conditions, $J_{y'}$ depends only weakly on x' inside the normal-metal layer as shown in Fig. 3. The variation of the order parameters just outside the normal-metal layer, and the corresponding supercurrent, decay on the length scale ξ. The induced magnetic field is screened perpendicular to the interface on the scale of the London penetration depth in the superconducting regions by smaller currents flowing in the opposite direction.

III. BOUND STATES IN THE NORMAL LAYER

Let us turn now to the microscopic view by considering the bound-state solutions to the Bogoliubov–de Gennes equation in the normal-metal layer[19] under the symmetric condition, i.e., the d-wave energy gap in D has the form $\Delta_d=|\Delta|\mathrm{sgn}(\cos[2(\beta-\alpha)])$, with the amplitude $|\Delta|$ equal to that of the gap of the s-wave superconductor in S. We take the Fermi momenta in S, N, and D to be equal and the transparency of the interfaces again to be high. Furthermore, we also neglect the suppression of the energy gap near the normal metal and assume the pairing interaction to be zero in N.

We assume in the following that the width L of the normal-metal layer is much larger than both of the coherence lengths, ξ_s and ξ_d. Furthermore, the temperature shall be low enough to satisfy $k_BT\ll|\Delta|$. Under these conditions mainly the bound states close to the Fermi energy are important for the Josephson current. Their energy is approximately $E_{\mu,\hat{\mathbf{k}}_F}(\varphi)\approx[\mu\pi\pm\varphi/2]\hbar v_F/L_{\hat{\mathbf{k}}_F}$, where μ is half-integer ($\pm 1/2$, $\pm 3/2$, ...) and $L_{\hat{\mathbf{k}}_F}$ denotes the effective direction-dependent thickness of the normal-metal layer, $L_{\hat{\mathbf{k}}_F}=Lk_F/(k_F^2-k_{y'}^2-k_z^2)^{1/2}$. The current carried by the bound state with momentum direction parallel to $\hat{\mathbf{k}}_F$ is given by $\mathbf{J}=2e\hat{\mathbf{k}}_F\partial F_{\hat{\mathbf{k}}_F}/\partial\varphi_{\hat{\mathbf{k}}_F}$ where $F_{\hat{\mathbf{k}}_F}$ is the free-energy contribution of these bound states. For the total current we obtain

$$\mathbf{J}=\int\frac{dk_{y'}}{2\pi}\frac{dk_z}{2\pi}\frac{2e\mathbf{k}_F}{m\pi L}\sum_{n=1}^{\infty}\frac{(-1)^{n+1}}{n}f_{n,\hat{\mathbf{k}}_F}\sin[n\varphi_{\hat{\mathbf{k}}_F}],\quad(3)$$

with the Fermi momentum $\mathbf{k}_F=(k_{x'},k_{y'},k_z)$ and $k_{x'}^2+k_{y'}^2+k_z^2\approx k_F^2$. The integral runs over all transverse momenta $k_{y'}^2+k_z^2\leq k_F^2$ and the sum over all possible numbers of multiple Andreev reflections n. The phase difference between S and D is direction dependent, and in the above expression $\varphi_{\hat{\mathbf{k}}_F}=\varphi_d-\varphi_s+(\pi/2)[1-\mathrm{sgn}(\cos[2(\beta-\alpha)])]$. The factors $f_{n,\hat{\mathbf{k}}_F}$ take the suppression due to thermal decoherence and impurity scattering[17] (broadening of the Andreev levels) into account,

$$f_{n,\hat{\mathbf{k}}_F}=\exp(-2nL_{\hat{\mathbf{k}}_F}/l)\frac{nL_{\hat{\mathbf{k}}_F}/\xi_T}{\sinh[nL_{\hat{\mathbf{k}}_F}/\xi_T]},\quad(4)$$

where we have introduced the normal-metal coherence length $\xi_T=\hbar v_F/(2\pi k_BT)$ in the clean limit and the mean free path l. For the orientation $\alpha=\pi/4$, the symmetry of $\varphi_{\hat{\mathbf{k}}_F}$ immediately leads to the phase dependence of the current anticipated in the Introduction. For currents perpendicular to the interface only contributions of even-n in the sum survive, while for the current component along the metal layer only the odd-n components occur.

The simplest case is that of zero temperature in the absence of impurities, so that $\xi_T=l=\infty$ and all $f_n\equiv 1$. In this limit the sums over n give sawtooth functions of the phase difference, $\mathrm{saw}[\varphi_{\hat{\mathbf{k}}_F}]=\{([\varphi_{\hat{\mathbf{k}}_F}+\pi]\mathrm{mod}2\pi)-\pi\}/\pi$. We obtain the Josephson currents perpendicular and parallel to the junction by angular integration in terms of $k_{y'}=k_F\sin(\theta)\sin(\beta)$ and $k_z=k_F\cos(\theta)$. Summing over the transverse momenta ($k_{y'}$, k_z) in this manner gives a slightly different result as compared to the isotropic orientational average used in Refs. 15 and 17. The physics, however, does not depend on the exact choice of measure for the angular integral. The result is

$$I_\perp=A_\perp J_0\left(\frac{1}{2}[\mathrm{saw}(\varphi+\pi)+\mathrm{saw}(\varphi)]+\frac{1}{\pi}\cos(2\alpha)[\mathrm{saw}(\varphi+\pi)-\mathrm{saw}(\varphi)]\right),\quad(5)$$

$$I_\parallel=A_\parallel J_0\frac{1}{\pi}\sin(2\alpha)[\mathrm{saw}(\varphi+\pi)-\mathrm{saw}(\varphi)].\quad(6)$$

Here $A_\perp$ and $A_\parallel$ denote the perpendicular and parallel cross section of the junction, and $J_0=ek_F^3/(6\pi mL)$. Note that the current density J_0 is inversely proportional to L, as in the GL calculation. The junction free energy $F(\varphi)$ is found by integrating $I_\perp$ with respect to the phase. It has two degenerate minima at phase differences $\varphi_0=\pm[\pi/2-\cos(2\alpha)]$, which correspond to a parallel current along the junction $I_\parallel=\pm A_\parallel J_0\sin(2\alpha)/\pi$. The ground state has $(s+e^{i\varphi_0}d)$ character in the normal-metal layer as in the phenomenological treatment, again reflecting $\mathcal{T}$ violation.

For nonzero temperature and in the presence of impurities, we evaluate $I_\perp$, $I_\parallel$, and the junction free energy F numerically. In Fig. 4 the equilibrium phase difference φ_0 across the junction is plotted as a function of orientation angle α for different temperatures ξ_T/L in the case $l=\infty$. We find that time-reversal symmetry is broken (φ_0

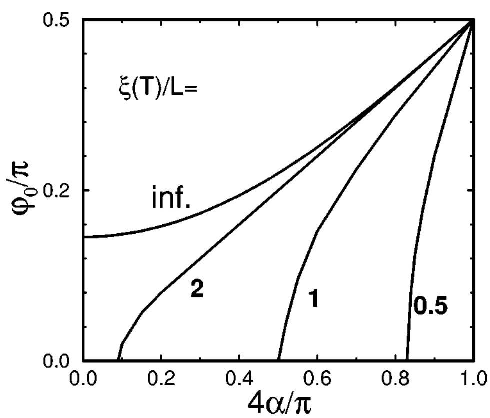

FIG. 4. The ground-state phase difference $\pm\varphi_0$ as a function of orientation angle α for temperatures corresponding to $\xi(T)/L=\infty$, 2, 1, and 0.5.

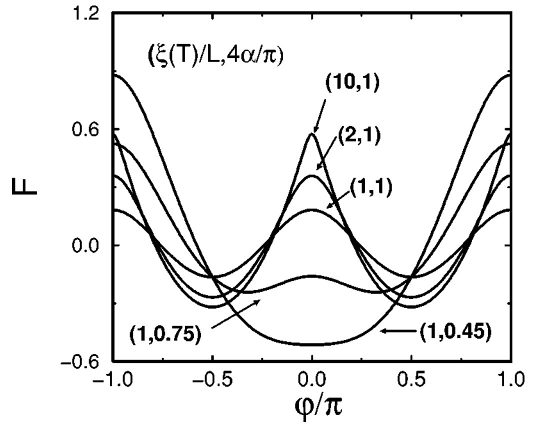

FIG. 5. The junction free energy F as a function of the phase for $\alpha=\pi/4$ and $\xi_T/L=$ 10, 2, 1, and for $\alpha=0.45\ \pi/4$ (one minimum) and $0.75\pi/4$ (two shallow minima) at $\xi_T/L=1$.

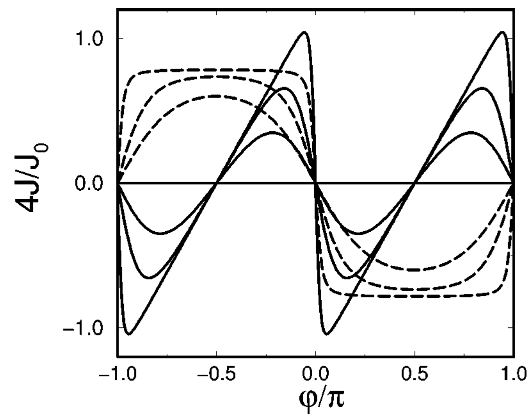

FIG. 6. The parallel (dashed line) and perpendicular (solid line) Josephson current densities as a function of phase difference for $\alpha=\pi/4$ and temperatures corresponding to $\xi_T/L=$ 10,2,1 (decreasing amplitude).

$\neq 0,\pm\pi$) only for low enough temperatures, or for the orientation angle α exceeding a critical value. For $\alpha=\pi/4$, however, $\varphi_0=\pm\pi/2$ for all $T<T_{cs},T_{cd}$ as in the GL treatment of Ref. 12. The resulting phase diagram is consistent with the one found by GL theories.[12] The time-reversal symmetry-breaking state extends even further than anticipated in the GL treatment. The result for the junction free energy F is plotted in Fig. 5, and for $I_\perp$ and $I_\parallel$ in Fig. 6. Both temperature and disorder smear the sharp sawtooth structures found at $T=0$ in the clean limit in a similar fashion, however, without removing the essential feature of the broken time-reversal symmetry.

IV. DISCUSSION

The arbitrary equilibrium phase difference across the junction, as discussed in the two previous sections, leads to experimentally observable effects. $\mathcal{T}$-violating junctions can lead to phase windings which are noninteger multiples of π, giving rise to non-half-integer flux quantization. Thus, it is possible to create devices including $\mathcal{T}$-violating junctions which generate a spontaneous arbitrary magnetic flux.[11,12] The observation of such a deviation from standard flux quantization is a clear sign of $\mathcal{T}$ violation. Furthermore, the presence of two degenerate equilibrium states allows for hysteresis effects, i.e., phase slips between the states with φ_0 and $-\varphi_0$. By applying a current through the junction one can switch between the two states. This effect corresponds to a phase slip with a fractional flux moving along the junction.[12] This leads to dissipation and the enhancement of microwave absorption as soon as the junction enters the $\mathcal{T}$-violating phase.

Direct observation of the spontaneous currents $I_\parallel$ or the field might be difficult, since they average to zero over rather small length scales (London penetration depth). The maximum magnetic field on both sides of the sandwich for the geometry with $\alpha=\pi/4$ can be estimated to be $\pm B_z=2J_0/c=\Phi_0/(4\pi\lambda^2)$, which is of order of the lower critical field H_{c1} of the superconductors. This value is comparatively large and would be renormalized by screening effects only if the width L of the junction exceeds the London penetration depth λ. Thus, a very sensitive probe with high spatial resolution would, in principle, be able to directly probe the magnetic field in the junction.

Extension of the above discussion to a DND junction is straightforward. The direction-dependent phase in the DND case depends on the orientations of both d-wave superconductors between which the normal metal is sandwiched. Assuming $0<\alpha_1<\alpha_2<\pi/4$ and $l,\xi_T\to\infty$, we obtain for instance for the perpendicular current

$$J_\perp=J_0\left(\frac{1}{2}\text{saw}(\varphi)-(\alpha_2-\alpha_1)[\text{saw}(\varphi+\pi)-\text{saw}(\varphi)]/\pi\right), \tag{7}$$

from which we extract a ground-state phase difference across the junction $\varphi_0=\pm 2(\alpha_2-\alpha_1)$ at zero temperature. The generalization to other order-parameter symmetries is straightforward as well.

In summary, we have demonstrated that the Andreev bound states in the normal-metal layer of an SND junction are the microscopic realization of local $\mathcal{T}$ violation and provide a clear understanding of the spontaneous current found in the phenomenological Ginzburg-Landau analysis. This observation allows for a more quantitative consideration of this effect, which will be important for future experimental investigations, in particular, also in the field of mesoscopic devices. Finally, we like to emphasize that this effect is only possible in connection with unconventional superconductivity and cannot occur for standard SNS junctions. Therefore, high-temperature superconductivity opens a door to interesting phenomena in normal-metal–superconductor interfaces.

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