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Superharmonic Josephson relations in unconventional superconductor junctions with a ferromagnetic barrier

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For misorientation of 45° in the *a*-*b* plane of Josephson junctions between two *d*-wave superconductors separated by a ferromagnet, unusual superharmonic current-phase relations (CPR) are predicted in the clean case and for high barrier transparency. Depending on the strength of the magnetic barrier influence, junctions with triply degenerate 0, $\pi/2$, and π equilibrium states exhibit CPR similar to $I \sim I_c \sin 4\phi$ at low temperature, whereas junctions with doubly degenerate 0 and π states or junctions with a nondegenerate state $\pi/2$ have CPR close to $I \sim \pm I_c \sin 2\phi$, respectively, at all temperatures. In the presence of an external magnetic field, the corresponding critical currents oscillate with periods of $\phi_0/4$ and $\phi_0/2$.

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Recently, devices containing Josephson junctions with nontrivial phase differences and nonsinusoidal current-phase relations (CPR) have been shown to be a promising system for observation of macroscopic quantum tunneling[1] and for realization of "silent" phase quantum bits [superconducting quantum interference devices (SQUIDs)].[2] In particular, studies of the combined influence of unconventional superconductivity and ferromagnetism in heterostructures have attracted great attention due to their scientific importance and potential for applications in quantum electronic.[3] Experimental progress is reported in several studies of superconductivity and ferromagnetism in bilayers[4] and superlattices[5] of *d*-wave oxide superconductors (D) and perovskitelike ferromagnetic oxides and of hybrid metallic ferromagnet/*d*-wave superconductor structures.[6] Theoretically, spin-polarized transport in ferromagnet (F)/unconventional superconductor heterostructures was studied by several authors[7] and Josephson current in D/F/D junctions was demonstrated[8] to be in general nonsinusoidal. In addition to the D/F/D junctions, unusual CPR and the possibility of π coupling (equilibrium phase $\phi_{eq}=\pi$) was predicted over 20 years ago[9] for conventional superconductor/ferromagnet/superconductor (S/F/S) junctions, due to the exchange-field-induced oscillation of the order parameter in the ferromagnetic barrier. The recent discovery of such π junctions[10] was followed by intense theoretical and experimental activity in this field.[11] Note that the absence of the second harmonic in some cases[12] was explained as due to the fact that the barrier is not in the clean limit, as it should be for second-order Josephson tunneling to dominate the CPR near 0-π transitions.[13]

However, junctions with a ferromagnetic barrier are not the only ones with nontrivial phase differences. π coupling was first observed in high-temperature superconductors junctions[14] and explained as due to a sign change of the order parameter on the Fermi surface for unconventional (*d*-wave) pairing. Theoretical investigations of the Josephson current in unconventional superconductor junctions have shown that the CPR are nonsinusoidal and that the energy minimum could correspond to an equilibrium phase not only equal to 0 or π, but varying between these values.[15] In a self-consistent treatment of pinhole junctions in *d*-wave superconductors (as a model of weak links) it was found[16] that the CPR are nonsinusoidal and that there are zones of electrode orientations where ϕ_{eq} continuously evolves from zero to π. Experimentally, nonsinusoidal CPR and half-fluxon periodicity were first found by Il'ichev *et al.*[17] in asymmetric 45° junctions between two *d*-wave superconductors. In $YBa_2Cu_3O_{7-\delta}$ (YBCO) SQUIDs with (100)/(110) boundaries, Schneider *et al.* measured half-fluxon periodicity of the critical current in an applied field, consistent with a strong second harmonic component of the CPR.[18] Unconventional CPR were also observed in YBCO dc SQUIDs with $0°-45°$grain boundaries by Lindstrom *et al.* One explanation of the pronounced effect of the second harmonic in the CPR could be that relatively large sections of interface are highly transparent and have a low degree of disorder.[19]

The recent experimental progress should provide the possibility of fabrication of clean D/F/D junctions with a high degree of barrier transparency, in search of novel phenomena in misoriented 0°–45° D/F/D junctions. In such structures, we predict the coexistence of 0 and π stable phases (doubly degenerate ground states) in finite intervals of the magnetic barrier strength, alternating with intervals of $\pi/2$ stable states. At low temperature, there are also triply degenerate states at the limits of the above intervals. The corresponding CPR are quite unconventional. This should open the possibility for SQUIDs with D/F/D junctions.

As in previous papers,[20–22] we consider Josephson junctions using the quasiclassical approach,[23] and assuming both superconducting electrodes and the ferromagnetic metal barrier clean with transparent barrier interfaces. As in the S/F/S case,[24] the main supercurrent transport mechanism here is via induced Andreev states in the barrier. In the ferromagnet barrier, there is a splitting of the conduction band and the electron with spin up (parallel to the magnetization) is lowered in energy by h, the exchange field energy, while the down-spin electron is raised in energy by the same amount.[25] The ferromagnetic barrier influence is measured by a parameter $Z=2hd/\hbar v_0$, where d is the barrier thickness and v_0 the Fermi velocity, assumed to be the same in the superconductor and the ferromagnet. The barrier is assumed perpendicular to the $\hat{a}$ axis in the $\hat{a}$-$\hat{b}$ plane of the left-hand superconducting electrode S_L, which is misoriented by $\theta=45°$ with respect to the right-hand one S_R (see Fig. 1). For $d_{x^2-y^2}$

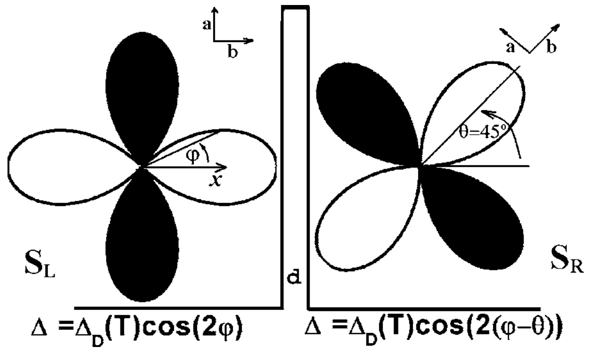

FIG. 1. Schematic picture of a junction with misoriented electrodes and $d_{x^2-y^2}$ symmetry of the gap with four node lines.

symmetry, the pair potential in S_L is $\Delta^L(\mathbf{v}_0)=\Delta_0(T)\cos 2\varphi$, where φ is the angle the quasiparticle momentum makes with the $\hat{a}$ axis, and here $\Delta_0(T)=\Delta_0 F(t)$, with $F(t)=\tanh(1.74\sqrt{1/t-1})$, and $t=T/T_c$. Similarly, in S_R, $\Delta^R(\mathbf{v}_0)=\Delta_0(T)\cos 2(\varphi-\theta)$. We take a step-function variation of the pair potential along the x axis perpendicular to the barrier, $\Delta(x)=\Delta^L\Theta(-d/2-x)+\Delta^R\Theta(x-d/2)$, where $\Delta^{L,R}=\Delta^{L,R}(\varphi)e^{\pm i\phi/2}$, ϕ being an intrinsic phase difference at the contact related to the passage of the supercurrent. This approximation should be valid for thin ferromagnetic barriers (of thickness smaller than or close to the superconducting coherence length ξ_0). Also, we expect that, as in the case of a pinhole junction,[16] a self-consistent calculation of the order parameter will not change the energy minima positions, only reducing the critical current amplitude. This expectation is corroborated by the fact that in clean ballistic junctions without interface scattering potential the magnitude of the exchange field in the ferromagnet has little effect on the order parameter profile near the interface.[26]

Solving the Eilenberger quasiclassical equations,[23] we found[20] the temperature Green's function in the barrier for one spin (down) orientation $g=g_\downarrow$. For $|x|\le d$ and $\omega_n=\pi k_B T(2n+1)$,

$$g(\varphi,\phi,\tilde{\omega}_n,\theta;\tilde{h},t)=-\frac{A+Be^{i\chi}}{A-Be^{i\chi}}, \tag{1}$$

where $\chi=\phi+(2/\pi)\tilde{h}\tilde{d}/\cos\varphi-i(2/\pi)(\tilde{\omega}_n/\cos\varphi)F(t)\tilde{d}$, $A=\tilde{\Delta}_L(\tilde{\omega}_n-\tilde{\Omega}^R)$, $B=\tilde{\Delta}^R(\tilde{\omega}_n+\tilde{\Omega}^L)$, and $\tilde{\Omega}^{L,R}=\sqrt{\tilde{\Delta}^{L,R}+\tilde{\omega}_n^2}$. Here and in the following we take $\hbar=c=k_B=1$ and introduce the dimensionless quantities $\tilde{\Delta}^{L,R}=\Delta^{L.R}/\Delta_0(T)$, $\tilde{\omega}_n=\omega_n/\Delta_0(T)$, $\tilde{h}=h/\Delta_0$, $\tilde{d}=d/\xi_0$, where the coherence length $\xi_0=v_0/\pi\Delta_0$. The Green's function for the opposite spin direction, $g=g_\uparrow$ is obtained by changing $h\to -h$.

The current density is expressed via the Green's functions in the barrier[22]:

$$\mathbf{j}(\phi,\theta;\tilde{h},t)=-2ieN_0T\sum_{\omega_n}\left\langle \mathbf{v}_0\frac{g(\tilde{h})+g(-\tilde{h})}{2}\right\rangle, \tag{2}$$

where $\langle\cdots\rangle$ means averaging over the Fermi surface (assumed circular). The supercurrent through the barrier of area S, $I=j_xS$, is given by

$$I(\phi,\theta;\tilde{h},t)/I_0=\frac{T}{\Delta_0(T)}2\lim_{m\to\infty}\sum_{n=0}^{m}2\mathcal{D}(\varphi_c)\times\int_{-\varphi_c}^{\varphi_c}\frac{\operatorname{Im} g(\tilde{h})+\operatorname{Im} g(-\tilde{h})}{2}\cos\varphi\, d\varphi. \tag{3}$$

The temperature-dependent normalizing current is $I_0=2\Delta_0(T)/eR_N$, where R_N is the normal state resistance of a corresponding weak link with normal metal barrier ($h=0$). It is given by $R_N^{-1}=e^2v_0N_0S$, N_0 being the density of states at the Fermi level. We model the barrier with a uniform probability distribution equal to 1 for all the quasiparticles propagating within an acceptance cone of angle $2\varphi_c$ about the interface normal, and zero outside the cone. Thus $2\mathcal{D}(\varphi_c)=2/\int_{-\varphi_c}^{\varphi_c}\cos\varphi\, d\varphi=1/\sin\varphi_c$. The angle φ_c depends on the dimensions of the barrier, which we assume thin and not too short. For any $\varphi_c>45°$ the supercurrent differs qualitatively only.

The equilibrium phase difference ϕ_{eq} at the contact can be found via minimization of the junction free energy $W(\phi)$. In reduced units

$$\tilde{W}(\phi)=\frac{1}{I_c}\int_0^{\phi}I(\phi')d\phi' \tag{4}$$

where $\tilde{W}=W/(\Phi_0I_c/2\pi)$, Φ_0 is the flux quantum, and I_c is the critical current, which for given ferromagnetic barrier depends on θ and the temperature t.

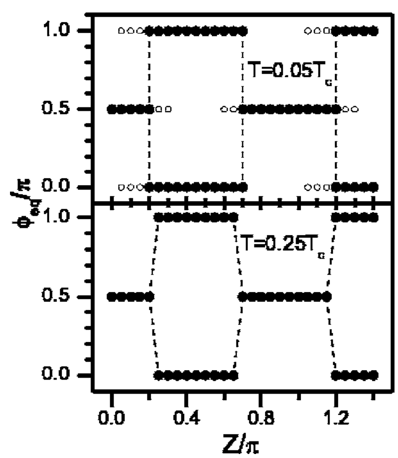

FIG. 2. Top: Equilibrium phase difference at the junction as a function of $Z=2\tilde{d}\tilde{h}/\pi$ at low temperature $T=0.05T_c$. Stable states, full circles; metastable states, open circles. Note the triply degenerate stable states 0, $\pi/2$, and π at $Z=Z_c/\pi\approx 0.2$, 0.7, and 1.2. In addition to the intervals of coexistence of stable and metastable states at low temperature, there are finite intervals of Z with doubly degenerate 0 and π stable states, alternating with intervals of a $\pi/2$ stable state at all temperatures. Bottom: Same as above, but at high temperature $T=0.25T_c$. Only stable states (full circles) are present. Note that the triplet states $(0,\pi/2,\pi)$ found at low temperature disappear.

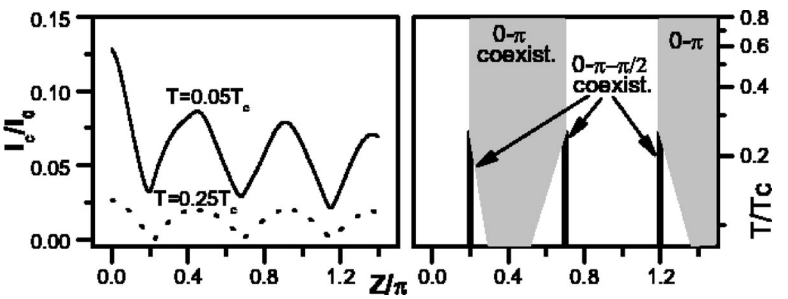

FIG. 3. Left: Normalized critical current as function of magnetic barrier influence Z at low and higher temperatures. Right: Phase diagram showing regions of T,Z space in which the 0-π and 0-π-$\pi/2$ coexistence can be observed.

The results of numerical calculations of the equilibrium phase for $\theta=\pi/4$, $\tilde{d}=1$, and $\varphi_c=80°$ as a function of $Z=2\tilde{d}\tilde{h}/\pi$ (in reduced units) are shown in Fig. 2. In D/F/D junctions, there are several additional features. First, there are doublet states, with finite coexistence intervals of stable 0 and π states, by contrast to the S/F/S junctions, where this coexistence is found only for a critical value of Z.[21] As in the S/F/S case,[21] there is coexistence of stable and metastable states at low temperature. However, in D/F/D junctions this is the coexistence of two stable and one metastable state (0 and π stable and $\pi/2$ metastable), or vice versa, in finite intervals of Z. Between the coexistence intervals, in a finite range of Z, $\phi_{eq}=\pi/2$ is stable. At low temperature, for some critical values of $Z=Z_c$ ($Z_c/\pi\approx 0.2$, 0.7,1.2) there are triplets of stable states, $\phi_{eq}=0,\pi/2,\pi$ (Fig. 2). These values of Z correspond to the (nonzero) minima in the critical current as a function of Z, Fig. 3 (left). The phase diagram in (T, Z) space showing the regions of stable doublet and triplet states is presented in Fig. 3 (right). Note that we do not present the results for $Z\gg\pi$, which would correspond to values of the exchange field so strong that the superconducting electrodes become decoupled. The accompanying plots of $W(\phi)$ and $I(\phi)$ at low temperature, for Z where only stable states are present, are given in Fig. 4. For comparison, the results for $Z=0$ are included in Figs. 4(a) and 4(b). For stable triplets, $W(\phi)$ has minima at 0, $\pi/2$, and π [Fig. 4(c)], and the CPR are similar to a deformed $I\sim\sin 4\phi$ curve [Fig. 4(d)]. For coexistence of stable 0 and π states [Fig. 4(e)], we find CPR similar to $I\sim\sin 2\phi$ [Fig. 4(f)], whereas for a stable $\pi/2$ state [Fig. 4(g)], they are similar to $I\sim\sin(2\phi+\pi)$ [Fig. 4(h)]. The corresponding plots for coexistence of stable and metastable states are given in Fig. 5. At high temperature the metastable states, as well as the triplet states with three-node CPR in the interval $0<\phi_{eq}<\pi$ disappear (see also Fig. 2). There are intervals of Z where the only stable state is $\phi_{eq}=\pi/2$, and the CPR are similar again to $I\sim\sin(2\phi+\pi)$. They alternate with intervals of coexistence of 0 and π states, with characteristic CPR similar to $I\sim\sin 2\phi$.

For experimental determination of the predicted superharmonic CPR, most pronounced at low temperature and for thin barriers ($d/\xi_0\leq 4$), phase-sensitive measurements are necessary. As in the experiments of Frolov *et al.*,[12] CPR could be obtained by including the junction in a rf SQUID configuration and by measuring the magnetic flux modulation due to an applied current in the absence of an external flux. Another manifestation of superharmonic CPR would be seen in the critical current modulation $I_c(H)$ by an external magnetic field.[27] However, in this case the periodicity of $\Phi_0/2$ would always be seen at high temperature, corresponding to both 0-π coexistence and the $\pi/2$ state, i.e., to $I\sim\pm\sin 2\phi$, respectively. At low temperature, for a suitable choice of Z, the appearance of triplet states with $I\sim\sin 4\varphi$ would be manifested as $\Phi_0/4$ periodicity in the $I_c(H)$ modulation.

Generally speaking, the nontrivial phase difference at the junction is due to the superconductor-ferromagnet proximity effect, which should be more pronounced in cleaner systems, where the oscillations of the pair-condensation amplitude are less damped by temperature than in the dirty case. On the other hand, interface nontransparency itself or the mismatch of Fermi wave vectors (FWVM) reduces the proximity effect.[11] However, this would change the above results only quantitatively.[13] In more realistic systems, lower interface

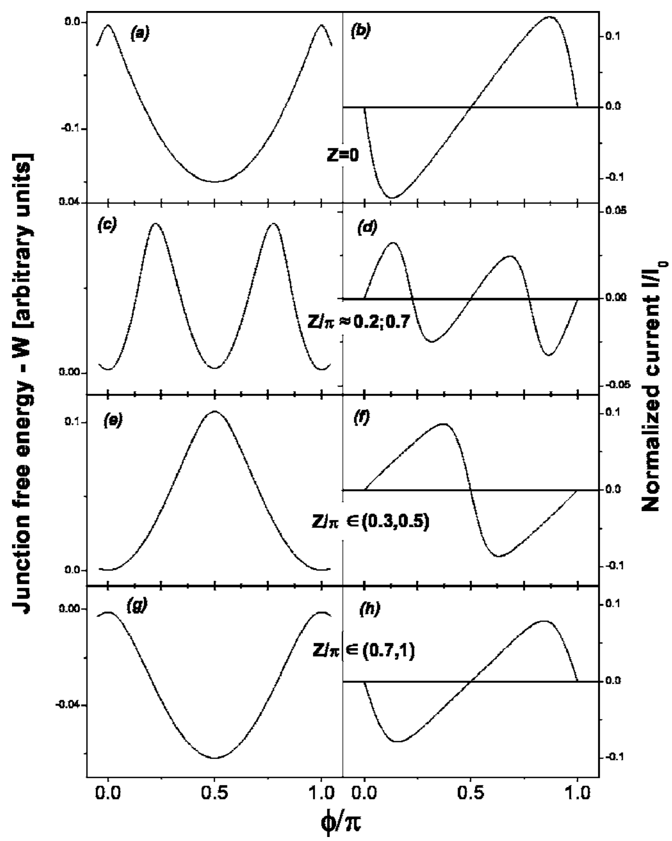

FIG. 4. Free energies and current-phase relations of junctions at low temperature $T=0.05T_c$ corresponding to stable states in Fig. 2 (see text). Note the unusual shapes of the CPR: (d) close to deformed $I(\phi)\sim\sin(4\phi)$ curve, (f) close to $I(\phi)\sim\sin(2\phi)$, and (h) close to $I(\phi)\sim\sin(2\phi+\pi)$.

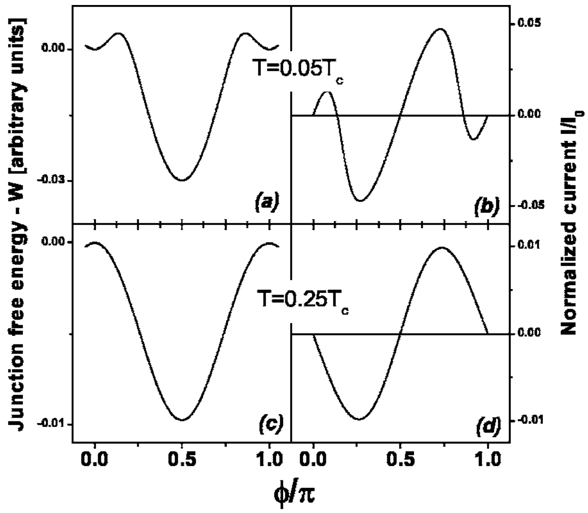

FIG. 5. Free energies and corresponding CPR for an example of coexistence of stable $\pi/2$ and metastable 0 and π states, $Z=0.1\pi$, at low and at higher temperatures.

transparency and FWVM could affect the positions of crossover between various equilibrium phases, and narrow or destroy the transition regions of coexistence of stable and metastable phases, as in the *s*-wave case.[13] However, we expect that the coexistence intervals of stable 0 and π phases should be visible. Thus, for comparison with experiments it would be desirable to have clean junctions with interfaces that are highly transparent, and with a low degree of disorder.[11,13,19] Progress in this direction has been made in 45° grain boundary YBCO junctions,[19] due to a special fabrication scheme. To obtain a high value of transparency in junctions with ferromagnetic barriers, a low roughness of the ferromagnet/superconductor interface and uniform concentration of ferromagnetic material along the interface are needed.[28] Transparency close to 1 was achieved in $Nb/Fe_{0.5}Si_{0.5}/Nb$.[29] For the F/D combination, promising systems are epitaxial $YBa_2Cu_3O_{7-\delta}/La_{2/3}Ca_{2/3}MnO_3$ structures.[4,5] However, understanding of the proximity effect in these systems requires further studies.[5]

In conclusion, the combined influence of *d*-wave symmetry in the electrodes and an exchange field in the barrier provides the possibility of Josephson junctions with equilibrium phases that include three-state (0-π-$\pi/2$) and two-state (0-π) coexistence. This could have a possible device application in quantum computing and for experimental study of the quantum superposition of macroscopically distinct states.[30]

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[1] T. Bauch *et al.*, Phys. Rev. Lett. **94**, 087003 (2005).
[2] T. Yamashita *et al.*, Phys. Rev. Lett. **95**, 097001 (2005); M. H. S. Amin *et al.*, Phys. Rev. B **71**, 064516 (2005).
[3] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005); I. F. Lyuksyutov and V. L. Pokrovsky, Adv. Phys. **54**, 67 (2005); E. Dagotto, Science **309**, 257 (2005).
[4] F. Perez *et al.*, Physica C **432**, 275 (2005); V. A. Vas'ko *et al.*, Phys. Rev. Lett. **78**, 1134 (1997); Z. Y. Chen *et al.*, Phys. Rev. B **63**, 212508 (2001).
[5] J. Stahn *et al.*, Phys. Rev. B **71**, 140509(R) (2005); Z. Sefrioui *et al.*, *ibid.* **67**, 214511 (2003); J. Chakhalian *et al.*, Nat. Phys. **2**, 244 (2006).
[6] M. Freamat and K. W. Ng, Physica C **400**, 1 (2003).
[7] J. X. Zhu, B. Friedman, and C. S. Ting, Phys. Rev. B **59**, 9558 (1999); S. Kashiwaya *et al.*, *ibid.* **60**, 3572 (1999); I. Žutić and O. T. Valls, *ibid.* **60**, 6320 (1999).
[8] Y. Tanaka and S. Kashiwaya, J. Phys. Soc. Jpn. **68**, 3485 (1999).
[9] A. I. Buzdin *et al.*, JETP Lett. **35**, 178 (1982).
[10] V. V. Ryazanov *et al.*, Physica C **341**, 1613 (2000); A. V. Veretennikov *et al.*, Physica B **284**, 495 (2000).
[11] A. A. Golubov *et al.*, Rev. Mod. Phys. **76**, 412 (2004).
[12] S. M. Frolov *et al.*, Phys. Rev. B **70**, 144505 (2004).
[13] Z. Radović, N. Lazarides, and N. Flytzanis, Phys. Rev. B **68**, 014501 (2003).
[14] D. J. Van Harlingen, Rev. Mod. Phys. **67**, 515 (1995).
[15] S. Yip, Phys. Rev. B **52**, 3087 (1995); M. Fogelstrom and S.-K. Yip, *ibid.* **57**, R14060 (1998).
[16] M. Fogelstrom *et al.*, Physica C **294**, 289 (1998).
[17] E. Il'ichev *et al.*, Phys. Rev. B **60**, 3096 (1999).
[18] C. W. Schneider *et al.*, Europhys. Lett. **68**, 86 (2004).
[19] T. Lindstrom *et al.*, Phys. Rev. Lett. **90**, 117002 (2003).
[20] R. Zikic, L. Dobrosavljević-Grujić, and Z. Radović, Phys. Rev. B **59**, 14644 (1999).
[21] Z. Radović, L. Dobrosavljević-Grujić, and B. Vujicic, Phys. Rev. B **63**, 214512 (2001).
[22] L. Dobrosavljević-Grujić and R. Zikic, Phys. Rev. B **68**, 092502 (2003).
[23] G. Eilenberger, Z. Phys. **214**, 195 (1968).
[24] M. J. M. de Jong and C. W. J. Beenakker, Phys. Rev. Lett. **74**, 1657 (1995).
[25] E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B **55**, 15174 (1997).
[26] K. Halterman and O. T. Valls, Phys. Rev. B **65**, 014509 (2001); J.-X. Zhu and C. S. Ting, Phys. Rev. B **61**, 1456 (2000).
[27] C. C. Tsuei and R. Kirtley, Rev. Mod. Phys. **72**, 969 (2000).
[28] M. Zareyan, W. Belzig, and Y. V. Nazarov, Phys. Rev. Lett. **86**, 308 (2001).
[29] O. Vavra *et al.*, Physica C **404**, 395 (2004).
[30] J. Q. You and F. Nori, Phys. Today **58** (11), 42 (2005).