

Odd Triplet Pairing in Clean Superconductor/Ferromagnet Heterostructures

Klaus Halterman,^{1,*} Paul H. Barsic,^{2,†} and Oriol T. Valls^{2,‡}

¹*Physics and Computational Sciences, Research and Engineering Sciences Department, Naval Air Warfare Center, China Lake, California 93555, USA*

²*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA*

(Received 4 April 2007; published 17 September 2007)

We study triplet pairing correlations in clean ferromagnet (F)/superconductor (S) nanojunctions, via fully self-consistent solution of the Bogoliubov–de Gennes equations. We consider FSF trilayers, with S being an s -wave superconductor, and an arbitrary angle α between the magnetizations of the two F layers. We find that contrary to some previous expectations, triplet correlations, odd in time, are induced in both the S and F layers in the clean limit. We investigate their behavior as a function of time, position, and α . The triplet amplitudes are largest at times on the order of the inverse Debye frequency, and at that time scale they are long-ranged in both S and F . The zero temperature condensation energy is found to be lowest when the magnetizations are antiparallel.

DOI: [10.1103/PhysRevLett.99.127002](https://doi.org/10.1103/PhysRevLett.99.127002)

PACS numbers: 74.45.+c, 74.25.Bt, 74.78.Fk

The proximity effects in superconductor/ferromagnet (S/F) heterostructures lead to the coexistence of ferromagnetic and superconducting ordering and to novel transport phenomena [1,2]. Interesting effects that arise from the interplay between these orderings have potential technological applications in fields such as spintronics [3]. For example, the relative orientation of the magnetizations in the F layers in FSF trilayers can have a strong influence on the conductivity [4–8], making them good spin valve candidates. Such trilayers were first proposed [9] for insulating F layers and later for metallic [10,11] ones.

This interplay also results in fundamental new physics. An outstanding example is the existence of “odd” triplet superconductivity. This is an s -wave pairing triplet state that is even in momentum, and therefore not destroyed by nonmagnetic impurities, but with the triplet correlations being odd in frequency, so that the equal time triplet amplitudes vanish as required by the Pauli principle. This exotic pairing state with total spin one was proposed long ago [12] as a possible state in superfluid ^3He . Although this type of pairing does not occur there, it is possible in certain FSF systems [1,2,13,14] with ordinary singlet pairing in S . This arrangement can induce, via proximity effects, triplet correlations with $m = 0$ and $m = \pm 1$ projections of the total spin. If the magnetization orientations in both F layers are unidirectional and along the quantization axis, symmetry arguments show that only the $m = 0$ projection along that axis can exist.

Odd triplet pairing in F/S structures has been studied in the dirty limit through linearized Usadel-type quasiclassical equations [2,13–16]. In this case, it was found that $m = 0$ triplet pairs always exist. They are suppressed in F over short length scales, just as the singlet pairs. The $m = \pm 1$ components, for which the exchange field is not pair-breaking, can be long-ranged, and were found to exist for nonhomogeneous magnetization. For FSF trilayers [2,17,18], the quasiclassical methods predict that the struc-

ture contains a superposition of all three spin triplet projections except when the magnetizations of the F layers are collinear, in which case the $m = \pm 1$ components along the magnetization axis vanish. It is noted in Ref. [1] that the existence of such effects in the clean limit has not been established and may be doubted. This we remedy in the present work, where we establish that, contrary to the doubts voiced there, induced, long-ranged, odd triplet pairing does occur in clean FSF structures.

Experimental results that may argue for the existence of long range triplet pairing of superconductors through a ferromagnet have been obtained in superlattices [19] with ferromagnetic spacers and in two superconductors coupling through a single ferromagnet [20,21]. Measurements [20] on a SQUID, in which a phase change of π in the order parameter is found after inversion, indicate an odd-parity state. Very recently, a Josephson current through a strong ferromagnet was observed, indicating the existence of a spin triplet state [21] induced by NbTiN, an s -wave superconductor.

In this paper, we study the induced odd triplet superconductivity in FSF trilayers in the clean limit through a fully self-consistent solution of the microscopic Bogoliubov–de Gennes (BdG) equations. We consider arbitrary relative orientation of the magnetic moments in the two F layers. We find that there are indeed induced odd triplet correlations which can include both $m = 0$ and $m = \pm 1$ projections. We directly study their time dependence and we find that they are largest for times of order of the inverse cutoff Debye frequency. The correlations are, at these time scales, long-ranged in both the S and F regions. We also find that the condensation energy depends on the relative orientation of the F layers, being a minimum when they are antiparallel.

To find the triplet correlations arising from the nontrivial spin structure in our FSF system, we use the BdG equations with the BCS Hamiltonian, \mathcal{H}_{eff} :

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & \int d^3r \left[\sum_{\delta} \psi_{\delta}^{\dagger}(\mathbf{r}) \left[-\frac{\nabla^2}{2m^*} - E_F \right] \psi_{\delta}(\mathbf{r}) \right. \\ & + \frac{1}{2} \left[\sum_{\delta, \beta} (i\sigma_y)_{\delta\beta} \Delta(\mathbf{r}) \psi_{\delta}^{\dagger}(\mathbf{r}) \psi_{\beta}^{\dagger}(\mathbf{r}) + \text{h.c.} \right] \\ & \left. - \sum_{\delta, \beta} \psi_{\delta}^{\dagger}(\mathbf{r}) (\mathbf{h} \cdot \boldsymbol{\sigma})_{\delta\beta} \psi_{\beta}(\mathbf{r}) \right], \end{aligned}$$

where $\Delta(\mathbf{r})$ is the pair potential, to be determined self-consistently; ψ_{δ}^{\dagger} , ψ_{δ} are the creation and annihilation operators with spin δ ; E_F is the Fermi energy; and $\boldsymbol{\sigma}$ are the Pauli matrices. We describe the magnetism of the F layers by an effective exchange field $\mathbf{h}(\mathbf{r})$ that vanishes in the S layer. We will consider the geometry depicted in Fig. 1, with the y axis normal to the layers and $\mathbf{h}(\mathbf{r})$ in the x - z plane (which is infinite in extent) forming an angle $\pm\alpha/2$ with the z axis in each F layer.

Next, we expand the field operators in terms of a Bogoliubov transformation, which we write as

$$\psi_{\delta}(\mathbf{r}) = \sum_n [u_{n\delta}(\mathbf{r})\gamma_n + \eta_{\delta}v_{n\delta}(\mathbf{r})\gamma_n^{\dagger}], \quad (1)$$

where $\eta_{\delta} \equiv 1(-1)$ for spin down (up), $u_{n\delta}$ and $v_{n\delta}$ are the quasiparticle and quasihole amplitudes, and the operators γ_n , γ_n^{\dagger} the usual Bogoliubov operators. This transformation diagonalizes \mathcal{H}_{eff} : $[\mathcal{H}_{\text{eff}}, \gamma_n] = -\epsilon_n\gamma_n$, $[\mathcal{H}_{\text{eff}}, \gamma_n^{\dagger}] = \epsilon_n\gamma_n^{\dagger}$. By taking the commutator $[\psi_{\delta}(\mathbf{r}), \mathcal{H}_{\text{eff}}]$, and with $\mathbf{h}(\mathbf{r})$ in the x - z plane as explained above, we have the following:

$$[\psi_{\uparrow}(\mathbf{r}), \mathcal{H}_{\text{eff}}] = (\mathcal{H}_e - h_z)\psi_{\uparrow}(\mathbf{r}) - h_x\psi_{\uparrow}(\mathbf{r}) + \Delta(\mathbf{r})\psi_{\uparrow}^{\dagger}(\mathbf{r}), \quad (2a)$$

$$[\psi_{\downarrow}(\mathbf{r}), \mathcal{H}_{\text{eff}}] = (\mathcal{H}_e + h_z)\psi_{\downarrow}(\mathbf{r}) - h_x\psi_{\downarrow}(\mathbf{r}) - \Delta(\mathbf{r})\psi_{\downarrow}^{\dagger}(\mathbf{r}). \quad (2b)$$

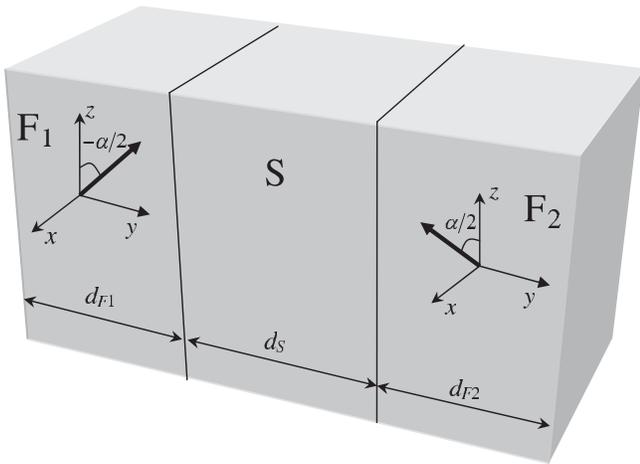


FIG. 1. Schematic of FSF junction. The left ferromagnetic layer F_1 has a magnetization oriented at an angle $-\alpha/2$ in the x - z plane, while the other ferromagnet, F_2 , has a magnetization orientation at an angle $\alpha/2$ in the x - z plane.

Inserting (1) into (2) and introducing a set $\boldsymbol{\rho}$ of Pauli-like matrices in particle-hole space yields the spin-dependent BdG equations:

$$[\rho_z \otimes (\mathcal{H}_0 \hat{\mathbf{1}} - h_z \sigma_z) + (\Delta(y)\rho_x - h_x \hat{\mathbf{1}}) \otimes \sigma_x] \Phi_n = \epsilon_n \Phi_n, \quad (3)$$

where $\Phi_n \equiv [u_{n\uparrow}(y), u_{n\downarrow}(y), v_{n\uparrow}(y), v_{n\downarrow}(y)]^T$ and $\mathcal{H}_0 \equiv -\partial_y^2/(2m) + \epsilon_{\perp} - E_F$. Here ϵ_{\perp} is the transverse kinetic energy and a factor of $e^{i\mathbf{k}_{\perp} \cdot \mathbf{r}}$ has been suppressed. In deriving Eqs. (3) care has been taken to consistently use the phase conventions in Eqs. (1). To find the quasiparticle amplitudes along a different quantization axis in the x - z plane, one performs a spin rotation: $\Phi_n \rightarrow \hat{U}(\alpha')\Phi_n$, where $\hat{U}(\alpha') = \cos(\alpha'/2)\hat{\mathbf{1}} \otimes \hat{\mathbf{1}} - i\sin(\alpha'/2)\rho_z \otimes \sigma_y$.

When the magnetizations of the F layers are collinear, one can take $h_x = 0$. For the general case shown in Fig. 1 one has in the F_1 layer $h_x = h_0 \sin(-\alpha/2)$ and $h_z = h_0 \cos(-\alpha/2)$, where h_0 is the magnitude of \mathbf{h} , while in F_2 , $h_x = h_0 \sin(\alpha/2)$, and $h_z = h_0 \cos(\alpha/2)$. With an appropriate choice of basis, Eqs. (3) are cast into a matrix eigenvalue system that is solved iteratively with the self-consistency condition, $\Delta(y) = g(y)f_3$ ($f_3 = \frac{1}{2}[\langle\psi_{\uparrow}(\mathbf{r})\psi_{\downarrow}(\mathbf{r})\rangle - \langle\psi_{\downarrow}(\mathbf{r})\psi_{\uparrow}(\mathbf{r})\rangle]$). In the F layers we have $g(y) = 0$, while in S , $g(y) = g$, g being the usual BCS singlet coupling constant there. Through Eqs. (1), the self-consistency condition becomes a sum over states restricted by the factor g to within ω_D from the Fermi surface. Iteration is performed until self-consistency is reached. The numerical process is the same that was used in previous work [22,23], with now the h_x term requiring larger four-component matrices to be diagonalized.

We now define the following time dependent triplet amplitude functions in terms of the field operators:

$$\tilde{f}_0(\mathbf{r}, t) = \frac{1}{2} [\langle\psi_{\uparrow}(\mathbf{r}, t)\psi_{\downarrow}(\mathbf{r}, 0)\rangle + \langle\psi_{\downarrow}(\mathbf{r}, t)\psi_{\uparrow}(\mathbf{r}, 0)\rangle], \quad (4a)$$

$$\tilde{f}_1(\mathbf{r}, t) = \frac{1}{2} [\langle\psi_{\uparrow}(\mathbf{r}, t)\psi_{\uparrow}(\mathbf{r}, 0)\rangle - \langle\psi_{\downarrow}(\mathbf{r}, t)\psi_{\downarrow}(\mathbf{r}, 0)\rangle], \quad (4b)$$

which, as required by the Pauli principle for these s -wave amplitudes, vanish at $t = 0$, as we shall verify. Making use of Eqs. (1) and the commutators, one can derive and formally integrate the Heisenberg equations of the motion for the operators and obtain

$$\tilde{f}_0(y, t) = \frac{1}{2} \sum_n [u_{n\uparrow}(y)v_{n\downarrow}(y) - u_{n\downarrow}(y)v_{n\uparrow}(y)]\zeta_n(t), \quad (5a)$$

$$\tilde{f}_1(y, t) = -\frac{1}{2} \sum_n [u_{n\uparrow}(y)v_{n\uparrow}(y) + u_{n\downarrow}(y)v_{n\downarrow}(y)]\zeta_n(t), \quad (5b)$$

where $\zeta_n(t) \equiv \cos(\epsilon_n t) - i\sin(\epsilon_n t) \tanh(\epsilon_n/2T)$.

The amplitudes in Eqs. (5) contain all information on the space and time dependence of induced triplet correlations throughout the FSF structure. The summations in Eqs. (5) are over the entire self-consistent spectrum, ensuring that f_0 and f_1 vanish identically at $t = 0$ and thus obey the

exclusion principle. Using a non-self-consistent $\Delta(y)$ leads to violations of this condition, particularly near the interface where proximity effects are most pronounced. Geometrically, the indirect coupling between magnets is stronger with fairly thin S layers and relatively thick F layers. We thus have chosen $d_S = (3/2)\xi_0$ and $d_{F1} = d_{F2} = \xi_0$, with the BCS correlation length $\xi_0 = 100k_F^{-1}$. We consider the low T limit and take $\omega_D = 0.04E_F$. The magnetic exchange is parametrized via $I \equiv h_0/E_F$. Results shown are for $I = 0.5$ (unless otherwise noted) and the magnetization orientation angle, α , is swept over the range $0 \leq \alpha \leq \pi$. No triplet amplitudes arise in the absence of magnetism ($I = 0$).

For the time scales considered here, the imaginary parts of $\tilde{f}_0(y, t)$ and $\tilde{f}_1(y, t)$ at $t \neq 0$ are considerably smaller than their real parts, and thus we focus on the latter, which we denote by $f_0(y, t)$ and $f_1(y, t)$. In Fig. 2, the spatial dependence of f_0 is shown for parallel magnetization directions ($\alpha = 0$) at several times $\tau \equiv \omega_D t$. The spatial range shown includes part of the F_1 layer (to the left of the dashed line) and half of the S layer (to the right). At finite τ , the maximum occurs in the ferromagnet close to the interface, after which f_0 undergoes damped oscillations with the usual spatial length scale $\xi_f \approx (k_{F1} - k_F)^{-1} \approx k_F^{-1}/I$. The height of the main peak first increases with time, but drops off after a characteristic time, $\tau_c \approx 4$, as seen in the inset, which depicts the maximum value of f_0 as a function of τ . As τ increases beyond τ_c , the modulating f_0 in F develops more complicated atomic scale interference patterns and becomes considerably longer ranged.

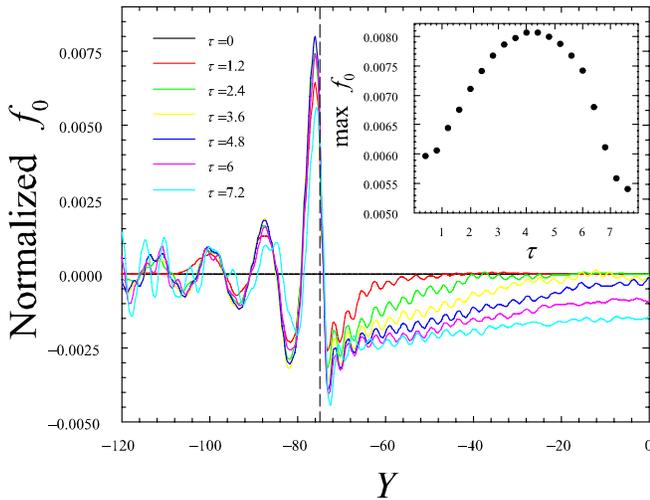


FIG. 2 (color online). The real part, f_0 , of the triplet amplitude \tilde{f}_0 , for a FSF trilayer at 7 different times. We normalize f_0 by the singlet bulk pair amplitude, Δ_0/g . The coordinate y is scaled by the Fermi wave vector, $Y \equiv k_F y$, and time by the Debye frequency, $\tau \equiv \omega_D t$. At $\tau = 0$, $f_0 \equiv 0$ as required by the Pauli principle. The interface is marked by the vertical dashed line, with an F region to the left and the S to the right. Half of the S region and part of the left F layer are shown. The inset shows the maximum value of f_0 versus τ .

S , we see immediately that f_0 is also larger near the interface. Since the triplet amplitudes vanish at $\tau = 0$, short time scales exhibit correspondingly short triplet penetration. The figure shows, however, that the value of f_0 in S is substantial for $\tau \gtrsim \tau_c$, extending over length scales on the order of ξ_0 without appreciable decay. In contrast, the usual singlet correlations were found to monotonically drop off from their $\tau = 0$ value over τ scales of order unity.

In the main plot of Fig. 3 we examine the spatial dependence of the real part of the $m = \pm 1$ triplet amplitude, f_1 . Normalizations and spatial ranges are as in Fig. 2, but now the time is fixed at $\tau = 4 \approx \tau_c$, and five equally spaced magnetization orientations are considered. At $\alpha = 0$, f_1 vanishes identically at all τ , as expected. For nonzero α , correlations in all triplet channels are present. As was found for f_0 , the plot clearly shows that f_1 is largest near the interface, in the F region. Our geometry and conventions imply (see Fig. 1) that the magnetization has opposite x components in the F_1 and F_2 regions. The f_1 triplet pair amplitude profile is thus antisymmetric about the origin, in contrast to the symmetric f_0 , implying the existence of one node in the superconductor. Nevertheless, the penetration of the f_1 correlations in S can be long-ranged. We find that f_1 and f_0 oscillate in phase and with the same wavelength, regardless of α . The inset illustrates the maximum attained values of f_0 and f_1 in F_1 as α varies. It shows that for a broad range of α , $\alpha \lesssim 3\pi/4$, the maximum of f_0 varies relatively little, after which it drops off rapidly to zero at $\alpha = \pi$. This is to be expected as the antiparallel orientation corresponds to the case in which the magnetization is in the x direction, which is perpendicular to the axis of quantization (see Fig. 1). The rise in the maximum of f_1 is monotonic, cresting at $\alpha = \pi$, consistent with the main plot. At this angle the triplet correlations extend considerably into the superconductor. At $\alpha = \pi/2$ the maxima coincide since the two triplet components are then identical

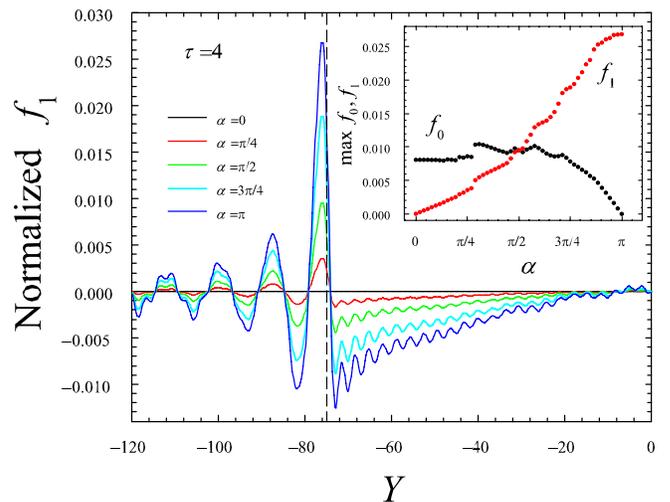


FIG. 3 (color online). Spatial and angular dependence of f_1 , at $\tau = 4 \approx \tau_c$ and several α . Normalizations and ranges are as in Fig. 2. Inset: maxima of f_0 and f_1 in F_1 versus α .

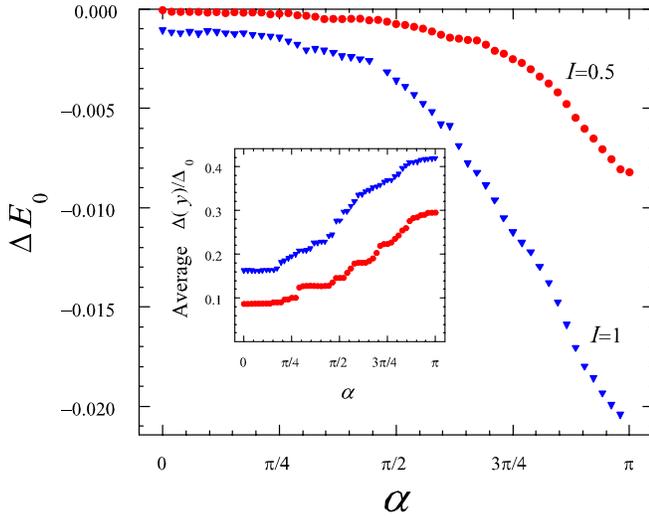


FIG. 4 (color online). The $T = 0$ condensation energy, ΔE_0 , normalized by $N(0)\Delta_0^2$ [$N(0)$ is the usual density of states], vs the angle α for two values of I . When the two magnetizations are antiparallel ($\alpha = \pi$) ΔE_0 is lowest. The inset shows the ordinary (singlet) pair potential averaged over the S region, normalized to the bulk Δ_0 .

throughout the whole space because the magnetization vectors have equal projections on the x and z axes. At $\alpha = \pi$ both magnetizations are normal to the axis of quantization z (see Fig. 1). By making use of the rotation matrix \hat{U} [see below Eqs. (3)] one can verify that the $m = \pm 1$ components with respect to the axis x along the magnetizations are zero.

We next consider the condensation energy, ΔE_0 , calculated by subtracting the zero temperature superconducting and normal state free energies. The calculation uses the self-consistent spectra and $\Delta(y)$ and methods explained elsewhere [23,24]. In the main plot of Fig. 4, we show ΔE_0 (normalized at twice its bulk S value) at two different values of I . The condensation energy results clearly demonstrate that the antiparallel state ($\alpha = \pi$) is in general the lowest energy ground state. These results are consistent with previous studies [8] of FSF structures with parallel and antiparallel magnetizations. The inset contains the magnitude of the spatially averaged pair potential, normalized by Δ_0 , at the same values of I . The inset correlates with the main plot, as it shows that the singlet superconducting correlations in S increase with α and are larger at $I = 1$ than at $I = 0.5$. The half-metallic case of $I = 1$ illustrates that by having a single spin band populated at the Fermi surface, Andreev reflection is suppressed, in effect keeping the superconductivity more contained within S .

Thus, we have shown that in clean FSF trilayers induced odd triplet correlations, with $m = 0$ and $m = \pm 1$ projections of the total spin, exist. We have used a microscopic

self-consistent method to study the time and angular dependence of these triplet correlations. The correlations in all 3 triplet channels were found, at times $\tau \equiv \omega_D t \gtrsim \tau_c$, where $\tau_c \approx 4$, to be long-ranged in both the F and S regions. Finally, a study of the condensation energy revealed that the ground state energy is always lowest for antiparallel magnetizations.

This project was supported in part by a grant of HPC resources from the ARSC at the University of Alaska Fairbanks (part of the DoD HPCM program) and by the University of Minnesota Graduate School.

*klaus.halterman@navy.mil

†barsic@physics.umn.edu

‡Also at Minnesota Supercomputer Institute, University of Minnesota, Minneapolis, MN 55455, USA.

otvalls@umn.edu

- [1] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).
- [2] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Rev. Mod. Phys. **77**, 1321 (2005).
- [3] Igor Žutić, Jaroslav Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).
- [4] J. Y. Gu *et al.*, Phys. Rev. Lett. **89**, 267001 (2002).
- [5] I. C. Moraru, W. P. Pratt, and N. O. Birge, Phys. Rev. Lett. **96**, 037004 (2006).
- [6] C. Bell, S. Turşucu, and J. Aarts, Phys. Rev. B **74**, 214520 (2006).
- [7] C. Visani *et al.*, Phys. Rev. B **75**, 054501 (2007).
- [8] K. Halterman and O. T. Valls, Phys. Rev. B **72**, 060514(R) (2005).
- [9] P. G. de Gennes, Phys. Lett. **23**, 10 (1966).
- [10] L. R. Tagirov, Phys. Rev. Lett. **83**, 2058 (1999).
- [11] A. I. Buzdin, A. V. Vdyayev, and N. V. Ryzhanova, Europhys. Lett. **48**, 686 (1999).
- [12] V. L. Berezinskii, Pis'ma Zh. Eksp. Teor. Fiz. **20**, 628 (1974) [JETP Lett. **20**, 287 (1974)].
- [13] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. Lett. **86**, 3140 (2001).
- [14] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B **68**, 064513 (2003).
- [15] T. Champel and M. Eschrig, Phys. Rev. B **72**, 054523 (2005).
- [16] A. F. Volkov, F. S. Bergeret, and K. B. Efetov, Phys. Rev. Lett. **90**, 117006 (2003).
- [17] T. Löfwander *et al.*, Phys. Rev. Lett. **95**, 187003 (2005).
- [18] Ya. V. Fominov, A. A. Golubov, and M. Yu. Kupriyanov, JETP Lett. **77**, 510 (2003).
- [19] V. Peña *et al.*, Phys. Rev. B **69**, 224502 (2004).
- [20] K. D. Nelson *et al.*, Science **306**, 1151 (2004).
- [21] R. S. Keizer *et al.*, Nature (London) **439**, 825 (2006).
- [22] K. Halterman and O. T. Valls, Phys. Rev. B **69**, 014517 (2004).
- [23] K. Halterman and O. T. Valls, Phys. Rev. B **70**, 104516 (2004).
- [24] I. Kosztin *et al.*, Phys. Rev. B **58**, 9365 (1998).