Pairing Symmetry Conversion by Spin-Active Interfaces in Magnetic Normal-Metal–Superconductor Junctions

Jacob Linder,[1] Takehito Yokoyama,[2] Asle Sudbø,[1] and Matthias Eschrig[3]

[1]*Department of Physics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway*
[2]*Department of Applied Physics, Nagoya University, Nagoya, 464-8603, Japan*
[3]*Institut für Theoretische Festkörperphysik and DFG-Center for Functional Nanostructures, Universität Karlsruhe, D-76128 Karlsruhe, Germany*

(Received 16 September 2008; published 13 March 2009)

We study the proximity-induced superconducting correlations in a normal metal connected to a superconductor when the interface between them is spin active and the normal metal is ballistic or diffusive. Remarkably, for any interface spin polarization there is a critical interface resistance, above which the conventional even-frequency proximity component vanishes completely at the chemical potential, while the odd-frequency component remains finite. We propose a way to unambiguously observe the odd-frequency component.

DOI: 10.1103/PhysRevLett.102.107008 PACS numbers: 74.20.Rp, 74.50.+r

Superconductivity and superfluidity are hallmarks of the wavelike character of matter and manifest themselves in vastly different systems, from ultracold dilute gases via cold metals and fluids to extremely dense protonic and neutronic matter. In all of these contexts, the symmetry of the order parameter is of profound importance. Over the past decades, the possibility of superconducting order parameters that change sign under a *time-coordinate* exchange of the two fermions comprising the Cooper pair has emerged in addition to the by now well studied varieties of orbital symmetries [1–5]. This so-called odd-frequency superconductivity [6] is distinct from the traditional even-frequency pairing in the Bardeen-Cooper-Schrieffer paradigm and may be induced by proximity effects in hybrid structures of superconductors and magnets [1].

In a broader context, proximity systems offer the possibility of controlling the physics of competing broken symmetries. The fundamental heterostructure for studying proximity-induced superconductivity is the superconductor–normal-metal ($S|N$) bilayer, where the normal metal or the interface may have magnetic properties. Among possible triplet pair correlations, in the diffusive limit odd-frequency pairs are favored [7], whereas in ballistic hybrid systems both odd- and even-frequency amplitudes compete [3,4]. As all known superconductors to date exhibit an even-frequency order parameter, the observation of proximity-induced effects that are particular to odd-frequency pairing would be of utmost interest.

There are two major difficulties associated with the detection of the odd-frequency state in superconductor-ferromagnet ($S|F$) bilayers. One is the usually short penetration depth into the ferromagnetic region, limited by the magnetic coherence length ξ_F, much less than the superconducting coherence length ξ_S [1]. Another problem is that odd-frequency pairs are only well defined when even-frequency correlations vanish in the ferromagnet. Clear-cut signatures of the former are therefore accessible only in a limited parameter regime [8].

The majority of work on superconducting proximity structures so far has been restricted to the diffusive limit and spin-inactive interfaces [9]. For a nonmagnetic bilayer, a minigap appears in the density of states of the normal metal. It scales with the Thouless energy of the normal layer and with the transmission probability of the interface. Such minigap structures are readily accessible experimentally [10]. For a spin-active interface, the transmission properties of spin-$\uparrow$ and spin-$\downarrow$ electrons into a metal are different, and this gives rise to both spin-dependent conductivities and spin-dependent phase shifts at the interface [11–15]. In this Letter, we show that a spin-active interface in an $S|N$ bilayer produces clear signatures of purely odd-frequency triplet pairing amplitudes that can be tested experimentally.

We consider the system shown in Fig. 1. The superconductor is conventional (even-frequency s-wave), while the interface is magnetic. We find that there is a dramatic change in the nature of proximity correlations when the spin-dependent phase shifts exceed the tunneling probability of the interface. The spin-active interface in an $S|N$ bilayer causes the even-frequency correlations to vanish at zero excitation energy, while odd-frequency correlations appear. At the same time, the minigap, one of the hallmarks

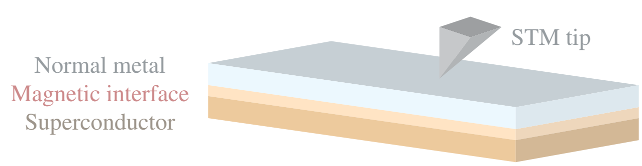

FIG. 1 (color online). Proposed experiment for observation of the odd-frequency component in a diffusive $N|S$ junction.

of the conventional proximity effect, is replaced by a low-energy band with an enhanced density of states. We focus on the density of states (DOS) in the normal region, which can be probed by tunneling experiments. Our findings suggest that it should be possible to detect the odd-frequency amplitude without any interfering effects of even-frequency correlations. Since the exchange field is absent in the normal metal, this resolves the two main difficulties associated with the experimental detection of odd-frequency correlations mentioned above.

We adopt the quasiclassical theory of superconductivity [16], where information about the physical properties of the system is embedded in the Green's function. For equilibrium situations, it suffices to consider the retarded Green's function $\hat{g}$ that is parameterized conveniently in the normal (N) region by a parameter θ_σ, allowing for both singlet and triplet correlations [8]. In the superconducting (S) region, we employ the bulk solution $\hat{g}_S = c\underline{\tau}_3 \otimes \underline{\sigma}_0 + s\underline{\tau}_1 \otimes (i\underline{\sigma}_2)$, with $c = \cosh(\theta)$, $s = \sinh(\theta)$, and $\theta = \text{arctanh}(\Delta/\varepsilon)$, $\underline{\tau}_i$ and $\underline{\sigma}_i$ being Pauli matrices in particle-hole and spin space, respectively.

We use the formalism described in Ref. [8] and consider first the diffusive limit. Then the orbital symmetry for all proximity amplitudes is reduced to s-wave, and hence the singlet component always has an even-frequency symmetry while the triplet component has an odd-frequency symmetry. The Green's functions are subject to boundary conditions, which assume at the $S|N$ interface in the tunneling limit the form [13,15] $2\gamma d\hat{g}_N\partial_x\hat{g}_N = [\hat{g}_S, \hat{g}_N] + i(G_\phi/G_T)[\underline{\tau}_0 \otimes \underline{\sigma}_3, \hat{g}_N]$ and at the outer surface read $\partial_x\hat{g}_N = \hat{0}$. Here $\gamma = R_B/R_N$, where R_B (R_N) is the resistance of the barrier (normal region), and d is the width of the normal region, while G_T is the junction conductance in the normal state. The boundary condition above contains an additional term G_ϕ compared to the usual nonmagnetic boundary conditions in Ref. [9]. This term is due to spin-dependent phase shifts of quasiparticles being reflected at the interface. G_ϕ may be nonzero even if the transmission $G_T \to 0$, corresponding to a ferromagnetic insulator [13]. We define the superconducting coherence length $\xi_S = \sqrt{D/\Delta}$ and Thouless energy $\varepsilon_{\text{Th}} = D/d^2$, where D is the diffusion constant, and assume that the inelastic scattering length l_{in} is sufficiently large, such that $d \ll l_{\text{in}}$.

The Usadel equation [17] reads $D\partial_x^2\theta_\sigma + 2i\varepsilon\sinh\theta_\sigma = 0$, with boundary condition $\gamma d\partial_x\theta_\sigma = (cs_\sigma - \sigma sc_\sigma) + i\sigma s_\sigma \frac{G_\phi}{G_T}$ at $x = 0$ and $\partial_x\theta_\sigma = 0$ at $x = d$. Here $c_\sigma = \cosh(\theta_\sigma)$ and $s_\sigma = \sinh(\theta_\sigma)$. For $\varepsilon = 0$ we find pairing amplitudes that are either purely (odd-frequency) triplet for $|G_\phi| > G_T$,

$$f_s(0) = 0, \qquad f_t(0) = G_T\text{sgn}(G_\phi)/\sqrt{G_\phi^2 - G_T{}^2}, \tag{1}$$

or purely (even-frequency) singlet for $|G_\phi| < G_T$,

$$f_s(0) = iG_T/\sqrt{G_T{}^2 - G_\phi^2}, \qquad f_t(0) = 0. \tag{2}$$

Thus, the presence of G_ϕ induces an odd-frequency component in the normal layer. The remarkable aspect of Eqs. (1) and (2) is that they are valid for any value of the width d below the inelastic scattering length and for any interface parameter γ. Thus, the vanishing of the singlet component is a robust feature in $S|N$ structures with spin-active interfaces, as long as $|G_\phi| > G_T$. Without loss of generality, we focus on positive values of G_ϕ from now on.

The DOS is given as $N(\varepsilon)/N_0 = \sum_\sigma \text{Re}\{c_\sigma\}/2$, yielding $N(0)/N_0 = \text{Re}\{G_\phi/\sqrt{G_\phi^2 - G_T{}^2}\}$. At zero energy, the DOS vanishes when $G_\phi < G_T$, which means that the usual minigap in $S|N$ structures survives. However, the zero-energy DOS is enhanced for $G_\phi > G_T$ since the singlet component vanishes there.

The full energy dependence of the DOS may be obtained only numerically. To model a realistic experimental setup, we fix $\gamma = 10$ and $d/\xi_S = 1.0$, although our qualitative results are independent of these particular choices. As a measure of the relevant energy scale, we define $\varepsilon_0 = \varepsilon_{\text{Th}}/(2\gamma)$. The results are shown in Fig. 2 to investigate the effect of the spin-dependent phase shifts. The low-energy DOS is strongly enhanced due to the odd-frequency amplitude when $G_\phi/G_T > 1$ ($G_\phi/G_T = 1.5$ in the figure). Conversely, the DOS develops a minigap around $\varepsilon = 0$ when $G_\phi/G_T < 1$ ($G_\phi/G_T = 0.5$ in the figure). The ratio G_ϕ/G_T depends on the microscopic barrier properties [15]. In the tunneling limit, one finds that G_ϕ can be considerably larger than G_T.

We suggest the following qualitative explanation for the mechanism behind the separation between even- and odd-frequency correlations. The superconductor induces a minigap $\propto G_T$ in the normal metal, while the spin-active barrier induces an effective exchange field $\propto G_\phi$. The situation in the normal metal then resembles that of a thin-film conventional superconductor in the presence of an in-plane external magnetic field [18], with the role of the gap and field played by G_T and G_ϕ, respectively. In that

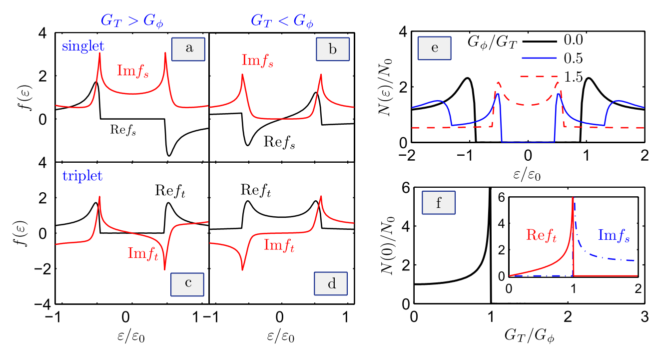

FIG. 2 (color online). The singlet and triplet proximity amplitudes induced in the normal metal are shown for $G_\phi/G_T < 1$ [in (a) and (c)] and $G_\phi/G_T > 1$ [in (b) and (d)]. In (e), we plot the energy-resolved DOS for several values of G_ϕ/G_T. Finally, (f) shows the zero-energy DOS as a function of G_T/G_ϕ, with the proximity amplitudes shown in the inset.

case, it is known that superconductivity is destroyed above the Clogston-Chandrasekhar limit [19], as the spin-singlet Cooper pairs break up. In the present case, we observe coexistence of the exchange field and spin-singlet even-frequency superconductivity as long as G_ϕ is below the critical value of $G_\phi = G_T$. At the critical point, the DOS varies as $1/\sqrt{|\epsilon|}$ and diverges at $\epsilon = 0$. However, for $G_\phi > G_T$ spin-singlet pairing is no longer possible at the chemical potential. It is then replaced by spin-triplet pairing, which must be odd in frequency due to the isotropization of the gap in the diffusive limit. Thus, there is a natural separation between even-frequency and odd-frequency pairing in the normal metal at a critical value of the effective exchange field G_ϕ.

The same effect occurs in the ballistic limit, as we now show. In this case, we can obtain the retarded Green's function using the formalism described in Refs. [14,20]. The Eilenberger equation in the normal region reads $iv_{Fx}\partial_x\hat{g} + [\varepsilon\underline{\tau}_3 \otimes \underline{\sigma}_0, \hat{g}] = \hat{0}$. For the boundary conditions, we use a scattering matrix describing the magnetic interface between the superconductor and the normal metal

$$\hat{S} = \begin{pmatrix} \underline{r}_S \cdot \exp(\frac{i}{2}\vartheta_S\underline{\sigma}_3) & \underline{t}_{SN} \cdot \exp(\frac{i}{2}\vartheta_{SN}\underline{\sigma}_3) \\ \underline{t}_{NS} \cdot \exp(\frac{i}{2}\vartheta_{NS}\underline{\sigma}_3) & -\underline{r}_N \cdot \exp(\frac{i}{2}\vartheta_N\underline{\sigma}_3) \end{pmatrix}, \quad (3)$$

with real reflection and transmission spin matrices $\underline{r}_S$, $\underline{r}_N$, $\underline{t}_{SN}$, and $\underline{t}_{NS}$. The spin mixing angles ϑ_S, ϑ_N, ϑ_{SN}, and ϑ_{NS} describe spin-dependent scattering phases [11]. Neglecting spin flip scattering, the transmission and reflection amplitudes are diagonal in spin space, and the relations $\underline{r}_S = \underline{r}_N \equiv \mathrm{diag}[r_\uparrow, r_\downarrow]$, $\underline{t}_{NS} = \underline{t}_{SN} \equiv \mathrm{diag}[t_\uparrow, t_\downarrow]$, $r_\uparrow^2 + t_\uparrow^2 = r_\downarrow^2 + t_\downarrow^2 = 1$, and $\vartheta_{NS} + \vartheta_{SN} = \vartheta_S + \vartheta_N$ follow from the unitarity of $\hat{S}$. Possible scalar phases are omitted in Eq. (3), as they play no role in the final results.

We next concentrate on subgap energies. The anomalous amplitudes can be decomposed into singlet and triplet components: $f = (f_s + f_t\underline{\sigma}_3)(i\underline{\sigma}_2)$. Defining $f_\sigma = (f_s + \sigma f_t)/2$, we obtain on the top of the normal overlayer ($x = d$) $f_\sigma(\varepsilon) = -\mathrm{sgn}(\alpha_\sigma)t_\uparrow t_\downarrow/\sqrt{\alpha_\sigma^2 - (t_\uparrow t_\downarrow)^2}$, with $\alpha_\sigma = \sin(2\varepsilon d/v_{Fx} + \vartheta_{\sigma+}) + r_\uparrow r_\downarrow \sin(2\varepsilon d/v_{Fx} + \vartheta_{\sigma-})$. Here $\vartheta_{\sigma\pm} = \frac{\sigma}{2}(\vartheta_N \pm \vartheta_S) \pm \arcsin(\varepsilon/\Delta)$, and ε has to be supplemented by an infinitesimally small positive imaginary part. The interface parameters and the Fermi velocity component in the x direction $v_{Fx} = v_F\cos\psi$ depend on the impact angle ψ. The relevant energy scale in the problem is the ballistic Thouless energy $\varepsilon_{\mathrm{Th}} = v_F/2d$. As we will show below, the DOS is nonzero only for $|\alpha_\sigma| > t_\uparrow t_\downarrow$, which for a sufficiently large impact angle always is fulfilled. Clearly, the most interesting regime concerns $\varepsilon/\varepsilon_{\mathrm{Th}} \sim |\vartheta_{\sigma\pm}| \sim t_\uparrow t_\downarrow$.

In the tunneling limit, for small excitation energies $\varepsilon/\varepsilon_{\mathrm{Th}} \ll 1$ and small spin mixing angles $\vartheta_{\sigma\pm}$, we obtain $\alpha_\sigma = (4\varepsilon d/v_{Fx} + \sigma\vartheta_N)$. In this case, due to $\vartheta_{\sigma+} + \vartheta_{\sigma-} = \sigma\vartheta_N$, only the spin mixing angle for reflection at the normal side of the interface enters and acts as an effective exchange field $b = \vartheta_N v_{Fx}/4d$ on the quasiparticles. Especially interesting is the case $\varepsilon = 0$, for which all proximity amplitudes are even in momentum. For $\varepsilon = 0$ we obtain $\alpha_\sigma = \sigma\vartheta_N$, and the pairing amplitudes are either purely (odd-frequency) triplet for $|\vartheta_N| > t_\uparrow t_\downarrow$,

$$f_s(0) = 0, \qquad f_t(0) = -t_\uparrow t_\downarrow \mathrm{sgn}(\vartheta_N)/\sqrt{\vartheta_N^2 - (t_\uparrow t_\downarrow)^2}, \quad (4)$$

or purely (even-frequency) singlet for $|\vartheta_N| < t_\uparrow t_\downarrow$,

$$f_s(0) = it_\uparrow t_\downarrow/\sqrt{(t_\uparrow t_\downarrow)^2 - \vartheta_N^2}, \qquad f_t(0) = 0. \quad (5)$$

Comparing with the results for the diffusive case, we find that G_ϕ/G_T corresponds to $-\vartheta_N/(t_\uparrow t_\downarrow)$.

In Fig. 3, we show results for the proximity amplitudes in the ballistic limit and focus on positive values of ϑ_N without loss of generality. A systematic expansion of all terms in the tunneling probability shows that in the tunneling limit the spin dependence of the transmission probabilities can be neglected, and only that of the phase shifts needs to be kept. Thus, we assume $t_\uparrow = t_\downarrow = t$. We model the dependence on the impact angle ψ as $t(\mu) = (t_0)^{1/\mu}$, $\mu = \cos\psi$, and assume for simplicity spin mixing angles independent of μ. The tunneling probability for normal impact is $T_0 = t_0^2$. In the case $T_0 < \vartheta_N$ at small energies, the odd-frequency triplet amplitude dominates, and it is the only nonzero amplitude at $\varepsilon = 0$. On the other hand, for $T_0 > \vartheta_N$ both singlet and triplet amplitudes contribute. This is due to the fact that for large impact angles the transmission probability $t(\mu)^2$ drops below the value for the spin mixing angle ϑ_N.

We turn now to the DOS. The general expression, assuming the bulk solution in the superconductor, is $N(\varepsilon)/N_0 = \mathrm{Re}\sum_{\sigma=\pm1}\int_0^1 |\alpha_\sigma|/\sqrt{\alpha_\sigma^2 - (t_\uparrow t_\downarrow)^2}d\mu$. In the

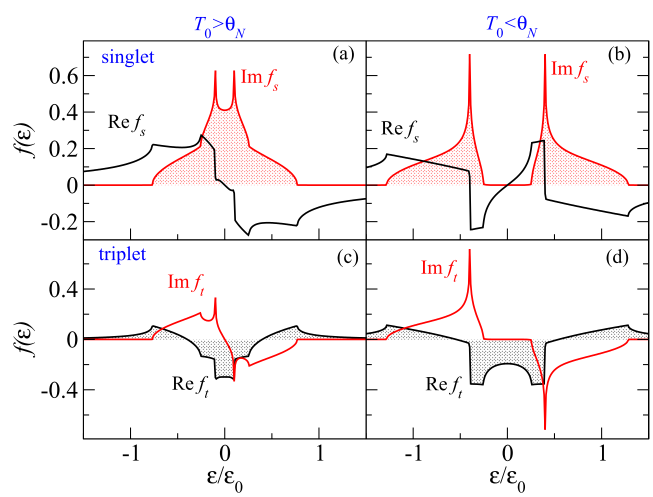

FIG. 3 (color online). Momentum-averaged proximity amplitudes at the surface of the normal layer. Parameters: $d = v_F/\Delta$ and $T_0 = 0.1$ (see text). (a),(c) $\vartheta_N = \vartheta_S = 0.05 < T_0$; (b), (d) $\vartheta_N = \vartheta_S = 0.15 > T_0$. Energy units are $\varepsilon_0 = T_0\varepsilon_{\mathrm{Th}}$. Even-frequency singlet components are shown in (a)–(b) and odd-frequency triplet components in (c)–(d).

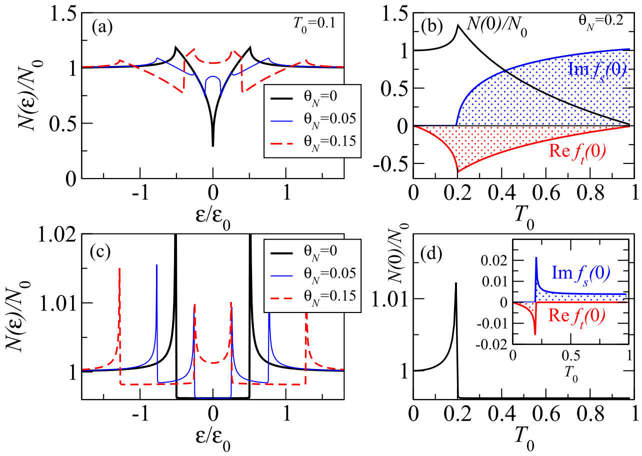

FIG. 4 (color online). (a) DOS as a function of energy at the top of the normal layer for fixed transmission probability $T_0 = 0.1$ and various values of $\vartheta_N = \vartheta_S$. The remaining parameters are as in Fig. 3. (b) DOS and proximity amplitudes at $\varepsilon = 0$ for $\vartheta_N = \vartheta_S = 0.2$ as a function of T_0. In (c) and (d), we show the results corresponding to (a) and (b) when assuming an (abrupt) tunneling cone with an opening angle of 10°.

tunneling limit, this simplifies again, and, provided that $|\vartheta_N| > t_\uparrow t_\downarrow$ for all impact angles, the DOS at the Fermi level is enhanced above its normal-state value $N(0)/N_0 = \int d\mu |\vartheta_N| / \sqrt{\vartheta_N^2 - (t_\uparrow t_\downarrow)^2}$.

In Fig. 4, we show results for the DOS. In Figs. 4(a) and 4(b), we assume the dependence on the impact angle as above, whereas in Figs. 4(c) and 4(d), we allow tunneling only in a narrow tunneling cone of 10°. The DOS for the cases of dominating triplet amplitudes and dominating singlet amplitudes differ qualitatively. In the case of a tunneling cone, this difference is most drastic, and a comparison with the results above shows that it is very similar to the diffusive case. In the right panels, where $\vartheta_N = \vartheta_S = 0.2$, we demonstrate that for $T_0 < 0.2$ only the odd-frequency triplet amplitude is present at the chemical potential, while the singlet amplitude is zero. The corresponding zero-energy DOS is enhanced in this region, whereas it is reduced in the region when singlet correlations are present at $\varepsilon = 0$.

The simplest experimental manifestation of the odd-frequency component is a zero-energy peak in the DOS [21–23]. In $S|F$ layers, where this phenomenon has been discussed, a clear peak at zero energy is often masked by the presence of singlet correlations f_s, which tend to suppress the DOS at low energy. This is not so in the system we consider, provided $T_0 < |\vartheta_N|$ in the ballistic limit or, equivalently, $G_T < |G_\phi|$ in the diffusive limit. This is ideal for an observation of the odd-frequency component, manifested as a zero-energy peak in the DOS.

The important factor with regard to isolation of the odd-frequency correlations at zero energy is the interface. The even-frequency correlations vanish when the interface transmission T_0 is sufficiently low. The parameters ϑ_N or, equivalently, G_ϕ can be increased by increasing the magnetic polarization of the barrier separating the superconducting and normal layers. By fabricating several samples with progressively increasing strength of magnetic moment $\vec{\mu}$ of the barrier, one should be able to observe an abrupt change at the zero-energy DOS above a certain strength of $\vec{\mu}$. Alternatively, one could alter T_0 by varying the thickness of the insulating region.

In summary, we have investigated the proximity effect in a $S|N$ bilayer with a spin-active interface. We find that, in both the ballistic and diffusive limits, the even-frequency correlations may vanish at zero energy, while odd-frequency correlations persist. This result is independent of the specific values for the layer thicknesses and barrier resistances, indicating that it is a robust and general feature of spin-active interfaces. Our findings suggest a way of obtaining unambiguous experimental identification of superconducting odd-frequency correlations.

A. Cottet and D. Huertas-Hernando are thanked for helpful communications. J. L. and A. S. were supported by the Norwegian Research Council Grants No. 158518/431, No. 158547/431 (NANOMAT), and No. 167498/V30 (STORFORSK). T. Y. was supported by the JSPS.

[1] F. S. Bergeret *et al.*, Rev. Mod. Phys. **77**, 1321 (2005).
[2] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).
[3] Y. Tanaka *et al.*, Phys. Rev. Lett. **99**, 037005 (2007); **98**, 037003 (2007).
[4] M. Eschrig *et al.*, J. Low Temp. Phys. **147**, 457 (2007).
[5] T. Yokoyama *et al.*, Phys. Rev. B **78**, 012508 (2008).
[6] V. L. Berezinskii, JETP Lett. **20**, 287 (1974).
[7] A. Volkov *et al.*, Phys. Rev. Lett. **90**, 117006 (2003).
[8] J. Linder *et al.*, Phys. Rev. B **77**, 174514 (2008).
[9] M. Kupriyanov *et al.*, Zh. Eksp. Teor. Fiz. **94**, 139 (1988); Yu. Nazarov, Superlattices Microstruct. **25**, 1221 (1999).
[10] H. le Sueur *et al.*, Phys. Rev. Lett. **100**, 197002 (2008).
[11] T. Tokuyasu *et al.*, Phys. Rev. B **38**, 8823 (1988); A. Millis *et al.*, Phys. Rev. B **38**, 4504 (1988).
[12] A. Brataas *et al.*, Phys. Rev. Lett. **84**, 2481 (2000); Phys. Rep. **427**, 157 (2006).
[13] D. Huertas-Hernando *et al.*, Phys. Rev. Lett. **88**, 047003 (2002).
[14] M. Eschrig *et al.*, Phys. Rev. Lett. **90**, 137003 (2003); M. Eschrig and T. Löfwander, Nature Phys. **4**, 138 (2008).
[15] A. Cottet *et al.*, Phys. Rev. B **72**, 180503 (2005).
[16] J. W. Serene *et al.*, Phys. Rep. **101**, 221 (1983).
[17] K. Usadel, Phys. Rev. Lett. **25**, 507 (1970).
[18] R. Meservey *et al.*, Phys. Rep. **238**, 173 (1994).
[19] A. M. Clogston, Phys. Rev. Lett. **9**, 266 (1962); B. S. Chandrasekhar, Appl. Phys. Lett. **1**, 7 (1962).
[20] M. Eschrig, Phys. Rev. B **61**, 9061 (2000); M. Fogelström, Phys. Rev. B **62**, 11 812 (2000); E. Zhao *et al.*, Phys. Rev. B **70**, 134510 (2004).
[21] Y. Asano *et al.*, Phys. Rev. Lett. **98**, 107002 (2007).
[22] T. Yokoyama *et al.*, Phys. Rev. B **75**, 134510 (2007).
[23] V. Braude *et al.*, Phys. Rev. Lett. **98**, 077003 (2007).