Anomalous diamagnetic response in multiband superconductors with broken time-reversal symmetry

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Within a Ginzburg-Landau formalism we establish analytically the necessary and sufficient conditions to realize a doubly degenerate superconducting ground state with broken time-reversal symmetry (BTRS) in a multiband superconductor. Using these results we analyze the ground state of a three-band superconductor in the cylindrical geometry in an external magnetic field. We show that depending on the interband coupling constants, a magnetic flux can induce current density jumps in such superconducting geometries that are related to adiabatic or nonadiabatic transitions from BTRS to time-reversal-symmetric states and vice versa. This unusual current-induced magnetic flux response can in principle be used experimentally to detect superconducting BTRS ground states as well as corresponding metastable excited states.

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I. INTRODUCTION

The phenomenon of superconductivity is characterized by the spontaneous breaking of a gauge symmetry. But in some cases simultaneously time-reversal symmetry (TRS) can be broken as well. Because of their unusual properties such superconductors (SCs) with broken time-reversal symmetry (BTRS) are attracting a lot of attention. For instance, recently the formation of new collective modes [1,2] (similar to the occurrence of Leggett modes in two-band SCs) and new topological excitations in the form of phase solitons, domains, and vortices that carry fractional magnetic flux values [3–12] have been discussed.

So far, BTRS superconductivity has been clearly detected in only a few cases. The most frequently cited example is Sr$_2$RuO$_4$ in which the order parameter was identified as a triplet chiral (Δ ∝ ρz + iρy) [13,14]. Furthermore, evidence for BTRS superconductivity was reported for the low-T phase of Th-doped UBe$_{13}$ [15], UPt$_3$ [16], and in SrPtAs based on muon measurements [17]. Theoretically superconductivity with BTRS has also been proposed for other compounds such as cuprate SCs at low temperature [18,19], transition-metal dichalcogenides [20,21], Na$_3$CoO$_2$·yH$_2$O [22], strongly doped graphene [23], and in some recently discovered Fe-based superconductors (FeSCs).

The FeSCs are of particular interest as BTRS superconductivity is anticipated for several dopings as a result of the multiband electron structure and strong repulsive interband couplings. Typically the Fermi surface of the non-SC parent compound consists of two or three hole-like pockets at the Γ = (0,0) point and two electron-like pockets around the M = (π,π) point. The resulting nesting at the vector Q = (π,π) connecting the Γ and M points drives the system to a spin density wave (SDW) state. With doping the SDW state melts, giving space for superconductivity. The natural symmetry of the order parameter in such a situation is given by the so-called s±, one, which causes a gap function with opposite signs at the electron and the hole pockets, respectively. In Ba$_{1-x}$K$_x$Fe$_2$As$_2$ the hole doping by K substitution leads at $x$ close to 1 to the vanishing of the electrons pockets and a change of the symmetry of the superconducting order parameter to nodal d-wave symmetry for pure KFe$_2$As$_2$ [24–27]. A BTRS state was proposed for two dopings: for $x \sim 0.7$, when the electron pockets vanish, and for $x \sim 1$, when a transition from s- to d-wave superconductivity is expected. The proposed intermediate pairing symmetries are $s + is$ and $s + id$ correspondingly [28,29].

However, as stressed above, still there are only a few compounds where superconductivity with BTRS was unambiguously observed. One of the reasons actually is the lack of simple experimental tools to identify it. The most common techniques to establish BTRS in superconductors are μSR and NMR; both suffer from restrictions related to the presence of impurities and other defects even in high-quality single crystals. In addition NMR requires considerable magnetic fields which may themselves significantly affect the superconducting ground state (GS), especially for low-temperature SCs, by paramagnetic pair-breaking effects and field-induced coexisting magnetic phases.

Here we first study in Sec. II the nature of possible SC ground states in zero magnetic field and the absence of currents. Within a Ginzburg-Landau approach we clarify the conditions for BTRS and explicitly show the twofold degeneracy of the corresponding ground states. We adopt the simplest model when the BTRS superconductivity, namely a three-band SC, is described approximately within a...
Ginzburg–Landau approach [30–35]. For a two-band model BTRS superconductivity is possible only in special cases such as dirty materials in the vicinity of the $s_\pm \rightarrow s_{++}$ transition [29].

In Sec. III we investigate the magnetic field response of superconducting cylinders with a BTRS order parameter and introduce it as a tool, which has not yet been considered in the literature to the best of our knowledge, to identify superconductivity with BTRS. In the three-band framework we investigate homogeneous current states in such a mesoscopically one-dimensional system and show that the diamagnetic, i.e., orbital-dominated, response depends directly on the nature of the underlying order parameter. Experimental verification of the characteristics that we predicted here could be used to identify multiband BTRS superconductivity.

II. GINZBURG–LANDAU APPROACH TO SUPERCONDUCTIVITY WITH BTRS

To describe the multiband superconductors we employ a general Ginzburg–Landau (GL) functional, which has been used previously only for particular cases (e.g., for two bands or three equivalent bands [36,37]; see the reviews Refs. [6,7] and references therein). We will provide a rather general solution for three nonequivalent bands with repulsive interband interactions being the most relevant case for BTRS physics in these systems. For the sake of simplicity we address only homogeneous states and isotropic order parameters. However, inhomogeneous states containing different topological defects and an explicit account of spin states can be treated straightforwardly within the same formalism [38]. The GL Gibbs energy density and the current density for three-band superconductors can be written in the following form:

$$\Delta G = \frac{1}{2} \sum_{i=1}^{3} \left[ \frac{1}{4m_i} \left(-i\hbar \nabla - \frac{2e}{c} A\right) |\psi_i| - a_i |\psi_i|^2 + \frac{b_i}{2} |\psi_i|^4 \right] + \frac{1}{8\pi} \int \left( \nabla A - \mathbf{H} \right)^2 \quad (1)$$

and

$$\mathbf{j} = -\sum_{i} \frac{i\hbar}{m_i} (\psi_i^* \nabla \psi_i - \psi_i \nabla \psi_i^*) - \frac{4e^2}{c} A \sum_{i} \frac{|\psi_i|^2}{m_i} \quad (2).$$

Here and below we consider the temperature regime below $T_c$ for the GL approach as valid; i.e., we ignore the region of strong fluctuations in the very vicinity of $T_c$ or in the case of very low temperatures. In the first integral in Eq. (1) the integration is performed over the superconductive region whereas in the second integral over the cylinder volume. The term $F_{\text{int}}$ describes the phase-sensitive Josephson-like interband coupling:

$$F_{\text{int}} = \sum_{i} \gamma_i^1 \psi_i^* \psi_2 + \gamma_{23} \psi_2^* \psi_3 + \gamma_{31} \psi_3^* \psi_1 + \text{c.c.}$$

$$= 2\gamma_{12} |\psi_1|^2 |\psi_2|^2 \cos(\phi_1 - \phi_2) + 2\gamma_{23} |\psi_2|^2 |\psi_3|^2 \cos(\phi_2 - \phi_3) + 2\gamma_{31} |\psi_3|^2 |\psi_1|^2 \cos(\phi_3 - \phi_1). \quad (3)$$

Here the order parameter is in general complex, i.e., $\psi_i = |\psi_i| e^{i\phi}$. In contrast, the interaction coefficients $\gamma_{ij}$ are real and can be positive or negative. It was shown for two-band superconductors that the sign of $\gamma_{12}$ fully determines the symmetry of the order parameter in the clean case. A repulsive interband interaction constant $\gamma_{12} < 0$ leads to unconventional symmetry and a ground state with zero-phase difference between the two bands (denoted as $s_{+}$ symmetry), while attractive interband interactions $\gamma_{12} > 0$ stabilize a ground state with a zero-phase difference between the their gap functions (denoted as $s_{++}$ symmetry). We keep the same sign convention in the case of three-band SCs considered here.

As a first step we examine the ground state in the absence of external magnetic fields. Then Eq. (4) can be rewritten as $F_{\text{int}} = \text{sgn}(\gamma_{12}) \gamma_{12} |\psi_2|^2 |\psi_1|^2 - \gamma_{23}^2 |\psi_3|^2 - \gamma_{31}^2 |\psi_1|^2$. The minimum of GL corresponds to the order parameter:

$$|\psi_i|^2 = \frac{\gamma_{13}^2 |\psi_3|^2 - \gamma_{23}^2 |\psi_2|^2}{2\gamma_{12}} \quad (4)$$

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and

$$\cos \theta = \cos(\phi_2 - \phi_1) = \frac{\gamma_{12}^2 |\psi_3|^2 - \gamma_{23}^2 |\psi_1|^2 - \gamma_{31}^2 |\psi_2|^2}{2\gamma_{12} |\psi_2| |\psi_3|} \quad (6)$$

$$\cos \phi = \cos(\phi_3 - \phi_1) = \frac{\gamma_{23}^2 |\psi_2|^2 - \gamma_{13}^2 |\psi_1|^2 - \gamma_{31}^2 |\psi_2|^2}{2\gamma_{13} |\psi_3| |\psi_3|} \quad (7)$$

The dependence of the interband interaction term $F_{\text{int}}$ of the GL free energy functional as a function of the phase differences for a fixed set $G_i$ is given in Fig. 1. Here we have introduced convenient new variables:

$$G_1 = \frac{\gamma_{12} |\psi_1|}{|\psi_2|}, \quad G_2 = \text{sgn}(\gamma_{23}), \quad G_3 = \frac{\gamma_{13} |\psi_3|}{|\psi_2|} \quad (8)$$

A BTRS state exists for zero external magnetic field only within a relative small volume in the six-dimensional parameter space $(|\psi_1|, \gamma_{ij})$. The corresponding projected regions onto the planes $G_1, G_2$, $G_3$ are shown in the form of tilted,
of the interband interactions in three-band superconductors does not provide a necessary and sufficient condition for the occurrence of a BTRS GS, contrary to some statements found in the literature [31,42,43]. To gain deeper insight into the nature of a BTRS GS, contrary to some statements found in the literature, we propose to apply a magnetic flux to a (topologically) doubly connected system. In particular, we consider a long and thin tube approximated by two concentric cylinders with the inner and outer radii \( R_1 \) and \( R_2 \), respectively (see Fig. 3). Its symmetry axis is denoted as the \( z \) axis in cylindrical coordinates \((r, \vartheta, z)\). An external constant magnetic field \( H \) is thought to be applied along the symmetry axis of such a cylinder: \( H = (0,0,H) \), where the vector-potential gauge is chosen as \( A = (0,A_{\vartheta}(r),0) \), \( A_{\vartheta}(r) = \frac{Hr}{2} \). We assume that the radius of the tube \( R \) and the thickness \( \delta \) satisfy the following conditions: \( R \gg \lambda, \xi \) and \( \delta \ll \lambda, \xi \), where \( \lambda(T) \) and \( \xi(T) \) are the London penetration depth and the coherence length, respectively. The first condition precludes the formation of magnetic vortices or any domains in the cylinder, while due the second one the self-induced magnetic fields are small and can therefore be ignored in our calculations. This means that we study only homogeneous solutions \( |\psi_i| = \text{constant} \), while the phase depends only on the polar angle \( \vartheta \). In the considered geometry the phase must fulfill the quantization condition \( \oint_\Gamma \nabla \psi_i = 2\pi n_i \), where the integral is taken over an arbitrary closed continuous contour \( \Gamma \) lying inside the cylinder and \( n = 0, \pm 1, \pm 2 \) are the phase winding (topological) numbers. Here we assume these winding numbers to be equal: \( n_1 = n_2 = n_3 \).

It is interesting to note that a similar experimental setup was proposed with the aim to detect a fractional flux plateau in the magnetization curve of a superconducting loop that is topologically the same as the one considered here [44]. These authors investigate numerically metastable phase kinks protected by a large energy barrier within a GL functional adopting certain special parameter values. The excited states with BTRS discussed in the following differ significantly from those solitonic states.

### III. BTRS AND TRS SUPERCONDUCTORS ON A CYLINDER AND IN MAGNETIC FIELD

In order to distinguish readily a three-band SC with a BTRS ground state from those traditional SCs with a TRS ground state, we propose to apply a magnetic flux to a (topologically) doubly connected system. In particular, we consider a long and thin tube approximated by two concentric cylinders with the inner and outer radii \( R_1 \) and \( R_2 \), respectively (see Fig. 3). Its symmetry axis is denoted as the \( z \) axis in cylindrical coordinates \((r, \vartheta, z)\). An external constant magnetic field \( H \) is thought to be applied along the symmetry axis of such a cylinder: \( H = (0,0,H) \), where the vector-potential gauge is chosen as \( A = (0,A_{\vartheta}(r),0) \), \( A_{\vartheta}(r) = \frac{Hr}{2} \). We assume that the radius of the tube \( R \) and the thickness \( \delta \) satisfy the following conditions: \( R \gg \lambda, \xi \) and \( \delta \ll \lambda, \xi \), where \( \lambda(T) \) and \( \xi(T) \) are the London penetration depth and the coherence length, respectively. The first condition precludes the formation of magnetic vortices or any domains in the cylinder, while due the second one the self-induced magnetic fields are small and can therefore be ignored in our calculations. This means that we study only homogeneous solutions \( |\psi_i| = \text{constant} \), while the phase depends only on the polar angle \( \vartheta \). In the considered geometry the phase must fulfill the quantization condition \( \oint_\Gamma \nabla \psi_i = 2\pi n_i \), where the integral is taken over an arbitrary closed continuous contour \( \Gamma \) lying inside the cylinder and \( n = 0, \pm 1, \pm 2 \) are the phase winding (topological) numbers. Here we assume these winding numbers to be equal: \( n_1 = n_2 = n_3 \).

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To illustrate the principle of identifying the BTRS for a given three-band SC, we consider a simple case and assume first that the equilibrium values of the order parameters are given but without adopting thereby the equality for the moduli of their interband interactions. Second, the strengths of the interband interactions coincide but, for instance, at least one of these interactions is repulsive. Since we are interested in a three-band superconductor with initial BTRS state we control the selection of parameters of interband interactions (see Figs. S1–S8 of the Supplemental Material [39]). Then we can write the GL free energy of the system in the form of the equation of the equation in zero magnetic field by substituting $a_i \rightarrow a_i + \kappa_i q^2$; i.e., an increase of $q$ acts in the same way as an increase of temperature. With this remark we can apply the considered above results for zero magnetic field.

To demonstrate the induced transition from a BTRS to a TRS state by an external magnetic field we consider a particular case of equal $a_i = a$, $b_i = b$, but keeping the $\kappa_i$ different and $\gamma_2 = \gamma_3 = -\gamma_3 = \gamma$. At zero $q$ a doubly degenerate BTRS state with $\phi = 2\pi - \theta = 5\pi/3$ and $\phi = 2\pi - \theta = \pi/3$ is realized. With increase $q$ first at $q_1$, which is the solution of the equation $\cos \phi_1 \theta_1 = 1$, we get transitions to TRS states (see Supplemental Material [39]). In the TRS state the minimization of the GL energy cannot be done analytically. In this case a numerical procedure must be applied. The dependence of the GL free energy Eq. (6) on the applied magnetic flux for different ratios of $\kappa_i$ is presented in Fig. 4. We track the evolution with magnetic flux of one of the ground states, namely for $\phi = 5\pi/3$, $\theta = \pi/3$. The full procedure can be found in the Supplemental Material [39]. We find that for a given value of $\tilde{\gamma}$ the ground state of a three-band superconductor under consideration exhibits always a BTRS. This means that despite the value of the trapped flux, by increasing the flux we will move along the bottom part of the solid curves (Fig. 4), following the route $10$–$1$–$2$–$\cdots$. But for a nonadiabatic, fast switched on magnetic flux, the three-band superconducting system can be excited and can be flipped to a metastable state with TRS. For instance, the previous ground state route $10$–$1$–$2$–$3$–$\cdots$ can be replaced by the path $10$–$1$–$L_1$–$2$–$3$–$\cdots$, where the dashed part $L_1$–$L_2$ corresponds to the above-mentioned metastable state with TRS of the three-band superconducting tube, or to a more complicated route, which will involve more excited states with TRS. Also we found that if $q_1 < 1/2$ then the transitions between BTRS and TRS states can occur without any excitation by an external magnetic flux. This means that solid (BTRS) and dashed (TRS) lines cross before $\Phi / \Phi_0 = 1/2$ (see the inset in Fig. 5). The phase diagram for a three-band superconductor, which determines the intervals of the parameters $\kappa_i$ for the transitions from a BTRS to a TRS state with an excitation and without one, is given in Fig. 5.

Further numerical examinations give that, if $\kappa_i > 1$ ($i > 2$), a nonadiabatic switching on of the increase of the magnetic flux can lead to a transformation of a three-band SC with a more complicated route.
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Fig. 6. Possible evolution of the phase differences $\phi$ (blue) and $\theta$ (green) for a three-band superconductor with nonadiabatic [(a), (b)] and adiabatic (c) transitions between BTRS and TRS states. The parameters are $\kappa_2 = 4$ and $\kappa_3 = 2$ [(a), (b)], $\kappa_2 = 0.25$ and $\kappa_3 = 0.5$ (b), and $\kappa_2 = 15$ and $\kappa_3 = 1.5$ (c).

BTRS GS into an excited state with TRS and an $s_{++}$ order parameter [see Figs. 6(a) and 6(c)], and then it relaxes again to a BTRS GS. If one or both $\kappa_i < 1$ ($i \geq 2$), then the increasing magnetic flux can transform a three-band SC with BTRS into an $s_{++}$ three-band SC and finally again to a BTRS state [see Fig. 6(b)].

From the experimental point of view transitions from the BTRS state to a TRS one and vice versa can be detected following the response of the current density on an applied magnetic flux (see Fig. 7). We found appropriate jumps in the $j(\Phi/\Phi_0)$ dependencies [see Figs. 7(b), 7(d), and 7(e)] induced by these transitions. Since in the present geometry changing of the magnetic field is equivalent to changing temperature, we expect also special features for the specific heat related to these BTRS to TRS transitions. Hence accompanying thermodynamic measurements might provide further support for identification of the BTRS states.

IV. DISCUSSION AND CONCLUSIONS

Based on these analytical and numerical calculations it is natural to suggest that such a behavior remains on a qualitative level the same also for other possible sets of interband interaction coefficients which admit the existence of frustrated states in the equilibrium state [45] (see Supplemental Material [39]). In other words jumps in the current dependencies in the magnetic-flux-driven regime can be expected for any three-band superconductor with a primordial (before switching on a magnetic field) BTRS state. Moreover with some restrictions it is reasonable to expect the same behavior also for other BTRS multiband superconductors, whose electronic structure and physical properties are described by more than three order parameters and where frustrated states are global ground states. Restrictions of the application of such method are connected with the special case of multiband superconductors with an even number of bands and all equal repulsive interband
interactions, where BTRS and TRS states have the same energy [46–48]. We believe that the presence of such jumps can be considered as an experimental proof for the detection of BTRS and frustration in unconventional three- and higher multiband superconductors. It is important to note that the detection method proposed here compares favorably with surface-sensitive techniques (interference or proximity based contacts) for the detection of properties related to the symmetry of the order parameter, because it probes the entire volume of the superconductor under examination. Based on our results we propose detecting the presence of frustration and BTRS in experiments with mesoscopic thin rings or tubes made from unconventional three-band superconductors, by measuring a generic current response on the applied magnetic flux.

It should be noted that currently the exact location of the soliton states on the energetic scale of a three-band superconductor is not known. Knowledge of all possible topological defects and their energies in the cases of three- and other multiband superconductors is very important for the detection of the BTRS phenomenon in order to distinguish the jumps connected with the presence of BTRS to TRS transitions and from the transitions from a BTRS ground state to excited soliton states. If the energy of phase-inhomogeneous solutions is higher than the homogeneous BTRS and TRS states then during the excitation one can in principle observe additional jumps on the current–magnetic flux dependencies due to relaxation processes from higher energetic levels (soliton states) to the ground state via metastable TRS states. Another situation is realized for solitons, whose energy is within the interval between homogeneous BTRS and TRS states. In this case during the excitation process a three-band superconductor can be promoted to a homogeneous TRS state as an intermediate state and then relax to the ground state via other intermediate states of solitonic nature. So also in this case additional jumps will also appear on the experimental dependencies. The last possibility can occur if the BTRS state is not a globally stable state and the ground state of a three-band superconductor already contains solitons. The realization of such a scenario was predicted recently for a three-band superconductor [4] based on nonrigorous stability considerations of phase kinks for an infinitely extended superconducting system. To the best of our knowledge a study of topological defects in three-band superconductors for a doubly connected finite superconducting system as considered here is still lacking. And it is not clear whether such solitons in a restricted geometry can also occur as globally stable phenomena. We will study this interesting but complex problem in more detail in the future. Another important and interesting case is when a TRS state is the ground state of a three-band superconductor. To understand the character and possible novel features of the diamagnetic response for possible TRS-BTRS transitions this problem will be addressed within the time-dependent Ginzburg-Landau theory in future studies. As a first step the investigation of soliton metastable states that are important for a more realistic description must be included. The calculations have been performed and the corresponding manuscript is now under preparation. Preliminary results show that soliton states are thermodynamically metastable and are located at an energy scale higher then the BTRS and even the TRS states. Also the cases of imperfect three-band superconductors with impurities as well as inhomogeneous states due to the presence of solitonic nonlinear excitations mentioned above all require a separate special analysis outside of the scope of the present paper and be left for future study.

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For two-band superconductors it was proven [49] that non-trivial topological states, which arise exactly for $n_1 \neq n_2$, are thermodynamically metastable and do not correspond to the ground state of the superconducting system. Hence, it is reasonable to apply such type of condition also to the case of three-band superconductivity considered here and put $n_1 = n_2 = n_3 = n$. Since the subject of our consideration is solely a homogeneous current state, here we do not consider the presence of topological defects such as phase solitons. In fact, solitonic solutions can be studied also within our approach. But the question of their local or global thermodynamic stability is still under debate (unfortunately, the arguments provided in Refs. [50,51] are mostly heuristic in nature and related to systems with open boundary conditions). A consideration of such topological defects in the case of periodic boundary conditions is out of the scope of the present paper.

See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.96.144513 for more information about mathematical details of the derivations given in the main text, additional examples of the phase diagrams showing the conditions of existence of BTRS states, as well as the example of a BTRS state in a mesoscopic ring imitating three-band superconductivity.


We note that the different behavior of the current density is not connected solely with the coinciding strength of the interband interactions. According to our analysis it holds for different values of the interband interaction coefficients, too, except for the case where these parameters vanish.


