Fractional Flux Plateau in Magnetization Curve of Multi-Component Superconductor Loop

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Time-reversal symmetry (TRS) may be broken in superconductors with three or more condensates interacting repulsively, yielding two degenerate states specified by chirality of gap functions. We consider a loop of such superconductor with two halves occupied by the two states with opposite chiralities. Fractional flux plateaus are found in magnetization curve associated with free-energy minima, where the two domain walls between the two halves accommodate different inter-component phase kinks leading to finite winding numbers in a part of the whole condensates around the loop. Fractional flux plateaus form pairs with their heights related to the flux quantum $\Phi_0 = hc/2e$. This phenomenon is a clear evidence of TRSB superconductivity, which in a general point of view provides a novel chance to explore relative phase difference, phase kink and soliton in ubiquitous multi-component superconductivity such as that in iron pnictides.

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\textbf{Introduction.---} Vortex with $2\pi$ phase winding in order parameters is a hallmark of macroscopic quantum phenomena such as superfluidity and superconductivity [14]. In superconductor, vortex is accompanied by a quantum of magnetic flux $\Phi_0 = hc/2e$ in a closed path with zero supercurrent. Since the quantization of magnetic flux is intimately related to the phase winding, superconductivity gap functions carrying intrinsic phase variation induced by unconventional pairing symmetry should leave unique consequences on the response to external magnetic field. By now several interesting examples are available. A tricrystal ring of cuprate superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ was observed to carry a half flux quanta $\Phi_0/2$, which is the signature for $d$-wave pairing symmetry [5]. In a ring-shaped setup composed by Nb and NdFeAsO$_{0.85}$F$_{0.12}$, flux jumps in odd-number multiple of half flux quanta were observed [6,7], which provides a support to the $S^+_- \pm$ pairing symmetry for iron-pnictide superconductors [6,13]. Half-valued fluxoid jumps in magnetization curve of a thin annular coil made of Sr$_2$RuO$_4$ were reported as consistent with the $p$-wave pairing symmetry [17].

The degree of freedom of relative phase difference in a two-component superconductor was first discussed by Leggett [18], and experimental observation on the collective Leggett mode was reported in MgB$_2$ [19,20]. Phase solitons in one of the two superconducting gap functions associated with fractional fluxoid jumps have been investigated [21,26].

Another interesting possibility in multi-component unconventional superconductivity was raised some time ago [27] that Josephson-like inter-component repulsions among three condensates generate frustrations in phases of gap functions, which breaks the time-reversal symmetry (TRS) even without external magnetic field and results in a pair of degenerate states specified by chirality of gap functions. Since the discovery of iron-pnictide superconductors where several orbitals of Fe contribute to multi superconducting condensates, this possibility becomes realistic and a considerable amount of subsequent works have been devoted to discuss its thermodynamic stability and various novel properties [28,42].

Recently it has been revealed by the present authors that in a Josephson junction between a conventional single-component superconductor and multi-component superconductor in TRSB state the critical current should be asymmetric with respect to current direction as the consequence of broken $\text{TRS}$ [43]. As a matter of fact, unequal critical currents in opposite current directions were observed experimentally in the Josephson junction between PbIn and BaFe$_{1.8}$Co$_{0.2}$As$_2$ [44]. Therefore, the TRSB state may have been realized already in iron-based superconductors, which is consistent with microscopic analysis where band structures and strongly correlated effects are taken into account [30]. To cross check this novel superconducting phenomenon becomes an important issue.
In this Letter, we address a new phase-sensitive property of the TRSB state. As schematically shown in Fig. 1 we consider a loop of multi-component superconductor where the two halves are occupied by two TRSB states carrying on opposite chiralities, accompanied by two domain walls associated with inter-component phase kinks. Adopting Ginzburg-Landau (GL) approach, we reveal explicitly that fractional flux plateaus appear in magnetization curve associated with free-energy minima, where the domain walls accommodate phase kinks among different components leading to $2\pi$ phase winding only in one or two of the three condensates. While the heights of fractional flux plateaus depend on material parameters and temperature, they form pairs with heights related to the flux quantum $\Phi_0$, which is a unique signature of TRSB state and can be used to confirm the state itself. In a more general point of view this provides a novel chance to explore relative phase difference, phase kink and soliton in ubiquitous multi-component superconductivity.

Fractional flux plateaus. — In order to reveal the essence of physics we first consider an isotropic TRSB state which is generated by three equivalent condensates with equal mutual repulsion. For simplicity all order parameters are taken as $s$-wave from now on. The two states in the loop are given by $\Psi = \{\psi_1, \psi_2, \psi_3\} = |\psi||1, e^{i2\pi/3}, e^{i4\pi/3}\}$ and $\Psi^*$ with opposite chiralities (see Fig. 1). Across each of the two domain walls between the left and right halves of the superconductor loop, there is a phase kink where inter-component phase difference between two of the three order parameters shrinks to zero and reopens in the opposite way continuously, resulting a sign reversal in the phase difference at the two sides of domain (see Fig. 2). We notice that the phase kink is a gauge-invariant object, which at equilibrium appears at the interface between two bulks of TRSB states with opposite chiralities.

When the two domain walls accommodate the phase kink between the same two condensates, such as that between condensate 1 and 2 defined as $D_{12}$ in Fig. 2(a), the phase rotation integrated anticlockwisely (indicated by $L$ in Fig. 2) over the two domain walls cancel each other, resulting in the same phase winding in all the three condensates. In this case, the flux trapped in the loop is an integer multiple of flux quantum. The situation is much different when the two domain walls accommodate different phase kinks, such as $D_{12}$ and $D_{32}$ in domain wall I and II respectively shown in Fig. 2(b). By inspection one sees that $\psi_2$ rotates $4\pi/3$ anticlockwisely over the two domain walls, while $\psi_1$ and $\psi_3$ rotate $-2\pi/3$. When the external magnetic field provides the additional phase rotation of $2\pi/3$, a state with $2\pi$ phase winding in $\psi_2$ and 0 in both $\psi_1$ and $\psi_3$ is stabilized. This yields a fractional flux quanta $\Phi_0/3$ in the loop.

A general case with three unequal condensates can be analyzed by the Ginzburg-Landau (GL) formalism. The GL free-energy functional of three-band superconductor with Josephson-like inter-component couplings is given by

$$F = \sum_{j=1,2,3} \left[ \alpha_j |\psi_j|^2 + \frac{\beta_j}{2} |\psi_j|^4 + \frac{\gamma_{jk}}{2m^*} \left( \nabla - \frac{eA}{c} \right) \psi_j \right]$$

where an inter-component coupling is repulsive when $\gamma_{jk}$ is negative. For $\gamma_{12}\gamma_{13}\gamma_{23} < 0$, a TRSB superconducting state appears when the coefficients in Eq. (1) satisfy conditions re-
we show one example of this GL analysis. When the magnetic field is negligibly small. This state remains stable till the magnetic field increases to such an extent that phase kinks unwind. At integer flux quanta \( \Phi = 2\pi \Phi_0 \) and integer \( \Phi = \Phi_0 \), the phase kinks are locked due to strong mutual repulsions except for \( \Phi = 0 \) case that phase kinks \( D_{ij} \) and \( D_{ik} \) are realized at domain wall I and II. The first term should be an integer multiple of \( 2\pi \) due to the single-valued wave function in the loop, and the second term is responsible to possible fractional flux plateau, with height determined by the quantities \( p_j = |\psi_j|^2/m_j^* \).

There are six different configurations of phase kinks in a three-component system. The two configurations \( |D_{ij}/D_{ik}\rangle \) and \( |D_{ik}/D_{ij}\rangle \) at the domain walls \( [I]/[II] \) take the same free energy. However, integrating phase differences for these two configurations along the closed path in anticlockwise way (see Fig. 2) results in opposite fractional values of \( 2\pi \) in the second term in Eq. (2). With difference in the first term in Eq. (2) equal to unity, the two configurations give two fractional magnetic plateaus satisfying \( \Phi_1 + \Phi_2 = \Phi_0 \) as shown schematically in Fig. 4(a) (see numerical results in Fig. 3(a)). Whether all the fractional flux plateaus can be stabilized should be determined by the free energy analysis. In the case shown in Fig. 3, \( F(D_{ij}) < F(D_{ik}) \) such that only the phase-kink pair \( D_{12} \) and \( D_{23} \) is stabilized, resulting in the two plateaus in the magnetization curve. In general, there are at most six

\[
\Phi = \frac{\Phi_0}{2\pi} \left[ \int_C p_1 \nabla \psi_1 + p_2 \nabla \psi_2 + p_3 \nabla \psi_3 \right] + \int_{DW} \frac{p_i \nabla \psi_{ij} + p_k \nabla \psi_{ik}}{p_i + p_j + p_k} \cdot dl.
\]

where \( p_j = |\psi_j|^2/m_j^* \) and "DW" indicates the integration over domain walls, presuming that the inter-component phase differences are locked due to strong mutual repulsions except for at the domain walls. In the second line of Eq. (2), the total magnetic flux is divided into two parts, corresponding to the

![FIG. 4. (a) Schematic magnetization curve for a symmetric superconducting loop with fractional flux plateaus displayed together with gauge-invariant phase kinks denoted by \( |D_{ij}/D_{ik}\rangle \) in the order of domain wall I and II. (b) Magnetization curve with two fractional flux plateaus for an asymmetric loop, where the width of side including domain II is enlarged to \( 12\lambda(0) \) from that given in Fig. 3. The parameters are the same as Fig. 3 except for \( \alpha_1' = 0.025 \), \( \alpha_2' = 0.028 \) and \( \alpha_3' = 0.022 \).](image)
In the present work we study the case that \( D_{ij}/D_{ik} \) may be unstable even though \([D_{ij}/D_{ij}]\) is stable associated with free-energy minimum. As shown in Fig. 4(b), the fractional flux plateau at \( \Phi = 0.74\Phi_0 \) remains stable for \( 0.017 \lesssim H/H_{c1}(0) \lesssim 0.023 \), while that at \( \Phi = 0.26\Phi_0 \) disappears in contrast to Fig. 3, since they are associated with different phase-kink configurations at domain wall I and II. Nevertheless, it is worth noticing that even in this asymmetric loop the plateau at \( \Phi = -0.26\Phi_0 \) is still stable for \( -0.020 \lesssim H/H_{c1}(0) \lesssim -0.014 \), since it is associated with the same phase-kink configuration with that at \( \Phi = 0.74\Phi_0 \) with a difference of \( \Phi_0 \) guaranteed by Eq. (2). It is clear that the symmetry in magnetization curve with respect to the direction of magnetic field is broken in the geometrically asymmetric superconductor loop (see Fig. 4(b)). The property that fractional flux plateaus in positive and negative magnetic fields are paired with the difference of \( \Phi_0 \) is robust, and can be taken as a crosscheck for fractional flux plateaus originated from the TRSB state.

Discussions.— In the present work we study the case that the left and right halves of the superconductor loop take the two TRSB states with opposite chiralities. This situation can be realized in experiments by cooling the whole system from temperature above \( T_c \) with laser heat pulse irradiated on region I and II \[48\]. The two halves condensate independently and by chance arrive at the different TRSB superconducting states, leading to the two domain walls at region I and II after releasing the irradiation. In order to check the stability of this configuration, we estimate the free energy of the whole system in terms of TDGL approach. As shown in Fig. 5, the state with two domain walls locating at the middle of the top and bottom sides of the loop corresponds to a free-energy minimum. The domain walls once generated should be stable since moving them outside the loop is prohibited by a large free-energy barrier which is produced by an elongated, single domain wall during the process of domain-wall relocating (see that at \( x_2 \) in the inset of Fig. 5). The stability of the present setup against relocating one of the two domain walls along the loop can be provided by widening the left and right arms of the loop. The increase in free energy in states with fractional flux and integer flux quantum upon application of external magnetic field is smaller by one order of magnitude than the free-energy barrier as seen in Fig. 5, which justifies the discussion on fractional flux plateau in the present work.

The amplitudes and inter-component phase differences of order parameters changes with temperature, leading to variation in stable domain-wall structures. As a result, both height and width of fractional flux plateau should change as temperature is swept. During field-cooling or field-heating processes, jumps between states with fractional flux and/or integer multiples of flux quantum can take place.

In previous studies on vortex states of TRSB superconductor it was discussed that vortex cores of different condensates can deviate from each other in space \[40, 49–52\]. However, without a closed path with zero supercurrent there is no fractional flux plateau in magnetization curve.

In conclusion, we have studied the magnetic response of a loop of three-component superconductor with two degenerate time-reversal symmetry broken states at two halves. When the two domain walls between the two halves accommodate different inter-component phase kinks, fractional flux plateaus appear in the magnetization curve which form pairs with their heights related to each other by the flux quantum. These properties are expected to be helpful for detecting experimentally the time-reversal symmetry broken superconducting state which may be realized in iron-pnictide superconductors, and in general provides a novel chance to explore relative phase difference, phase kink and soliton in ubiquitous multi-component superconductivity.

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