Test Experiment for Time-Reversal Symmetry Breaking Superconductivity

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A new experiment is proposed to probe the time-reversal symmetry of a superconductor. It is shown that a time-reversal symmetry breaking superconductor can be identified by the observation of a fractional flux in connection with a Josephson junction in a special geometry.

During the last few years one of the important issues in the field of high temperature superconductivity was the experimental identification of the symmetry of the superconducting order parameter. Various theories suggest unconventional superconductivity due to Cooper pairing in the “d-wave” channel with a pair wave function of the form

\[ \psi(k) = \cos k_x - \cos k_y. \]

The phase characteristics of this state (the sign change under exchange of \( x \)- and \( y \)-coordinate) is the basis for a series of experiments probing directly the intrinsic phase difference of \( \pi \) between different momentum directions in the pair wave function [1]. It was shown that the presence of a d-wave pairing state could also be responsible for the occurrence of the peculiar paramagnetic response in granular Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Wohleben effect) [2]. Although by now a considerable number of experiments demonstrate convincingly the d-wave symmetry of the superconducting state at least for YBa$_2$Cu$_3$O$_7$ (YBCO) [3], this issue is still controversial as other experiments seem to be incompatible with a d-wave state [4]. Recently, Kirtley et al. reported the observation of vortices on grain boundaries in a film of YBCO, which carry a flux smaller than a standard flux quantum \( \Phi_0 (= hc/2e) \) (even smaller than \( \Phi_0/2 \) as it could originate from the intrinsic \( \pi \)-phase shift of a d-wave state) [5]. Subsequently, this observation was interpreted as an indication that the superconducting state breaks the time-reversal symmetry \( T \) if not in the bulk then at least near the grain boundaries [6]. Besides the high temperature superconductors, the heavy fermion systems UPt$_3$ and U$_{1-x}$Th$_x$Be$_{13}$ (0.018 < \( x \) < 0.045) are good candidates to have a \( T \)-violating superconducting phase [7]. In this letter we propose a new experiment to probe the time-reversal symmetry of a superconducting state. In the following we will examine a concrete model which may directly apply to the high temperature superconductors. However, most of our conclusions are also valid for other superconductors. We consider a thin film of the superconducting material, which is c-axis oriented and untwinned if any orthorhombic distortion is present as in YBCO. (Twinning leads to various complications which we will discuss elsewhere.) We cut two small separate holes in the film, which are connected by a straight homogeneous Josephson junction (see Fig.1). We assume that the junction is sufficiently strong such that the Josephson penetration depth \( \lambda_J \) is much smaller than the distance between the two holes. Thus, if magnetic field threads the two holes then the magnetic flux of each hole is separately a well-defined quantity. Obviously the geometry of the arrangement requires that the total flux of both holes together is an integer multiple of \( \Phi_0 \). However, we claim that it is possible to find an arbitrary amount of magnetic flux in each hole if certain conditions are satisfied. Let us assume that the flux through one of the two holes is \( \Phi \). If we applied the time-reversal operation to the system then this flux would change sign, \(-\Phi\). If the superconducting state (apart from the currents flowing to sustain the flux in the hole) is invariant under time-reversal, the original and the
inverted flux should differ by an integer multiple of $\Phi_0$. Thus, $\Phi = -\Phi + n\Phi_0$ which leads to $\Phi = n\Phi_0/2$. Hence $\Phi$ can only be an integer (for even $n$) or half-integer (for odd $n$) multiple of $\Phi_0$. The latter is possible only under special conditions which are not included in our discussion \[3\]. Thus, in order to obtain a flux different from these two cases, the superconducting state must break the time-reversal symmetry. Next, we reflect the system at a mirror plane which includes the junction and is parallel to the $c$-axis. This operation also changes the sign of $\Phi$. Hence, the same argument as before applies. Both the time-reversal and the reflection symmetry have to be violated in order to find an arbitrary flux $\Phi$ in the hole. It is reasonable to assume that the violation of reflection symmetry is connected with asymmetric properties of the junction and its vicinity. On the other hand, $T$ may be violated in the whole superconductor or, alternatively, only locally near the junction as proposed by Sigrist, Bailey and Laughlin (see also Ref.8) \[3\]. To be concrete let us now look at the example of a superconductor with a two-component order parameter, $\eta = (\eta_1, \eta_2)$. The first component has the above introduced d-wave symmetry, while the second is completely symmetric (s-wave). Under tetragonal crystal symmetry $\eta_1$ and $\eta_2$ are generally not degenerate and have different (bare) transition temperatures $T_{c1}$ and $T_{c2}$, respectively. For our purpose it is sufficient to study this system on a phenomenological level. The Ginzburg-Landau free energy has the following general form $F = F_1 + F_2 + F_{12}$ with

$$F_i = \int d^2r [A_i(T)|\eta_i|^2 + \beta_i|\eta_i|^4 + K_i|D\eta_i|^2]$$

and

$$F_{12} = \int d^2r \left[ \gamma|\eta_1|^2|\eta_2|^2 + \delta (|\eta_1|^2|\eta_2|^2 + h.c.) \right]$$

$$+ K' \sum_{\alpha=x,y} s_{\alpha} ((D_{\alpha}\eta_1)^*(D_{\alpha}\eta_2) + h.c.),$$

where we neglect the degree of freedom related to the $z$-coordinate, $s_x = +1, s_y = -1$, and $A_i(T) = a(T-T_{c1}) \ (i = 1, 2)$. The coefficients $\beta_i$, $\gamma$, $\delta$, $K_i$ and $K'$ are phenomenological parameters. The symbol $D = \nabla - 2\pi A/\Phi_0$ denotes the gauge invariant derivative where $A$ is the vector potential. In case of $T_{c1} > T_{c2}$, we find $|\eta_1|^2(T) = -A_1(T)/2\beta_1$ and $\eta_2 = 0$ for $T_{c1} > T > T^*$, while both components are finite for $T < T^*$. The temperature $T^*$ represents the renormalized transition temperature for $\eta_2$, which is defined by $A_1(T^*)/(\gamma + 2\delta \cos(2\theta)) = 2\beta_1 A_2(T^*)$. The relative phase $\theta = \phi_1 - \phi_2$ is determined by the sign of $\delta$ (we parametrize $\eta_j = u_j e^{i\phi_j}$). If $\delta > 0$ the (two-fold degenerate) low temperature state has $\theta = \pm \pi/2$ and violates $T$, i.e., it is the so-called $s + id$-state. If the superconductor has a two-component order parameter, the property of a Josephson junction is described by four coupling terms leading to the local current-phase relation

$$J = \sum_{i,j=1,2} J_{ij} \sin(\phi_{ib} - \phi_{ja})$$

with subscripts $a$ and $b$ indicating the two sides of the junction (Fig.1). On the other hand, the current density in the bulk superconductor with $\theta = \pi/2$ is given by

$$J_\alpha = \frac{4\pi c}{\Phi_0} \left[ R \left( \frac{\partial \varphi}{\partial \alpha} - \frac{2\pi}{\Phi_0} A_\alpha \right) + K' s_\alpha \sum_{i,j=1,2} \varepsilon_{ij} u_i \frac{\partial u_j}{\partial \alpha} \right],$$

where $\varphi = \phi_1 + \phi_2 + \pi/2$, $R = \sum_{i=1}^2 K_i u_i^2$, and $\varepsilon_{ij} = -\varepsilon_{ji} \ (\alpha = x, y)$. The last term gives a finite contribution only if the magnitude of the order parameter is varying in space. This is the case, in general, in the vicinity of the junction, i.e., $u_j = u_j (\vec{x} = \vec{r} \cdot \vec{n})$, where $\vec{n}$ is the normal vector of the junction (we choose $\vec{x} = 0$ on the junction). We calculate now the flux in one of the two holes by encircling it on a path $C$ starting at the junction on side $a$ and ending just
on the other side \( b \) (Fig.1). If \( C \) is far enough from the hole the current disappears and we can use Eq.(4) to evaluate the path integral of \( \nabla \varphi - 2 \pi A / \Phi_0 \) along \( C \). Assuming that the junction has no extension along \( n \) we obtain the flux

\[
\frac{\Phi}{\Phi_0} = n + \frac{\chi}{2\pi} = n + \frac{\chi_0 + \chi_1}{2\pi}
\]

(5)

with \( \chi_0(= \varphi_b - \varphi_a) \) determined from the condition of vanishing current through the junction using Eq.(3) \( (\chi_0 = \arctan[(J_{12} - J_{21})/(J_{11} + J_{22})]) \) and

\[
\chi_1 = \left[ \int_{0^+}^{\infty} + \int_{-\infty}^{0^-} \right] d\tilde{x} K' \frac{R(\tilde{x})}{\tilde{x}} (n_x^2 - n_y^2) \sum_{i,j} \varepsilon_{ij} u_i(\tilde{x}) \frac{\partial u_j}{\partial \tilde{x}}(\tilde{x}).
\]

(6)

from Eq.(4). Since the integrand is finite only near the junction we have restricted the integration to the straight line part of \( C \) on either side of the junction (Fig.1). It is easy to see that both, \( \chi_0 \) and \( \chi_1 \), vanish if the system is symmetric with respect to reflection at the junction \( (\tilde{x} \to -\tilde{x}) \) because in this case \( J_{12} = J_{21} \) and the contribution to the integrals of Eq.(6) of the two sides cancel each other. Of course, \( \chi_0 \) and \( \chi_1 \) vanish also if \( T \) is conserved \((\theta = 0 \ and \ \pi) \). However, in case that both symmetries are broken, \( \chi_0 \) and \( \chi_1 \) are finite and can have any value depending on the system parameters such that \( \Phi \) is arbitrary and non-zero for any integer winding number \( n \). The flux \( \Phi' \) in the other hole is given by the requirement that the sum of both fluxes have to be added to give an integer multiple of \( \Phi_0 \). Our treatment applies whenever the superconducting state breaks the time-reversal symmetry everywhere in the bulk or only locally at the junction. In either case there would be a transition temperature \( T^* \), lower than the onset temperature of superconductivity, above which \( T \) is conserved and the flux quantization of the holes is standard. Hence, we expect that the flux \( \Phi \) changes with temperature. In particular, for an experiment in zero external field \( \Phi \) would be zero for \( T > T^* \) and become spontaneously finite immediately below \( T^* \). Our discussion is based on the assumption that the junction is homogeneous on its entire length. Because in reality it is difficult to satisfy such a condition, we examine briefly the problem of an inhomogeneous junction where the coupling \( J_{ij} \) and the order parameter \( u_i(\tilde{x}) \) vary with the position \( \tilde{y} \) along the junction. Thus, also the two phases \( \chi_0 \) and \( \chi_1 \) depend on \( \tilde{y} \). The length scale of their variation has a lower bound at the coherence length \( \xi \) of the superconductor. If we assume that \( \chi_0(\tilde{y}) \) and \( \chi_1(\tilde{y}) \) are fixed, we may describe the inhomogeneous junction by a the well-known sine-Gordon equation for the effective phase difference \( \chi \) of the junction.

\[
\frac{\partial^2 \chi}{\partial \tilde{y}^2} = \lambda J^{-2}(\tilde{y}) \sin(\chi - \chi_0(\tilde{y}) - \chi_1(\tilde{y}))
\]

(7)

where \( \lambda J^{-2} = (c\Phi_0/16\pi\lambda)[(J_{11} + J_{22})^2 + (J_{12} - J_{21})^2]^{1/2} \) with \( \lambda \) as the London penetration depth perpendicular to the junction. The local field on the junction is given by

\[
B_z(\tilde{y}) = \frac{\Phi_0}{4\pi\lambda} \frac{\partial \chi}{\partial \tilde{y}}
\]

(8)

Therefore an inhomogeneous junction carries a distribution of magnetic field whose magnitude depends on the variation of \( \chi \). The characteristic length scale of variation for \( \chi \) is \( \lambda J \) (the screening length along the junction) which is much larger than \( \xi \). Variations of \( \chi_0 \) and \( \chi_1 \) on the length scales much shorter than \( \lambda J \) do not affect \( \chi \). Instead of following these variation \( \chi \) takes a value which is an average of \( \chi_0 + \chi_1 \) over a length of the order of \( \lambda J \). Therefore a junction which does not change its properties averaged over a length \( \lambda J \) or larger can be considered as homogeneous for our purpose. The orientation of the junction within the crystal lattice has a large influence on the boundary conditions. The d-wave order parameter is most strongly affected, if \( n \) is close to the (1,1)-direction so that in case of a \( T \)-conserving bulk d-wave supercondutor the conditions to find local \( T \)-violation are optimal here. On the other hand, we expect that the fluctuations of the junction properties are enhanced for this direction, because the Josephson
coupling for the d-wave component \((J_{11})\) is reduced. Therefore, it can be characteristic for junctions with \(n\) close to \((1, 1)\) to have a varying magnetic field distribution over their whole length. We turn now briefly to the possibility of a device with a variable flux at fixed temperature. The arrangement in Fig.1 can be modified by placing a metallic gate over the whole length of the Josephson junction (Fig.2). A voltage on the gate changes the tunnelling properties of the junction as well as the order parameter in its vicinity. Thus the quantity \(\chi_0\) and \(\chi_1\) can be tuned by the gate voltage so that a change of \(\Phi\) results according to Eq.(5). If the bulk superconducting state is time-reversal symmetric, the gate may be used to modify the junction in order to introduce a local \(T\)-violation as described in Ref.5 and 8. In principle this arrangement could be used to switch a magnetic field in the holes on and off by variation of the gate voltage. In summary, we have discussed an arrangement of holes and a Josephson junction of mesoscopic length scales, which allows one to identify a superconductor with broken time-reversal symmetry by observation of a fractional magnetic flux. Various tools like the SQUID scanning or the electron holography microscope could be applied for the detection of such fluxes. Of course, variations of this arrangement are possible. It may be simplified by expelling one of the two holes out of the superconductor such that only one hole is remaining which is then connected by the junction to an edge of the film. The holes may also be arbitrarily small. We are grateful to A. Furusaki, J.R. Kirtley, K. Kuboki, D.K.K. Lee, P.A. Lee, T.M. Rice and K. Ueda for stimulating discussions. M.S. acknowledges a fellowship financed by the Swiss Nationalfonds and Y.B.K. is supported by the NSF grant No. DMR-9022933.

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Figures Caption

**Figure 1:** Two holes (shaded) connected by a Josephson junction (dashed line) in a superconducting film. The contour \(\mathcal{C}\) from the side \(a\) to the side \(b\) is used to evaluate the flux in the hole on the right hand side.

**Figure 2:** Schematic cross-section through the junction with a gate on the top. The asymmetry of the gate ensures that the reflection symmetry is broken.