Time-reversal symmetry breaking in superconductors: A proposed experimental test

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We propose an experiment to test for broken time-reversal symmetry in the high-temperature superconductors. We configure a superconducting quantum interference device (SQUID) in a manner similar to that used recently to test for $d_{x^2-y^2}$ symmetry, but where one junction is on a crystal face of arbitrary angle. Assuming a $d_{x^2-y^2}+i\epsilon d_{xy}$ symmetry, we calculate the magnetic diffraction patterns for the SQUID for various values of ϵ and angle. For any nonzero ϵ , i.e., time-reversal symmetry breaking, we find a nonzero circulating current spontaneously arises in the SQUID even in zero applied magnetic field and zero bias current.

Among the more exotic theories of the hightemperature cuprate superconductors are those based on anyons and related gauge theories that break timereversal symmetry.¹ A variety of tests have been carried out to test for broken time-reversal symmetry in these superconductors.²⁻⁴ To date, such symmetry breaking has not been established. However, none of these tests was specifically related to the superconducting properties of these materials. Recent clarification of the form of the superconducting order parameter predicted by these gauge theories now permits construction of much more direct tests. Here we propose a generalization of the recent experiment of the University of Illinois group^{5,6} based on superconducting quantum interference devices (SQUID's) to test for d-state symmetry of the order parameter in Y-Ba-Cu-O that can in principle test for broken time-reversal symmetry. Moreover, the test provides a very direct and physically transparent example of the consequences of broken time-reversal symmetry in superconductors. Thus it is of conceptual value in its own right.

The order parameter we specifically consider in this paper has $d_{x^2-y^2}+i\epsilon d_{xy}$ symmetry. Time-reversal symmetry breaking is reflected in the fact that no gauge can be found in which this order parameter is pure real. A detailed derivation of this order parameter using anyon methods is impractical in a short paper of this kind. We shall therefore simply assert that it is implicit in the anyon description and consider the experimental consequences of its existence as an illustration of the kind of behavior allowed by a *T*-violating ground state.

Consider the dc SQUID configuration shown in Fig. 1. The junctions of the SQUID are formed on the faces of a crystal of a high- T_c superconductor. The remaining wiring is made from a low- T_c , s-state superconductor. Junction J_1 is formed on the face of the crystal at an angle θ with respect to the face containing a second junction J_2 . J_2 is taken to be on either a (100) or (010) face of the crystal, i.e., on a face normal to either the *a* or *b* axis. Photoemission data have established that the maximum of the order parameter lies along the a and b axes.⁷ The (001), i.e., c axis, is normal to the plane of the figure.

Let us now evaluate the critical current of this SQUID. Assuming that the barriers in J_1 and J_2 are identical and that the coupling process involves electrons moving primarily in the normal direction, we obtain from the relevant order parameters,

$$\Delta_1(\theta) = \Delta_0[(\cos^2\theta - \sin^2\theta) + i\epsilon(2\sin\theta\cos\theta)], \qquad (1)$$

$$\Delta_2 = \Delta_0 , \qquad (2)$$

the junction critical currents,

$$I_{J_1}(\theta) = I_0 [(\cos^2\theta - \sin^2\theta)^2 + \epsilon^2 (4\sin^2\theta \cos^2\theta)]^{1/2}, \qquad (3)$$

$$I_{J_2} = I_0 , \qquad (4)$$

where θ is the wedge angle shown in Fig. 1. From Kirchhoff's current law and the requirement that the superconducting order parameter be single valued around the SQUID loop, we then obtain the standard equations for a dc SQUID:



FIG. 1. Configuration of high- T_c -s-wave superconducting dc SQUID.

$$I = I_{J_i} \sin \phi_1 + I_{J_2} \sin \phi_2 , \qquad (5)$$

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$$\phi_1 - \phi_2 + \frac{2\pi\Phi}{\Phi_0} + \delta(\theta) = n 2\pi , \qquad (6)$$

where $\phi_{1,2}$ are the gauge-invariant macroscopic quantum phase differences across J_1 and J_2 , Φ is the magnetic flux in the loop, and

$$\delta(\theta) = \tan^{-1}(\epsilon \tan 2\theta) \tag{7}$$

is the phase difference across the high- T_c superconductor due to the assumed *d*-state symmetry of the order parameter. In the University of Illinois experiment, $\theta = \pi/2$ and therefore $\delta = \pi$. When both superconductors have *s*state symmetry, $\delta = 0$. The dependence of δ on θ is shown graphically in Fig. 2(a) for various values of ϵ .

For the particular case $\epsilon = 1$, $I_{J_1} = I_{J_2} = I_0$, independent of θ , and Eqs. (5) and (6) can be solved analytically in the limit of weak shielding (i.e., $I_0L \ll \Phi_0$, where L is the inductance of the SQUID) for the net critical current through the SQUID as a function of the phase difference δ and the applied flux Φ_a . The result is

$$I_c = 2I_0 \left| \cos \left[\frac{\pi \Phi_a}{\Phi_0} + \frac{\delta}{2} \right] \right| \tag{8}$$

which is shown in Fig. 3 for $\theta=0$, $\pi/4$, and $\pi/2$. The case $\theta=\pi/4(\delta=\pi/2)$ is particularly interesting because it corresponds to the angle at which $\Delta(\theta)$ is pure imaginary. We note that in this case the so-called magnetic diffraction pattern $I_c(\Phi_a)$ is asymmetric. This asymmetry is a consequence of the broken time-reversal symmetry. For $\delta=\pi$, as in the University of Illinois experi-



FIG. 2. (a) Phase difference across the high- T_c crystal vs the angle of the first junction. This is plotted as a function of ϵ , the amount of id_{xy} symmetry in order parameter. (b) Calculated SQUID loop current vs the angle of the first junction, as a function of ϵ .

ment, the curve is shifted by $\Phi_a/\Phi_0 = \frac{1}{2}$ but is still symmetric. This is because this result is only sensitive to the real part of the order parameter in Eq. (3), which has a sign change but does not break time-reversal symmetry.

An even more striking consequence of the broken-time reversal symmetry is seen by calculating the circulating loop current I_L in the SQUID in the absence of any bias current or applied flux

$$I_L(I=0,\Phi_a=0,\epsilon=1)=I_0\sin\theta.$$
 (9)

Hence a spontaneous circulating current arises in the SQUID due to broken time-reversal symmetry for any $0 < \theta < \pi/2$. Note that I_L in Eq. (9) is indeterminate at $\theta = \pi/2$.⁸ For $\theta = \pi/4$, $\delta = \pi/2$ for all ϵ , and it is easy to show that $I_L(\theta = \pi/4) = \epsilon/(1 + \epsilon^2)^{1/2}$. For general values of ϵ , Eqs. (3) to (6) must be solved numerically. The results are shown in Figs. 2 and 3.



FIG. 3. Critical current of the SQUID versus applied flux, for (a) $\theta = 0$, (b) $\theta = \pi/4$, and (c) $\theta = \pi/2$. In the $\theta = 0$, $\pi/2$ cases, the critical current is unaffected by ϵ . For $\theta = \pi/4$, the curves are shown for various values of ϵ .

We emphasize that the spontaneously generated supercurrents described here are fundamentally different from those generated in rings containing so-called π junctions, and proposed as an explanation of the Wohlleben effect.^{6,9} In that case broken time-reversal symmetry arises only when the inductance of the SQUID is large. The symmetry breaking here occurs for junctions in the small inductance limit and for any $0 < \theta < \pi/2$, and $\epsilon \neq 0$.

We conclude with some observations relevant to experiment. The results obtained here do not depend on there being a true energy gap in the $\theta = \pi/4$ direction, i.e., along the nodes of a $d_{x^2-y^2}$ symmetry order parameter. It is well known that pair breaking can eliminate an energy gap but not destroy the order parameter. The results do depend on the Josephson coupling being predominantly due to electrons moving normal to the junction. Coupling processes that involve electrons with transverse momentum will tend to reduce the Josephson coupling due to the effect of lobes of the order parameter of opposite sign. Nonidentical barriers change the results only quantitatively. We have not considered the possibility of domain formation.

The execution of this experiment is not easy at present

due to the difficulty in cleaving, polishing or patterning a crystal at an angle θ between 0° and 90°, such that the crystal face is not composed of many facets at angles not equal to θ locally.

Since twinning is noted by Wollman *et al.* not to affect the results of their experiment, we propose that *c*-axis thin films (which are known to have 90° twins) can be used. As long as the twin boundary is not a weak link the order parameter symmetry should be coherent across the twin. Characterization of the angled face via microscopy is very important in this experiment to determine the directional coupling of the order parameter.

In the likely situation that the barriers are different for the two junctions due to geometry or scattering considerations, the result in the $\theta = \pi/4$ case [Fig. 2(b)] will be simply to affect the magnitude of the modulation but not the offset of the maximum. The situation is qualitatively similar for $\theta \neq \pi/4$.

This work was supported by the NSF-MRL program and the AFOSR. The authors thank Aharon Kapitulnik for helpful remarks.

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FIG. 1. Configuration of high- T_c -s-wave superconducting dc SQUID.