Broken time-reversal symmetry in a SQUID based on chiral superconducting Sr$_2$RuO$_4$

R. Ishiguro$^{1,2}$, T. Sakurai$^2$, M. Yakabe$^2$, T. Nakamura$^{3,4}$, S. Yonezawa$^1$, S. Kashiwaya$^3$, H. Takayanagi$^{2,6}$, and Y. Maeno$^3$

$^1$Center for Emergent Matter Science, RIKEN, Wako, Saitama 351-0198, Japan
$^2$Department of Applied Physics, Faculty of Science, Tokyo University of Science, Tokyo 125-8585, Japan
$^3$Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan
$^4$Institute for Solid State Physics, the University of Tokyo, Chiba 277-8581, Japan
$^5$National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8568, Japan
$^6$International Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), Tsukuba 305-0044, Japan

ryosuke.ishiguro@riken.jp

Abstract. Unconventional superconductors involve not only gauge-symmetry breaking but also orbital- and spin-symmetry breaking. Superconducting Sr$_2$RuO$_4$ is known as a spin-triplet chiral $p$-wave and a topological superconductor with broken time-reversal symmetry (BTRS). Kerr-effect and muon-spin-rotation ($\mu$SR) measurements have shown that the bulk superconducting state of Sr$_2$RuO$_4$ features BTRS in the orbital part; hence, it is called the chiral state. BTRS in the response of superconducting junctions or SQUIDs would appear as the shifts of magnetic interference patterns. However, it is problematic to distinguish whether the shift originates in the residual magnetic field (trapped vortex) or the effects of BTRS. Here, we show that the magnetic interference patterns of a SQUID based on Sr$_2$RuO$_4$ are explicitly asymmetric with respect to the direction of both the bias current and the applied magnetic field; namely, there is no inversion symmetry. This indicates that the superconducting state of Sr$_2$RuO$_4$ undoubtedly breaks the time-reversal symmetry of the SQUID.
1. Introduction

The BCS superconducting transition is associated with gauge-symmetry breaking at the transition temperature $T_c$, above which the system is invariant under gauge transformation. Unconventional condensates, such as superfluid $^3$He, $p$-wave superconductors, and $d$-wave superconductors, are more complicated, as they include both spin and orbital degrees of freedom. Therefore, the transition involves not only gauge-symmetry breaking but also orbital- and spin-symmetry breaking. In these superconducting states, time-reversal symmetry can be spontaneously broken.

Superconducting Sr$_2$RuO$_4$ is known as a strong candidate for a spin-triplet chiral $p$-wave and a topological superconductor with broken time-reversal symmetry (BTRS) [2-4]. A BTRS state arising from the possible magnetization of the orbital component of the superconducting wave function of the chiral $p$-wave superconducting Sr$_2$RuO$_4$ has been decisively demonstrated by Kerr-effect and muon-spin-rotation ($\mu$SR) measurements [5, 6]. Associated with the bulk BTRS of a topological superconductor, it is expected that its edge, namely its surface state, also breaks TRS as dictated by the bulk-edge correspondence, there is also a possibility of BTRS originating in the phase winding of the chiral superconducting wave function around the edge of the chiral superconductor [3, 7-10]. The wave function phase at the edge of the chiral superconductor is continuously changing with the angle of the surface normal vector [3, 11]. Therefore, superconducting hybrid junctions, or rings containing the superconducting junctions, consisting of $s$-wave and chiral superconductors have the possibility of exhibiting BTRS with a spontaneous supercurrent [3, 7-10]. The Josephson current through a superconducting junction is ordinarily proportional to $\sin \phi$, where $\phi$ is the phase difference between two superconductors separated by the junction. However, if BTRS exists, the Josephson current is represented by a linear combination of $\sin \phi$ and $\cos \phi$ [12].

A number of transport- and phase-sensitive measurements have been conducted on $p$- and $d$-wave superconductors to confirm the presence of BTRS [7, 9, 10, 13-16]. However, it is difficult to establish the existence of BTRS because of the subtle differences between the shifts originating in the residual magnetic field (trapped vortex) and those caused by the effects of BTRS. Here, we present the magnetic interference patterns of a SQUID consisting of a hybrid superconducting loop consisting of the $s$-wave superconductor niobium (Nb), Sr$_2$RuO$_4$, and Nb/Ru/Sr$_2$RuO$_4$ junctions. The observed interference patterns are explicitly asymmetric with respect to the direction of both the bias current and the applied magnetic field. This asymmetry clearly demonstrates the existence of BTRS.

2. Experimental methods

We fabricated hybrid Nb–Sr$_2$RuO$_4$ dc SQUIDs, which are composed of Nb, Sr$_2$RuO$_4$, and two Nb/Ru/Sr$_2$RuO$_4$ junctions, by photolithographically building a Nb bridge between the two individual Ru islands on the $ab$-plane surface of eutectic Sr$_2$RuO$_4$-Ru shown in Fig.1 [1, 17]. The Sr$_2$RuO$_4$-Ru eutectic is known to develop a filamentary superconducting phase around the Ru islands for temperatures below about 3K, which is twice the transition temperature of bulk Sr$_2$RuO$_4$ ($T_c$-bulk = 1.5 K) [17]. The Nb/Ru/Sr$_2$RuO$_4$ junctions, which were made by depositing an $s$-wave superconductor on Ru metal in eutectic Sr$_2$RuO$_4$-Ru, are known to behave as “topological junctions” [11, 18]. The measurements reported herein were performed at temperatures well below $T_c$-bulk. Our purpose in using the eutectic material is to establish good electrical contact between the $s$-wave and chiral $p$-wave superconductors in order to exploit the proximity effect. Finally, the dc-SQUID shape was formed by the focused ion beam technique (FIB) to restrict the supercurrent path in the Sr$_2$RuO$_4$. The details of the fabrication of similar-type SQUIDs with Nb/Ru/Sr$_2$RuO$_4$ junctions and of the herein presented SQUID have been described elsewhere [19] and in a very recently submitted paper[1]. The relative angle between the two Nb/Ru/Sr$_2$RuO$_4$ junction surfaces was approximately $\pi$. In this angle condition, only $\pi$ SQUID behavior is expected [8] and not BTRS. However, we observed BTRS behavior but no $\pi$ SQUID features. We consider that this is related to the existence of a small number of chiral domains, each of which is a superconducting regions with the same chirality.

For the measurements, a helium-3 evaporative refrigerator with a base temperature of 0.3 K was used.
3. Results and discussion

Figure 2 shows magnetic interferences pattern for the SQUID for both a positive and negative current at 0.6 K. The details of these interference patterns will be presented elsewhere[1]. It must be noted that these interference patterns are explicitly asymmetric with respect to the directions of both the bias current and the applied magnetic field; that is, there is no inversion symmetry. Usually, BTRS in hybrid SQUIDs or junctions shifts the origin (zero magnetic field) of the interference patterns [7-10]. However, it is difficult to distinguish between external residual magnetic fields and the intrinsic effects of BTRS from this shift. In the present case, the asymmetry of the observed interference pattern cannot be explained only by the shift of the origin of the external magnetic field. Therefore, this asymmetry cannot be due to the influence of the magnetic field formed by trapped flux or the test current but must be caused by the existence of \( \cos \phi \) components in the Josephson current due to BTRS. Therefore, the asymmetry of the interference pattern constitutes strong evidence of the expected BTRS originating in the wave function phase winding of the chiral \( p \)-wave superconductor \( \text{Sr}_2\text{RuO}_4 \).

This reproducibility of this asymmetric interference pattern is very good; the pattern does not depend on the direction of the field sweep. In contrast, the asymmetric patterns slightly change upon thermal cycling above the superconducting transition temperature, as shown in Fig. 3. We assume that this change is influenced by the reconstruction of the chiral domains [1, 7, 18, 20]. Since it is difficult to explain BTRS only by the relative angle of two junctions, \( \pi \), we believe that the origin of the displayed BTRS is related to the relative position and phase of the chiral domains present in the \( \text{Sr}_2\text{RuO}_4 \) part of the SQUID.

**Figure 1.** (a) SEM image of the SQUID. (b),(c) A possible configuration of the chiral domain in the SQUID.
Figure 2. Evidence of broken time-reversal symmetry. The positive and negative critical currents of SQUID are asymmetric. The asymmetries increase with decreasing temperature. The different colors indicate the field sweeping directions, as shown by the arrows.

Figure 3. Thermal cycling of the magnetic interference patterns of SQUIDs at 0.6 K above the superconducting transition temperature, measured on different days[1].
4. Conclusion
We fabricated a hybrid SQUID based on the chiral p-wave superconducting Sr$_2$RuO$_4$ and an s-wave superconductor and showed that the hybrid SQUID features BTRS originating in the phase winding of the chiral superconductor and certainly breaks the time-reversal symmetry.

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