Superconducting transition of Ru in SQUIDs with Nb/Ru/Sr$_2$RuO$_4$ junctions

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Abstract. We have measured transport properties and magnetic responses of direct current superconducting quantum interference devices (dc-SQUIDs) with Nb/Ru/Sr$_2$RuO$_4$ junctions on the same Ru inclusion of a Sr$_2$RuO$_4$-Ru eutectic crystal. We observed a superconducting transition of the Ru at 0.50 K and another transition with anomalous interference patterns of the SQUID at 0.40 K. The second transition led to change of the SQUID structure due to energy competition of alternative supercurrent paths. Phase shifts of the peak of the Fraunhofer diffraction pattern were observed below the transition temperatures, implying topological effects due to a chiral p-wave state or its competition with a s-wave state.

1. Introduction

Layered perovskite Sr$_2$RuO$_4$ is one of strong candidates of a spin-triplet chiral p-wave superconductor [1–3]. In recent ten years, Ru-Sr$_2$RuO$_4$ eutectic crystal has attracted research interest because it exhibits an inhomogeneous superconducting phase at 3 K (3-K phase), different from the bulk phase which appears at 1.5 K [4]. A theoretical research [5] shows that a 3-K phase nucleates as a state conserving time reversal symmetry at the interface with Ru inclusion and evolves into a state breaking time reversal symmetry with decreasing temperature. A number of transport measurements [6–9] revealed novel properties of 3-K phase around Ru inclusions, following the theoretical prediction. However the generation mechanism of this enhanced superconductivity has not yet been completely identified [10].

The eutectic crystal is also useful for investigation of a competition between a s-wave state of Ru and a p-wave state of Sr$_2$RuO$_4$ at sufficiently low temperatures below the superconducting transition temperature $T_c^{Ru}$ of Ru inclusion (0.49 K); it is helpful to understand a superconducting state of a 3-K phase as well as a bulk phase [11]. A topological mismatch of s- and p-wave phases in the eutectic crystal is expected to cause Josephson vortex nucleation in Ru inclusion [12]: a frustrating coupling of the order parameters of both states induces
a spontaneous magnetic flux distribution at the interface of Ru inclusion, growing up with decreasing temperature to form a single vortex state at the Ru center.

A topological superconducting junction can be easily fabricated using the eutectic crystal and an s-wave superconductor [8,9]. We have so far fabricated dc-SQUIDs with Nb/Ru/Sr$_2$RuO$_4$ junctions using Ru-Sr$_2$RuO$_4$ eutectic crystal [13] and measured its transport properties [14]. In this study, a dc-SQUID whose two junctions are composed on the same Ru inclusion was prepared. We measured a superconducting transition of the Ru inclusion and magnetic responses of the SQUIDs below the transition in order to investigate the effects due to a topological mismatch of s- and p-wave phases. We discuss anomalous interference patterns of the SQUIDs observed below the transition.

2. Experimental
The dc-SQUIDs consist of Nb, Sr$_2$RuO$_4$ and two Nb/Ru/Sr$_2$RuO$_4$ junctions on the ab-plane surface of a Sr$_2$RuO$_4$-Ru eutectic crystal, cut from a crystalline rod grown by a floating-zone method [4]. This device was fabricated by the same technique as used in previous studies [13, 14]: the eutectic crystal is coated with an SiO$_2$ layer of 300-nm thickness and two small windows are made on the small parts above Ru inclusion for contacting a niobium electrode using a photo lithography and a reactive ion etching. The structure of the device is similar to one in previous studies [13, 14], except for the point that two junctions are located on each edge of the same Ru inclusion as shown in Fig. 1. A size of the Ru inclusion is about 1 $\mu$m $\times$ 20 $\mu$m and hence the distance of junctions is about 20 $\mu$m. Above the Ru superconducting transition $T_{c}$Ru, a SQUID loop should be formed so that a current flows on Sr$_2$RuO$_4$ in the vicinity of the Ru-Sr$_2$RuO$_4$ interface, for example, as indicated by a red line in Fig. 1 (b). Therefore it is expected that the effective SQUID-loop area consists of SiO$_2$ cover area surrounded by the Nb bridge, some part of the Ru inclusion area and the penetration depth $\lambda$ of a magnetic flux into Nb and Sr$_2$RuO$_4$ which is order of 100 nm at sufficiently low temperatures below 1 K, as seen in Fig. 2 (a). Below $T_{c}$Ru, on the other hand, the junctions change from S(s-wave)-N-S(p-wave) to S(s-wave)-S(s-wave)-S(p-wave) and a supercurrent is expected to flow on the whole Ru inclusion, forming a superconducting ring with Nb/Ru(s-s) junctions and a single Ru/Sr$_2$RuO$_4$(s-p) junction, as seen in Fig. 2 (b).

For four-terminal measurements, two of the four Nb electrodes with a thickness of 600 nm are directly linked to the SQUID line as seen in Fig. 1 and the others deposited on the other Ru inclusions (see Ref. [13, 14]). The device was placed in a dilution refrigerator and cooled below 1 K. We measured IV characteristics as shown in Fig. 3 and estimated the Josephson maximum current $I_c$ as a function of external magnetic field at each temperature. Since $I_c$ depending on a superconducting gap $\Delta(T)$ increases up to order of mA at sufficiently low temperatures as seen in Fig. 3, a considerably large current needs to be driven. In that case, heat generated by the current at the junctions and electrical wires made of a normal metal near the device is an inevitable problem; we actually performed continuous measurements of IV characteristics at each temperature and found a gradual decrease of $I_c$ due to heating despite the temperature is
Figure 2. Schematics of flowing path of a current on the SQUID. (a) above the superconducting transition temperature of $T_{Ru}^c$. (b) below $T_{Ru}^c$, the case in which a path on superconducting Ru is more favorable. (c) below $T_{Ru}^c$, the case in which a path through two junctions is more favorable. Crosses indicate superconducting junction.

Figure 3. Current-Voltage characteristics of the SQUID at 0.55 K and zero magnetic field. Josephson maximum current $I_c$ is determined alternatively as the current indicated in $I^+_c$ and $I^-_c$ where a finite voltage suddenly appears in each current direction.

set constant. Therefore we verified effects of heating with continuous measurements of $I_c$ and finally fixed measurement intervals of 5 min - 15 min, depending on temperature.

3. Results and discussion
Figure 4 shows the Josephson maximum current $I^+_c$ estimated from IV characteristics (Fig. 3) as a function of external magnetic field $B$ at 0.55 K. Periodic patterns can be seen in a Fraunhofer diffraction pattern and its period $\Delta B_0$ is approximated to be 0.1 mT, which corresponds to a flux quantum $\Phi_0$ divided by the effective SQUID-loop area. Though the precise depth of the Ru inclusion is unidentified, it is speculated that only a part of the Ru near its surface contributes to the effective loop area shown in Fig. 2 (a) because the Ru is surrounded by Sr$_2$RuO$_4$ as shown in Fig. 1. Therefore, assuming $\lambda$ of Nb and Sr$_2$RuO$_4$ to be 100 nm, the effective loop area is approximated to be $\sim 0.5 \mu m \times 20 \mu m$, consistent with the observed period $\Delta B_0$.

The field at the maximum of $I^+_c$ shifts in the negative direction from $B = 0$ mT as seen in Fig. 4. We found that the field at the minimum of $I^-_c$ in Fig. 3 is also shifted by almost the same amount of field in the positive(opposite-sign) direction. These results indicate that a self-induced flux by a bias current causes this peak shift.

Figure 5 shows temperature dependence of SQUID diffraction patterns at 0.55 K - 0.35 K. Josephson maximum current $I^+_c$ gradually increases with decreasing temperature, due to a temperature dependence of energy gap of both superconductors at the junction. A period $\Delta B_0$ of the SQUID pattern also shows a very weak temperature dependence indicating gradual decrease of the effective SQUID-loop area which magnetic fluxes thread through. This can be attributed to a temperature dependence of a penetration depth $\lambda$. However we found the excess increase of $\Delta B_0$ below 0.50 K, whose increase rate is approximately 30 % from 0.50 K to 0.40 K. When Ru undergoes a superconducting transition, the Meissner screening at the superconducting Ru area reduces the effective SQUID area. Therefore the large increase of a period evidently indicates that a superconducting transition of the Ru inclusion occurs at about 0.50 K.

A junction structure changes from S-N-S to S-S-S at $T_{Ru}^c$ as mentioned in the section 2 and the system is likely to form series circuits with a superconducting ring with Nb/Ru(s-s) junctions and a single Ru/Sr$_2$RuO$_4$(s-p) junction as seen in Fig. 2 (b). It is not clear of which junction $I_c$
is smaller (more detectable) just below $T_{c\text{Ru}}$; in either case, the proximity effect at the junction is expected to become weak compared with the case of the original S-N-S junction, because of a small gap of the Ru near $T_{c\text{Ru}}$. If it is the case, $I_c$ should decrease according to decrease of a tunneling rate through the junction. However $I^+_c$ seems to increase noticeably below $T^R_{c\text{Ru}} = 0.50$ K as seen in Fig. 5. This result suggests that a coupling of superconducting pair functions at the original Nb/Ru/Sr$_2$RuO$_4$ junction is still strong: a superconducting path through the Ru inclusion newly appears between two junctions at the Ru transition but, just below $T^R_{c\text{Ru}}$, a critical current of this thin superconducting path on the Ru is too small to be energetically more favorable than the paths to bulk Sr$_2$RuO$_4$ through the two original junctions. This scenario means that the system forms a SQUID with Nb/Ru/Sr$_2$RuO$_4$(s-s-p) junctions at 0.50 K, i.e., the SQUID structure changes from (a) not to (b) but to (c) shown in Fig. 2. A transition from S-N-S to S-S-S junction implies that a pair function of superconducting Ru strengthens a coupling between superconducting states of Nb and Sr$_2$RuO$_4$ and consequently $I^+_c$ increases, consistent with our present result.

A critical supercurrent of Ru inclusion increases with decreasing temperature and may exceed the Josephson maximum current at the Nb/Ru/Sr$_2$RuO$_4$ junctions at a certain temperature, resulting in change of the supercurrent path from Fig. 2 (c) to (b). One can find that the periodic SQUID pattern starts to collapse below about 0.40 K as seen in Fig. 5. This anomalous characteristics were not observed for the previous SQUID with two junctions connected on two different Ru inclusions [13, 14]. It is therefore plausible that the SQUID undergoes the second transition at which a SQUID structure changes into a Nb/Ru superconducting ring with a single Ru/Sr$_2$RuO$_4$ junction shown in Fig. 2 (b). Furthermore, the field value of the peak of the Fraunhofer pattern largely shifts in the positive direction below about 0.40 K as seen in Fig. 5. Such large shifts were never observed at temperatures higher than 0.40 K. Magnetic fluxes trapped in holes, defects or impurities in a superconductor cause a large Fraunhofer-peak shift [15]. In this case, $I_c$ modulation would show hysteresis between up and down sweeps of applied field, and $I^+_c$ and $I^-_c$ modulations obtained with each current direction would show the Fraunhofer peak of which the field shifts in the same sign direction. In contrast, our results revealed no hysteresis and the negative shift of $I^-_c$-modulation peak which is opposite to the case of $I^+_c$, indicating different mechanisms. The peak shift dependent on field direction implies effects of a magnetic flux induced by a bias current due to an asymmetric structure of the SQUID. The structure of our present sample seems almost symmetric in the case of Fig. 2 (c) but can become
asymmetric in the case of Fig. 2 (b) if the supercurrent flows asymmetrically only through a particular part of the Ru/Sr$_2$RuO$_4$ junction. These pictures are consistent with our results that the large peak-shift was observed only below 0.40 K; in other words it can be an evidence that structure of the SQUID changed from (c) to (b) at 0.40 K. The asymmetric current flow at the Ru/Sr$_2$RuO$_4$ (s-p) junction might be caused by a spontaneously generated current as a result of competition of s- and p-wave states, which possibly leads to a Josephson vortex nucleation [12]. However it should be noted that topological properties of Sr$_2$RuO$_4$ such as odd-parity pairing symmetry [16] or chiral domains [7, 9] can also cause phase shifts in the interference pattern. Further theoretical and experimental approaches are needed for clarification.

4. Conclusion
We fabricated dc-SQUID with Nb/Ru/Sr$_2$RuO$_4$ junctions on the same Ru inclusion of Ru-Sr$_2$RuO$_4$ eutectic crystals and observed two transitions in the magnetic interference patterns of the SQUID at very low temperatures. One corresponds to the superconducting transition of the Ru inclusion at 0.50 K, which changes the effective SQUID-loop area due to Meissner screening, resulting in an increase of a period of the interference patterns. It also strengthens a coupling of pairing states at the junction, resulting in an increase of a Josephson maximum current. The other is the collapse of the periodic pattern of the SQUID and its phase shifts at 0.40 K, where a supercurrent path changes due to the energy balance between a Josephson current through the junction and a critical supercurrent through the Ru inclusion. The anomalous phase shifts below 0.40 K raise a possibility of effects due to topological properties of a chiral p-wave state or topological mismatch between p- and s-wave phases.

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