Magnetization measurements of Sr$_2$RuO$_4$-Ru eutectic microplates using dc-SQUIDs

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Abstract. We report magnetization measurements of Sr$_2$RuO$_4$-Ru eutectic microplates using micro-dc-SQUIDs. Sr$_2$RuO$_4$ is considered as a chiral p-wave superconductor and hence Sr$_2$RuO$_4$-Ru eutectic becomes in an unstable state with a superconducting phase frustration between a chiral p-wave state of Sr$_2$RuO$_4$ and a s-wave state of Ru. To compensate the frustration, a single quantum vortex is spontaneously formed at the center of the Ru inclusion at sufficiently low temperatures. However, such a spontaneous vortex state has not been experimentally observed yet. In this study, we prepared a micro-dc-SQUID and a Sr$_2$RuO$_4$-Ru eutectic microplate containing a single Ru-inclusion at the center of the microplate. We performed magnetization measurements down below the superconducting transition temperature of the Ru inclusion to investigate the spontaneous Ru-center vortex state.

1. Introduction
Unconventional superconductivity is one of attractive subjects because it exhibits orbital- and spin-symmetry breaking [1]. Among unconventional superconductors, Sr$_2$RuO$_4$(SRO) is the strong candidate of a spin-triplet chiral p-wave pairing with broken time reversal symmetry in the orbital state [2, 3, 4]. This property exhibits a continuous phase winding from 0 to 2$\pi$ following the momentum vector around $p_z$ axis. SRO-Ru eutectic crystal [5] is useful for research for chiral superconductivity of SRO because the Ru inclusion is surrounded by the SRO: the superconducting SRO shows a phase winding along the SRO/Ru interface due to its chiral property while the superconducting Ru has a constant phase, leading to a topological phase frustration between a s-wave state of the Ru and a chiral p-wave state of the SRO at the interface [6, 7, 8, 9]. The frustration causes a spontaneous supercurrent flowing through the interface to...
Figure 1. (a) Electron micrograph of the magnetization device. (b)(c) Top and side views of the device. The plus and minus signs in (b) indicates a flux direction in the Ru-center vortex state, corresponding to the arrows in (c).

minimize the interface free energy [10, 11]. With decreasing temperature, the spontaneous-current profile gradually changes according to growth of the frustrating coupling of both states, finally forming a single vortex state at the center of a Ru-inclusion [10]. The Ru-center vortex accompanies a quantized flux distributed along the SRO/Ru interface to compensate the phase frustration (see Fig.1(c) in Ref.[10]). Therefore detection of the spontaneous Ru-center vortex state can be one of evidences of chiral superconductivity of SRO. However, it has not been experimentally observed yet.

We have so far fabricated dc-SQUIDs with Nb/Ru/SRO junctions using SRO-Ru eutectic crystal and investigated the effects due to a topological mismatch of s- and chiral p-wave phases [9, 12]. However, an evidence of the Ru-center vortex state has not been obtained. Investigation of the vortex state requires a simpler high-sensitivity device to detect a magnetic flux directly. Recently, we have succesfully developed magnetization measurement device composed of a micro-dc-SQUID and a SRO microplate [13]. This device is useful to detect the magnetic flux induced on the SRO-Ru eutectic microplate. In this study, We prepared a micro-dc-SQUID and a SRO-Ru eutectic microplate with one Ru inclusion and performed magnetization measurements below the superconducting transition temperature of the Ru inclusion.

2. Experimental
Figure 1 exhibits the magnetization measurement device composed of the SRO-Ru eutectic microplate and the micro-dc-SQUID. A SRO-Ru eutectic crystal was fabricated into a micrometer-sized square plate with a focused Ga-ion beam (FIB) technique, based on our previous studies [13, 14]. The microplate was milled so that a cylindrical-shaped Ru inclusion was located nearly in the center of the plate as seen in Fig. 1 (a). Diameter of the Ru inclusion was less than 1µm. We checked that a depth of proximal Ru-cylinders in our eutectic crystal is larger than 725 nm,
corresponding to a height of our present microplate, and hence we sure that the Ru-inclusion cylinder in the microplate reaches the bottom.

The micro-dc-SQUID consists of Al/AlOx/Al tunnel-type Josephson junctions. The design of the SQUID was typically the same as those used in a previous study [13], wherein the Josephson junctions were located at each corner of the SQUID loop to prevent any undesirable magnetic flux from intruding into the SQUID electrodes. The electrode width was 100 nm and the inner square loop length was 1 μm x 1 μm. An SiO2 mask 100 nm thick was sputtered onto the SQUIDs to prevent the Ga ions from reaching the SQUIDs during the FIB process.

The Ru-center vortex state consists of a magnetic flux quantum Φ0 at the Ru center and a quantized flux distributed along the SRO/Ru interface [10]. The direction of the vortex flux at the center of the Ru is opposite to that of the flux at the interface, satisfying flux conservation. Therefore a flux circulates in a very small scale in the vicinity of the plate surface as shown by the arrows and the signs in Figs 1 (b) and (c). This makes it hard to detect a flux induced by the vortex. In this study, the milled eutectic microplate was picked up using a Tungsten nano-probe of a nano-manipulator system and directly mounted onto the SiO2-masked SQUID so that the center of the microplate was slightly displaced from the center of the SQUID loop, as in Figs 1 (b) and (c). This condition is expected to enable detection of the vortex state.

The device was cooled down below 1 K with a dilution cryostat holding an applied magnetic field H at zero. We measured current-voltage characteristics and subsequently a Josephson critical current Ic as a function of the field applied along c-axis of the microplate. We also measured a SQUID without a eutectic as a reference, separated by about 40 μm from the SQUID with the eutectic microplate.

3. Results and discussion

Figure 2 shows the Josephson critical current Ic of the SQUID with the eutectic microplate as a function of an external magnetic field μ0H at each temperature. The depicted field range is lower than the field of 3.3 mT where a quantum vortex intrusion into the microplate was observed. The modulation period for the reference SQUID was measured to be 1.71 mT at 0.04 K, while the period of the SQUID with the eutectic microplate was estimated to be about 2 mT at all temperature range as seen in Fig. 2. This difference is attributed to be the exclusion of the magnetic flux from the loop of the SQUID owing to the Meissner effect of the superconducting SRO-Ru. The period ΔB and the offset Boff of the modulation patterns were estimated from fitting the data in Fig 2 by

\[ I_c \propto \cos[\pi(\Phi - \Phi_{\text{off}})/\Phi_0] = \cos[\pi(\mu_0 H - B_{\text{off}})/\Delta B], \]

where a flux density B is obtained from a flux threading the SQUID Φ divided by the effective SQUID-loop area. Fig. 3 exhibits a temperature dependence of ΔB and Boff. ΔB monotonically increases with decreasing temperature, which means a partial exclusion of a flux out of the SQUID loop. An external flux penetrates the eutectic microplate by the penetration depth $\lambda^{\text{SRO}}_c$ in good which is temperature dependent [9, 15]. The decrease of the depth with decreasing temperature leads to the exclusion of the flux, qualitatively in good agreement with the behavior in the lower panel of Fig. 3. However, one can see a kink at about 0.425 K. This indicates that the Ru-inclusion undergoes the superconducting transition at $T_{\text{cRu}} = 0.425$ K and subsequently the flux was pushed out further. In other words, a temperature dependence of the penetration depth of the Ru $\lambda^{\text{Ru}}_c$ reflects the excess increase of ΔB below $T_{\text{cRu}}$.

In contrast, the upper panel in Fig. 3 shows no remarkable change of Boff below $T_{\text{cRu}}$. The offset Boff includes contribution of spontaneous magnetization. Therefore, if the Ru-center vortex state exists below $T_{\text{cRu}}$, the SQUID should detect the excess flux induced by the vortex, resulting in a shift on the temperature dependence of Boff. According to our numerical simulation of magnetic flux distribution solving Maxwell and London equations using the commercial package COMSOL (the detail will be described elsewhere [16]), the total flux by the Ru-center vortex detectable for the SQUID was estimated to be 0.09Φ0 in our present device geometry.
Figure 2. Josephson critical current $I_c$ as a function of external magnetic field at 0.8 K - 0.04 K.

Figure 3. Temperature dependence of the period $\Delta B$ (lower panel) and the offset $B_{off}$ (upper panel) of the $I_c$ modulation. The broken line indicates the temperature corresponding to a kink in the lower panel.

However, our present result in Fig. 3 shows almost constant $B_{off}$ and the overall shift of $B_{off}$ at the measured temperature range corresponds to be less than 0.001$\Phi_0$. Therefore we conclude that the spontaneous Ru-center vortex state does not exist in our present eutectic microplate.

Our result showing absence of the Ru-center vortex state implies a weak coupling of proximity-induced superconductivity between SRO and Ru. A phase frustration between SRO and Ru superconducting states causes a spontaneous supercurrent at the SRO/Ru interface [10, 11]. If the superconducting coupling at the SRO/Ru interface is weak, the interface free energy in the spontaneous current state becomes smaller than that in the Ru-center vortex state and hence the Ru-center vortex state cannot be stably formed [10]. This feature at the SRO/Ru interface is consistent with the previous tunneling measurement on an SRO/Ru/Au junction which revealed suppression of the superconducting gap of the SRO in the Ru-inclusion [6].

We note that possibilities of non-chiral superconductivity of SRO or influence of a chiral domain state cannot be ruled out yet. Clarification of a magnetic flux distribution in the eutectic microplate and a phase frustration of both superconductivity requires further theoretical and experimental approaches.

4. Conclusion

We measured magnetization of Sr$_2$RuO$_4$-Ru eutectic microplates using micro-dc-SQUIDs at temperatures down below the superconducting transition temperature $T_{c}^{Ru}$ of the Ru inclusion. With decreasing temperature, the magnetic flux is gradually excluded from the microplate according to decrease of the penetration depth of superconducting Sr$_2$RuO$_4$ and pushed out further below $T_{c}^{Ru} = 0.425$ K owing to the Meissner diamagnetism of the superconducting Ru. Our present results show that spontaneous magnetization such as a Ru-center vortex state is
absent in the present microplate, implying weak proximity-induced superconductivity at the Sr$_2$RuO$_4$/Ru interface.

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