Anomalous Transport through the \( p \)-Wave Superconducting Channel in the 3-K Phase of \( \text{Sr}_2\text{RuO}_4 \)

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Using microfabrication techniques, we extracted individual channels of 3-kelvin (3-K) phase superconductivity in \( \text{Sr}_2\text{RuO}_4\)-Ru eutectic systems and confirmed odd-parity superconductivity in the 3-K phase, similar to pure \( \text{Sr}_2\text{RuO}_4 \). Unusual hysteresis in the differential resistance-current and voltage-current characteristics observed below 2 K indicates the internal degrees of freedom of the superconducting state. A possible origin of the hysteresis is current-induced chiral-domain-wall motion due to the chiral \( p \)-wave state.

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Most superconductors have a spin-singlet pairing state, including high-\( T_c \) cuprates. Spin-triplet pairing superconductors are quite rare, and the superconducting transition temperature (\( T_c \)) is generally low (~1 K). Layered perovskite \( \text{Sr}_2\text{RuO}_4 \) (SRO) is one of the best candidates for spin-triplet pairing with a \( T_c \) of 1.5 K, and the superconducting vector order parameter is similar to that of the superfluid \( ^3\text{He}-\text{A} \)—the so-called chiral \( p \)-wave state [1]. The pure SRO phase (1.5-K phase) has been well studied, but SRO-Ru eutectics in which Ru lamellae are embedded are not well understood. The SRO-Ru eutectic is called the 3-kelvin (3-K) phase [2–4] because of the remarkable enhancements of \( T_c \) up to 3 K. However, the enhancement mechanism of \( T_c \) and the pairing symmetry of the 3-K phase have not been understood clearly. Since the 3-K phase is the interface superconductivity in the SRO region between SRO and Ru [4], the volume fraction of the superconducting state is very low compared with that of the pure phase. Thus, it is difficult to determine the pairing symmetry by the ordinary method, i.e., Knight shift by NMR. It is extremely important to determine the pairing symmetry of the 3-K phase, because \( T_c \) of 3 K is the highest among spin-triplet superconductors if the 3-K phase is established as a spin-triplet superconductor. Moreover, the 3-K phase enables us to reveal nanoscale physics in inhomogeneous spin-triplet superconductivity.

Thus far, several experiments have been performed to investigate the pairing symmetry of the 3-K phase. In tunnel junction experiments, Mao et al. [5] and Kawamura et al. [6] observed the zero-bias conductance peak due to Andreev resonance reflecting non-\( s \)-wave superconductivity [7]. Hooper et al. [8] reported the \( c \)-axis transport characteristics, which is interpreted in terms of a complex Josephson network with anomalous asymmetric features in their current-voltage characteristics. Although these results imply an internal phase of the superconducting order parameter, they do not necessarily indicate odd-parity superconductivity of the 3-K phase.

In this study, we investigated the \( ab \)-plane differential resistance-current (\( dV/dI-I \)) and voltage-current (\( V-I \)) characteristics of the 3-K phase by controlling the number of Ru inclusions by a microfabrication technique using a focused ion beam (FIB). We extracted individual channels which connect the 3-K phase region—normal-state region of the SRO–3-K phase region as the superconductor—normal-metal–superconductor junction at \( 3 \, \text{K} > T > 1.5 \, \text{K} \) [Fig. 1(b)]. We then determine the pairing symmetry of the 3-K phase from the temperature dependence of the critical current (\( I_c \)). Finally, we show quite unusual hysteresis in \( dV/dI-I \) and \( V-I \) below 2 K, which strongly indicates the internal degrees of freedom, mostly due to the chiral \( p \)-wave state.

Eutectic crystals of SRO-Ru were grown in an infrared image furnace by the floating zone method [9]. The transport was measured using a standard four-probe technique. The sample neck between the voltage-lead contacts was milled by a FIB to reduce the number of Ru inclusions in this region. The details of the sample preparation and measurement system are described elsewhere [10]. For sample A, the neck region was \( 70 \times 70 \times 35 \, \mu\text{m}^3 \) (sample A-1) before the FIB process [Fig. 1(a), top]. Next, it was successively milled narrower and thinner (samples A-2 → A-3). The final dimensions of the neck were \( 20 \times 20 \times (\leq 10) \, \mu\text{m}^3 \) (sample A-3) [Fig. 1(a), bottom]. In Fig. 1(c), \( \sim 1 \, \mu\text{m} \) thick and 1–6 \( \mu\text{m} \) long Ru inclusions are visible as black bars. Only two pieces of Ru inclusions appear on the topmost surface in the neck region of sample A-3. Thus, there should be only a few Ru inclusions in the neck region, including a few hidden below the surface. Figure 1(d) shows the differential resistance at zero-bias current (\( R \))-temperature curves of samples A-1, 2, and 3, respectively. For clarity, \( R \) is normalized at 4.2 K.
as shown in Fig. 1(e). We can see that the component of the 3-K phase is more dominant in sample A-3 than in sample A-1. Thus, the milling process can change the ratio of the 1.5-K:3-K phase. Note that it is not always the case that the component of the 3-K phase becomes dominant by milling.

In Figs. 2(a) and 2(b), we show $dV/dI$ vs $I$ curves normalized to 4.2 K for samples A-1 and A-3. Below ~3 K, $dV/dI$ curves show dip structures ($dV/dI \to 0$) near the zero-bias current, reflecting the 3-K phase superconductivity. Below 1.6 K, the $dV/dI$ curves show the flat zero resistance, reflecting the 1.5-K phase superconductivity in addition to a path formation due to the proximity effect of the 3-K phase superconductivity. Increasing the bias currents beyond some critical values makes the $dV/dI$ values larger. In particular, for sample A-3, more characteristic kinks were observed in the $dV/dI$ curves than for sample A-1. With a filament (nonuniform) model in which local superconducting channels connect Ru islands, the characteristic kinks are naturally explained by the superconducting linkage channels in series with their own critical currents, as illustrated in the inset in Fig. 2(b). Thus we can extract the $I_c$ of each channel for sample A-3. In sample A, it is difficult to separate the channels without FIB milling possibly because similar linkage channels are averaged; i.e., sample A-1 hides the individuality of the channels.

Figure 3 shows the temperature dependence of $I_c$ focusing only on the most pronounced kinks [the arrows in Fig. 2(b)] for three different types of samples cut from

FIG. 2 (color online). Normalized $dV/dI$ vs $I$ as a function of temperature for samples A-1 (a) and A-3 (b), respectively. The open and solid symbols denote the different sweep directions, from zero to max. (open), max. to min. (solid), and min. to zero (open). A series of the most pronounced kinks are denoted by arrows (average currents of the $dV/dI$ peaks in upward and downward sweep directions). Inset: Characteristic kinks in the $dV/dI$ curves are explained by superconducting linkage channels with critical currents ($I_{c1} < I_{c2} < \ldots$) in series. The red (black) line corresponds to a schematic of $dV/dI$ ($V-I$).

FIG. 3 (color online). Critical currents ($I_c$) vs normalized temperature for samples A-3, B, and C, respectively. The three curves are traces of the most pronounced kinks in each series of $dV/dI$ curves [for sample A-3, the arrows in Fig. 2(b)].
the same crystal rod. A continuous monotonic increase is seen in the value of $I_c$ with the temperature decreasing through $T_{c0}$ which is defined as the zero-resistivity point in Figs. 1(d) and 1(e). The temperature dependence of $I_c$ should reflect the pairing symmetry of the 3-K and 1.5-K phases. From the experiment by Jin and co-workers [11], $I_c$ of Josephson junctions (JJs) in Pb-SRO-Pb increases below 7.2 K ($T_c$ of Pb) with decreasing temperature but decreases below 1.3 K ($T_c$ of SRO). This result is explained by the transition from 0 to the $\pi$ junction, which forms below 1.3 K due to the difference in parity of the pairing symmetry (Pb has s-wave and SRO has $p$-wave pairing symmetry) either as a first-order [12] or a second-order process [13]. Assuming a similar relation, we deduce that the 3-K and 1.5-K phases have the same parity, such as s-s-s or p-p-p in the 3-K-1.5-K-3-K phase configuration, excluding $\pi$-junction configurations. Therefore, considering an odd parity in the 1.5-K phase, the 3-K phase also has an odd parity in the local superconducting channel. This is an experimental proof of the assumption in a phenomenological theory by Sigrist and Monien (SM) [14], which states that the filamentary phase at 3 K has $p$-wave pairing symmetry.

In the $dV/dI-I$ curves, we observed unusual hysteresis below 2 K, as seen for sample A-3 [Fig. 2(b)]. The hysteresis curves are more pronounced at temperatures below $T_{c0}$ and tend to appear for small samples after FIB milling. To demonstrate the anomalous behavior in $dV/dI-I$ data clearly, we show V-I curves obtained by the ac method as well as $dV/dI-I$ curves for sample D (the neck region is $3 \times 10 \times 5 \mu m^3$(length $\times$ thickness)) at 1.3 K in Figs. 4(a) and 4(b). The V-I data show discontinuous points at $\pm 3$ and $\pm 4.3$ mA when sweeping up of absolute value of the dc current [Fig. 4(a)]. To explain this behavior, we show a schematic of the V-I data in Fig. 4(c). When we increase the dc current from 0, a finite voltage appears at $I_{c0}$ like usual JJs. Here the $I_{c0}$ is defined as a critical current at which a finite $dV/dI$ value is observed. Surprisingly, the voltage suddenly drops at a threshold $I_{th1} > I_{c0}$ denoted by the arrow. With further increasing of the dc current, a similar voltage drop appears at $I_{th2}$. We note that one can measure only positive $dV/dI$ by the ac method [Fig. 4(b)] because the switching occurs instantaneously at $I_{th1}$, $I_{th2}$, etc. That is, the $dV/dI$ value around the threshold reflects that just before the switching or just after the switching.

Here we emphasize that $I_{th}$ is not a critical current from the dc to ac Josephson effect as seen in the inset in Fig. 2(b) because of the following anomalous features. (i) At $I_{th}$, the voltage discontinuously decreases when the V-I curve switches to the next branch. (ii) It switches to a lower-$R_p$ (normal resistance) branch that has a larger $I_c$. (iii) The hysterisis loop shows the opposite direction compared to typical JJs. In serially connected typical JJs, e.g., the c-axis intrinsic JJs of high-$T_c$ cuprate Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ [15], the voltage absolutely increases and $R_p$ becomes larger after the zero-voltage state changes to the finite-voltage state. Furthermore, in typical JJs, $I_c$ when sweeping up from 0 is always higher than $I_{th}$ when sweeping down [16], which is obviously contrary to the data shown in Fig. 4(d). Therefore, the anomalous switchings at $I_{th}$ above $I_c$ are never explained in terms of serially connected JJs. In other words, the anomalous switching phenomena occur in the identical channel. Thus, it cannot be explained without considering the internal degrees of freedom of the superconducting state.

One of the possible explanations of the unusual hysteresis is due to the chirality of the superconducting state taking account of the 1.5-K phase being a chiral $p$-wave ($p_x \pm ip_y$) superconductor. The chiral state has two types of distinct domains in the superconducting state. If a superconducting linkage is formed from two antiparallel domains, there should exist a domain wall (DW) between them. No DW is formed between parallel domains. Let us assume that a local linkage channel contains both parallel and antiparallel domains, as depicted in Fig. 5. Each critical current is expected to be proportional to each cross section, i.e., $I_c \propto S_{\text{filament}}$ or $I_c \propto S_{\text{DW}}$, where $S_{\text{filament}}$ is the cross section at which a weak link forms between parallel domains and $S_{\text{DW}}$ is that between antiparallel domains. Generally, a DW is likely to be formed and pinned at defects in the sample. Once a DW forms, dc current which transfers a Cooper pair with a given chiral state ($p_x, -ip_y$) to the opposite chiral state ($p_x + ip_y$) induces the DW
motion as a backaction. Assuming the spatial variation of the cross section of the linkage channel, the movement of the DW varies $I_c$ during the current sweep. If the DW is pinned at defects at low-bias currents, the DW slides to the next metastable position when the dc current beyond the threshold $I_{th1}$, $I_{th2}$, etc., in Fig. 4(c) is applied. Thus, the branch of the $V-I$ curve switches, which causes the anomalous hysteresis. As a small $S_{DW}$ should be energetically favorable because the different order parameters overlap each other at the DW, it is reasonable that $I_c$ is lower than $I_{c0}$. On the other hand, no hysteresis would appear in a channel between parallel domains without a DW. In short, the $dV/dI$-$I$ curve shows two types: no hysteresis without a DW and hysteresis with a DW moving in alternate directions. The DW motion is analogous to the current-driven DW motion in magnetic wires [17]. Here we note that the chiral domain picture is consistent with the SM’s prediction of the 2nd transition at $T_2^c$ (3 K $> T_2^c$ $> 1.5$ K) with time reversal symmetry breaking. We also note that no hysteresis has been reported [8], possibly because $I_c$ of averaged channels smeared it under a large number of Ru inclusions.

Recent Kerr effect [18] and JJ [19] experiments suggest the presence of the chiral domains for the 1.5-K phase. However, the estimated domain size is $\sim 50-100$ $\mu$m [18] and $\sim 1$ $\mu$m [19], respectively. Thus, a consensus about the domain size is not established yet. In our experiment for the 3-K phase, we estimate the domain size to be $\sim 10$ $\mu$m from the neck region of the sample if the chiral domain scenario is correct. We emphasize that our four-probe configuration is not sensitive to surface or interface states. Thus, our result reflects the intrinsic properties of the superconducting state in SRO removing experimental ambiguity as much as possible. Further experimental work is needed to verify the presence of the chiral domains in the 3-K phase and make further discussions about the effects of the chiral domains on the Josephson current [20].

In summary, we revealed the superconducting nature of the 3-K phase in Sr$_2$RuO$_4$ by transport measurements on microfabricated samples. We confirmed that the 3-K phase has odd-parity pairing symmetry, similar to the 1.5-K phase, from the monotonic temperature dependence of the critical currents. The unusual hysteresis of the differential resistance or the voltage-current characteristics in the sweeping current observed below 2 K indicates the internal degrees of freedom of the superconducting pairing, i.e., the chiral $p_x \pm ip_y$ state. The domain wall motion induced by the dc current is a possible origin of the hysteresis. This is a new discovery of the dynamic response of the superconducting order parameter of Sr$_2$RuO$_4$.

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