Current-induced superconducting anisotropy of Sr$_2$RuO$_4$

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(Dated: November 12, 2021)

In the unconventional superconductor Sr$_2$RuO$_4$, unusual first-order superconducting transition has been observed in the low-temperature and high-field region, accompanied by a four-fold anisotropy of the in-plane upper critical magnetic field $H_{c2}$. The origin of such unusual $H_{c2}$ behavior should be closely linked to the debated superconducting symmetry of this oxide. Here, toward clarification of the unusual $H_{c2}$ behavior, we performed the resistivity measurements capable of switching in-plane current directions as well as precisely controlling the field directions. Our results reveal that resistive $H_{c2}$ under the in-plane current exhibits an additional two-fold anisotropy. By systematically analyzing $H_{c2}$ data taken under various current directions, we succeeded in separating the two-fold $H_{c2}$ component into the one originating from applied current and the other originating from current imperfection in the sample. The former component, attributable to vortex flow effect, is weakened at low temperatures where $H_{c2}$ is substantially suppressed. The latter component is enhanced in the first order transition region, possibly reflecting a change in the nature of the superconducting state under high magnetic field.

INTRODUCTION

The layered perovskite superconductor Sr$_2$RuO$_4$ with the transition temperature $T_c$ of 1.5 K [1] has been extensively studied due to its unconventional pairing state. Its Fermi surface has a relatively simple topology, consisting of three cylindrical sheets ($\alpha$, $\beta$, and $\gamma$) [2, 3] with its well-characterized Fermi-liquid behavior. Recent nuclear magnetic resonance (NMR) experiments using low rf pulses revealed a technical problem in previous reports and clarified a reduction of the $^{17}$O Knight shift in the superconducting states [4–6]. Recent polarized neutron scattering experiments performed under lower fields also revealed the reduction in the magnetic susceptibility in the Ru site [7]. These results cannot be explained in the framework of the traditional spin-triplet superconductivity scenario. Zero-field muon spin rotation ($\mu$SR) as well as magneto-optic Kerr effect experiments showed evidence for time-reversal-symmetry breaking (TRSB) [8, 9]. Recent $\mu$SR experiments under uniaxial stress reveal stress-induced splitting between the onset temperatures of superconductivity and TRSB [10] whereas such splitting does not occur as long as the tetragonal symmetry is preserved [11]. Furthermore, ultrasound measurements show that the superconducting order parameter of Sr$_2$RuO$_4$ consists of at least two components [10, 12, 13]. More recently, it is revealed that the NMR spectrum near the upper critical magnetic field $H_{c2}$ exhibits characteristic “double-horn” structure, indicating the superconducting spin smecticity [14]. This result suggests that Sr$_2$RuO$_4$ is a strong candidate for the formation of the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state [15, 16].

Toward clarification of the debated superconducting order parameter of Sr$_2$RuO$_4$, properties near $H_{c2}$ is of primary importance. Indeed, the superconducting transition becomes the first-order transition (FOT) under magnetic fields aligned accurately in the $ab$ plane and below 0.8 K [17, 18]. Considering the recent revival of the NMR data, this first-order transition can be well interpreted as a consequence of the Pauli-paramagnetic pair-breaking effect, which is not allowed in the traditional chiral-$p$-wave spin-triplet scenario. More interestingly, in the same temperature region, $H_{c2//ab}$ exhibits a clear four-fold in-plane anisotropy [18–20]. Although this four-fold anisotropy preserves the tetragonal crystalline symmetry, its origin and relation to the first-order transition have not been clarified. The motivation of our study is to reveal the relationship between the $H_{c2//ab}$ anisotropy and FOT.

We focus on the anisotropy of $H_{c2}$ in the $ab$ plane under in-plane currents. First of all, $H_{c2}$ under currents is expected to show the same four-fold symmetry as that under zero current [18–20]. Secondly, since type-II superconductors under currents and magnetic fields above the lower critical field $H_{c1}$ exhibit vortex flow resistivity caused by the Lorentz force [21], $H_{c2}$ in the $ab$ plane of Sr$_2$RuO$_4$ is also expected to exhibit current-induced two-fold symmetry depending on the relative angle $\phi_H - \phi_I$ between the magnetic field $H$ and current $I$. Here, $\phi_H$ and $\phi_I$ are the in-plane angles of the field and current with respect to the crystal axes, respectively. Thirdly, $H_{c2}$ under currents may also depend on $\phi_I$. Furthermore, these anisotropies of $H_{c2}$ under currents are expected to show different behavior between the FOT and second-order transition (SOT) regions due to the difference in the dominant pair-breaking mechanisms. Thus, we consider the direction and strength of the current as new parameters of controlling $H_{c2}$, and extensively investigate the temperature dependence of the anisotropies of $H_{c2}$.

In order to clarify the electric current effect on the anisotropy of $H_{c2}$, it is crucial to identify and eliminate the effects of technical origins. First, because the value of $H_{c2//c}$ is about one twentieth less than that of $H_{c2//ab}$ [20], the presence of a small misalignment of in-plane magnetic field itself can lead to an apparent two-fold anisotropy of $H_{c2}$ in the $ab$ plane. Second, since the distribution of currents depends on the shape and distortion of the sample device, they can cause...
apparent anisotropy dependent on the direction of the current $\phi_I$. To avoid detecting these extrinsic anisotropies, we established a measurement procedure in which magnetic fields are applied in the $ab$ plane accurately and precisely, and currents can be switched in various crystal orientations.

In this work, we measured resistivity of a micro-structured single-crystalline Sr$_2$RuO$_4$ sample under various in-plane field and current directions. The sample was processed by focused ion-beam (FIB) as shown in Fig. 1(a); this structured sample allows us to switch the current in the [100], [010], [110] and [110] directions. We find that, with a given current direction, $H_{c2}$ exhibits two-fold anisotropy as a function of the in-plane field angle, in addition to the ordinary four-fold anisotropy. Careful analysis reveals that the two-fold component consists of two contributions: the one caused by external current likely due to vortex-flow effect, and the other originating from sample inhomogeneity probably introduced by the FIB process. We revealed that the latter is enhanced below 0.8 K, namely in the FOT regime, attributable to a change in the superconducting order parameter.

**EXPERIMENTAL**

In this study, we used Sr$_2$RuO$_4$ single crystals grown with the floating-zone method [23]. Before the micro-fabrication of the resistivity device, $T_c$ of a thin crystal (batch: C432; 500 $\mu$m $\times$ 100 $\mu$m $\times$ 10 $\mu$m) was measured to be 1.43 K (see Supplemental Material (SM) [24]), defined as the peak of the imaginary part of the AC susceptibility measured using a compact susceptometer [25] that fits inside a commercial refrigerator (Quantum Design; PPMS adiabatic demagnetization refrigerator option). The crystalline orientation of the sample was determined by x-ray Laue pictures. The crystal was placed on a single-crystalline SrTiO$_3$ substrate (Shinkosha), which has a thermal contraction match with that of Sr$_2$RuO$_4$ [26, 27]. After the electrical contact between the Sr$_2$RuO$_4$ crystal and eight electrodes, made of high-temperature-cure silver paint (Dupont, 6838), were established similarly to previous studies [28], the surface of the crystal was protected by evaporating a 0.5-$\mu$m layer of SiO$_2$. We then used a focused ion beam (FIB) instrument (JEOL, JIM-4501) with a Ga ion beam to fabricate the current-direction switchable resistivity device. Figure 1(a) shows a scanning electron microscope (SEM) image of the device taken from the $c$-axis direction. As shown in the figure, the widths of the arms along the [100] and [010] directions are both 20 $\mu$m, and those along the [110] and [110] directions are 10 $\mu$m. The device thickness is 10 $\mu$m.

The Sr$_2$RuO$_4$ device was cooled down to 0.12 K with a $^3$He-$^4$He dilution refrigerator (Oxford Instruments, Kelvinox 25). For controlling the field orientation at low temperatures, we used a vector magnet system (Cryomagnetics, VSC-3050) with orthogonally arranged SC magnets that generate a horizontal field $H_z$ of up to 5 T and a vertical field $H_r$ of up to 3 T to control the polar field angle $\theta_{lab} = \arctan(H_z/H_r)$ [29]. This vector magnet allows control of $\theta_{lab}$ with a typical precision of $\Delta\theta_{lab} = 0.005^\circ$ at $\mu_0H = 1$ T. Moreover, the magnet set is placed on a horizontal rotation stage, which can control the azimuthal field angle $\phi_{lab}$ with a precision of $\Delta\phi_{lab} = 0.001^\circ$. The quasi-two-dimensional (2D) anisotropy of $H_{c2}$ of Sr$_2$RuO$_4$ ($H_{c2}//ab$) allows us to align the field to the $ab$ plane accurately and precisely: by rotating magnetic field close to $H_{c2}//ab$, we can find a sharp drop of resistivity when the field is exactly parallel to the $ab$ plane (see SM [24]).

The in-plane alignment was done based on the known four-fold anisotropy of the in-plane $H_{c2}$ (i.e. $H_{c2}//[110] > H_{c2}//[100]$ below 0.8 K) [19, 20]. All the data shown in this paper are

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**FIG. 1.** Current-direction switchable device of single-crystalline Sr$_2$RuO$_4$ for $H_{c2}$-anisotropy measurements. (a) Scanning electron microscope (SEM) image of the device after the focused ion-beam (FIB) process. FIB-cut trenches are visible as black lines with white edges. The orange broken line indicates the original shape of the Sr$_2$RuO$_4$ crystal. To obtain low contact resistance, we used high-temperature-cure silver paint (Dupont, 6838). (b) Schematics of the four configurations of resistivity measurements with different current directions under magnetic fields. The [100] direction is defined as $\phi = 0^\circ$ and the [010] direction as $\phi = 90^\circ$ for both the field angle $\phi_H$ and current angle $\phi_I$. (c) Resistive transition of the device measured on warming under zero field. The superconducting transition temperature $T_c$ (1.45 K) remains close to the value for the cleanest samples (1.5 K) [22] even after the FIB process, and $T_c$ does not depend on current below 200 $\mu$A.

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| $I$ (μA) | 50 | 100 | 150 | 200 |
|----------|----|-----|-----|-----|
| $R$ (mΩ) | 0.1 | 0.05 | 0.0 | 0.0 |

| $H$ (T) | 0.0 | 0.5 | 1.0 | 1.5 | 2.0 |
|---------|-----|-----|-----|-----|-----|
| $\mu_0H = 0$ T | 0.05 | 0.0 | 0.0 | 0.0 | 0.0 |
plotted using the field angles $\phi_H$ and $\theta_H$ defined in the sample coordinate.

To measure the sample resistance $R$, we employed a DC method with current sign reversal to avoid influence of voltage offsets such as thermoelectric voltages. We used a combination of a current source (Keithley, 6221) and a nanovoltmeter (Keithley, 2182A). We measured $R$ against various parameters: temperature $T$, magnitude and azimuthal angle of magnetic field $H$ and $\phi_H$, current amplitude and direction $I$ and $\phi_I$. Both $\phi_H$ and $\phi_I$ are defined with respect to the [100] direction as shown in Fig. 1(b). We note that for a given current value, the current density for an orthogonal measurement ($I//[100]$ or [010]) is half of that for a diagonal measurement ($I//[110]$ or [1T0]) due to the larger width of the arm along the [100] or [010] directions.

Figure 1(c) shows the temperature dependence of $R$ under zero field. The resistivity reaches zero at $T_c = 1.45$ K. The consistent $T_c$ values before and after FIB assure that a possible damage or strain due to FIB is minimal. In this study we used the current values below 200 $\mu$A for which $T_c$ does not change under zero field.

RESULTS AND ANALYSIS

To investigate the anisotropy of the resistive $H_{c2}$ in the $ab$ plane under in-plane currents, we measured $R(H)$ under various field and current conditions. We show in Fig. 2 representative field dependence of the resistance for $I//[100]$. Additional raw data are shown in SM [24]. The in-plane $H_{c2}$, defined with the deviation from $R = 0$ as indicated by the arrows, clearly depends on the in-plane field direction and on the current strength.

Figures 3(a) and 3(c) compare the $\phi_H$ dependence of the in-plane $H_{c2}$ for $I//[010]$ and $I//[110]$ measured at 200 $\mu$A and 0.40 K. Under in-plane currents, $H_{c2}(\phi_H)$ clearly shows not only the known four-fold anisotropy but also an additional two-fold anisotropy. This is also evident in the raw data in Fig. 2: For example, $H_{c2}$ for $\phi_H = 45^\circ$ is noticeably larger than that for $\phi_H = 135^\circ$, although these two conditions should be equivalent considering the tetragonal crystalline symmetry if the current were absent. We have confirmed that this two-fold behavior is not caused by the misalignment of fields with respect to the $ab$ plane (Fig. S2 in SM [24]). Moreover, comparing Figs. 3(a) and (c), we notice that this two-fold components of $H_{c2}$ are altered by changing the current direction $\phi_I$. For example, the $H_{c2}$ value at $\phi_H = 0^\circ$ is smaller than that at $90^\circ$ for $I//[010]$ (Fig. 3(a)), whereas they are opposite for $I//[110]$ (Fig. 3(c)). This fact also evidences that the external current plays an important role in the observed two-fold behavior.

To analyze the two-fold behavior quantitatively, we fit the $H_{c2}(\phi_H)$ data with a combination of two and four-fold sinusoidal functions with a constant offset:

$$\mu_0 H_{c2}(\phi_H) = a_0 + a_2 \cos 2\phi_H + a_4 \cos 4\phi_H + b_2 \sin 2\phi_H + a_4 \cos 4\phi_H. \quad (1)$$

We comment that the four-fold cosine term represents the known $H_{c2}$ anisotropy under zero current ($H_{c2}([110]) > H_{c2}([100])$) [19, 20] and we have checked that the four-fold sine term is negligible even under currents. As exemplified by the solid curves in Figs. 3(a) and (c), the fitting was successful for the $H_{c2}(\phi_H)$ data sets. The obtained value of $a_4$ is $\sim -0.011$ T and $\sim -0.010$ T for the data in Fig. 3(a) and (c). These values are consistent with the previous studies [19, 20]. Notice that the quantity $\Delta(\mu_0 H_{c2}) = \mu_0 H_{c2}([110]) - \mu_0 H_{c2}([100])$ in the literature corresponds to the quantity $-2a_4$ of our study. Then, to extract the two-fold component $H_{c2}^2$, we subtracted the fitted four-fold component $a_4 \cos 4\phi_H$ and the offset $a_0$ from the data. The results are shown in Figs. 3(b) and (d). Here, the colored curves represent the two-fold terms obtained by the fitting, whereas the black solid and broken curves are decomposed $\cos 2\phi_H$ and $\sin 2\phi_H$ components, respectively. Between $I//[100]$ and [010], $H_{c2}^2$ is clearly different with the
FIG. 3. $H_{c2}$ anisotropy of Sr$_2$RuO$_4$ under in-plane current. Additional data are shown in SM [24]. (a) Raw $H_{c2}$ data as a function of $\phi_H$ under current along the [010] direction. The data are well fitted with a combination of four-fold and two-fold sinusoidal function (Eq. (1)) as shown with the pink solid curve. (b) two-fold component of $H_{c2}$ under $I \parallel$[010] (pink squares), extracted from the raw $H_{c2}$ data by subtracting the fitted four-fold component and the constant offset. The pink curve present the two-fold sinusoidal fitting, whereas the solid and broken black curves show the decomposed cosine and sine terms of the pink curve, respectively. (c) Raw $H_{c2}$ data (circles) and fitting result (solid curve) under current along the [100] direction. Compared to (a), the data suggest that current direction $\phi_H$ switches the sign of the difference between $H_{c2}$ at $\phi_H = 0^\circ$ and that at $\phi_H = 90^\circ$. (d) two-fold component of $H_{c2}$ under $I \parallel$[100] together with the two-fold fitting (orange curve) and the decomposed cosine (black solid curve) and sine (black broken curve) components. (e) $I$-induced two-fold component of $H_{c2}$ obtained by taking the difference between the data in (b) and (d). This only contains the two-fold component that is switched by the change of the current direction. (f) $I$-independent two-fold component of $H_{c2}$ obtained by taking the summation of the data in (b) and (d). This only contains the two-fold component that is not switched by the change of the current direction.

We present the $T$-dependence of the obtained fitting parameters $a_2$ and $b_2$ in eq. 1 for various current directions in Fig. 4. (Behavior of $a_0$ and $a_4$ will be discussed later.) For the coefficient of the $\cos 2\phi_H$ component $a_2$, its magnitude under the orthogonal current directions, namely $I \parallel$[100] and $I \parallel$[010], increases on warming while its sign is switched by current-direction change. This is indeed expected for a current-induced effect, where by symmetry $H_{c2}^{(2)}$ should behave as $\propto \cos 2(\phi_H - \phi_f)$. However, unlike $a_2$, $b_2$ under all current-directions have an additional term reaching nearly 0.005 T on cooling to 0 K.
This is again consistent with the \( \cos 2(\phi_H - \phi_I) \) behavior for the current-induced \( H_c^{(2)} \), since \( \cos 2(\phi_H - \phi_I) = \pm \sin 2\phi_H \) for \( \phi_I = \pm 45^\circ \) and thus the cosine component should be zero. Figure 4(b) shows the temperature dependence of the coefficients \( b_2 \). At high temperatures, the behavior of \( b_2 \) is similar to that of \( a_2 \) except for an exchange of the roles between the orthogonal and diagonal current directions: \( b_2 \) is finite and its sign depends on the current direction //\{110\} or \{1\overline{1}0\}, but is nearly zero for \( I//\{100\} \) and \{010\}. Thus this high-temperature behavior in \( b_2 \) again indicates \( H_c^{(2)} \propto \cos 2(\phi_H - \phi_I) \) behavior. However, at lower temperatures, \( b_2 \) tends to converge to a finite value \( b_2(T \to 0) \sim 0.005 \) T irrespective of the current directions. Such convergence reveals an additional two-fold component that is not caused by external current.

From these analyses, we revealed that the \( H_c^{(2)} \) data have current-independent and current-dependent contributions, which we hereafter express as \( H_c^{(2\text{ind})} \) and \( H_c^{(2\text{dep})} \). The former corresponds to the low-temperature current-direction independent behavior in \( b_2 \) (Fig. 4(b)) and the latter corresponds to the \( \cos 2(\phi_H - \phi_I) \) term as seen in the behavior common to \( a_2 \) and \( b_2 \) discussed in the previous paragraph. Further analyses and interpretations of these contributions will be provided in the next section.

Before closing this section, we comment on possible heating effects due to current. We estimate the upper limit of the actual sample temperature from the current dependence of the coefficient \( a_0 \) by assuming that its variation with current is solely due to Joule heating (see SM [24]). This estimation indicates that the temperature increase is at most 0.2 K in the lowest temperature region and less than 0.1 K above 0.4 K.

**DISCUSSION**

As explained in the previous section, we extracted two-fold anisotropy of \( H_c \) of Sr$_2$RuO$_4$ under current. We reveal systematically how this additional anisotropy depends on current-strength, current-directions, and temperature. As a well-known effect, external current induces vortex flows in the mixed state by the Lorentz force, leading to the appearance of \( H_c \) field parallel to current; As a result, this effect is expressed as a two-fold cosine-like component \( \propto \cos 2(\phi_H - \phi_I) \), whose sign is negative under field perpendicular to current (i.e. \( \phi_H - \phi_I = \pm 90^\circ \)) and positive under field parallel to current (i.e. \( \phi_H = \phi_I \)).

For ordinary type-II superconductors (both 2D and 3D) in which the main pair-breaking effect is the ordinary orbital effect, the vortex-flow effect is expected to be observed. However, in Sr$_2$RuO$_4$, the FOT occurs below 0.8 K, indicating that the dominated pair-breaking effect is not the orbital effect. Thus, the two-fold cosine-like component can behave differently between the FOT and the SOT regions.

In order to extract the current-dependent term including the vortex-flow effect from the \( H_c^{(2)} \) data, we take the difference between the \( H_c^{(2)}(\phi_H, \phi_I = 90^\circ) \) data and \( H_c^{(2)}(\phi_H, \phi_I = 0^\circ) \) data. This process is motivated by the following expectation. We expect that the two-fold component \( H_c^{(2)}(\phi_H, \phi_I) \) contain current-independent term \( H_c^{(2\text{ind})}(\phi_H) \) and current-dependent term \( H_c^{(2\text{dep})}(\phi_H - \phi_I), \) i.e.

\[
H_c^{(2)}(\phi_H, \phi_I) = H_c^{(2\text{ind})}(\phi_H) + H_c^{(2\text{dep})}(\phi_H - \phi_I), \tag{2}
\]

Then, due to the two-fold nature, \( H_c^{(2\text{dep})} \) should change sign upon a current-direction switching of 90°, as

\[
H_c^{(2\text{dep})}(\phi_H - (\phi_I + 90^\circ)) = -H_c^{(2\text{dep})}(\phi_H - \phi_I). \tag{3}
\]

In contrast, \( H_c^{(2\text{ind})} \) is, by definition, independent of the 90° current direction switching. Thus, by calculating

\[
A_{\text{orth}} H_c^{(2)} \equiv H_c^{(2)}(\phi_H = 90^\circ) - H_c^{(2)}(\phi_H = 0^\circ), \tag{4}
\]

\( H_c^{(2\text{ind})} \) is eliminated and we can expect that \( A_{\text{orth}} H_c^{(2)} \approx 2H_c^{(2\text{dep})}(\phi_H = 90^\circ) \). A representative \( A_{\text{orth}} H_c^{(2)} \) is shown in Fig. 3(e). Similarly,

\[
A_{\text{diag}} H_c^{(2)} \equiv H_c^{(2)}(\phi_H = 45^\circ) - H_c^{(2)}(\phi_H = -45^\circ) \tag{5}
\]

should satisfy \( A_{\text{diag}} H_c^{(2)} \approx 2H_c^{(2\text{dep})} \) (Fig. S4), providing an independent evaluation of \( H_c^{(2\text{dep})}(\phi_H = 45^\circ) \). On the other hand, we can extract the current-independent term \( H_c^{(2\text{ind})}(\phi_H) \) by evaluating sums of the datasets with 90° current switching:

\[
\Sigma_{\text{orth}} H_c^{(2)} \equiv H_c^{(2)}(\phi_H = 90^\circ) + H_c^{(2)}(\phi_H = 0^\circ) \tag{6}
\]

and

\[
\Sigma_{\text{diag}} H_c^{(2)} \equiv H_c^{(2)}(\phi_H = 45^\circ) + H_c^{(2)}(\phi_H = -45^\circ). \tag{7}
\]

In these sums, current-dependent terms are eliminated and we expect \( \Sigma_{\text{orth}} H_c^{(2)} \approx \Sigma_{\text{diag}} H_c^{(2)} \approx 2H_c^{(2\text{ind})} \). A representative \( \Sigma_{\text{orth}} H_c^{(2)} \) is shown in Fig. 3(f) and \( \Sigma_{\text{diag}} H_c^{(2)} \) in Fig. S4.

In Figs. 3(e), 3(f), S4, one can clearly see that \( A_{\text{orth}} H_c^{(2)} \) and \( A_{\text{diag}} H_c^{(2)} \) are dominated by \( \cos 2(\phi_H - \phi_I) \) \( \phi_H = 90^\circ \) and \( \phi_I = 90^\circ \) and \( \phi_I = -90^\circ \). To perform quantitative analysis, we fitted (1/2)\( \Sigma H_c^{(2)} \) by \( A_2 \cos 2\phi_H + B_2 \sin 2\phi_H \) and (1/2)\( \Sigma H_c^{(2)} \) by \( C_2 \cos (\phi_H - \phi_I) \) \( \phi_H = 90^\circ \), \( \phi_I = -90^\circ \). The resultant parameters are plotted in Figs. 5(c), (d), S6(a) and S6(b). We confirmed that the coefficients evaluated from the orthogonal and diagonal conditions agree with each other, manifesting the validity of the analyses. We then indeed find \( A_2 \) and \( B_2 \) are small and do not exhibit temperature dependence. In contrast, \( B_2 \) and \( C_2 \) plotted in Figs. 5(c) and (d) are much larger than \( A_2 \) or \( B_2 \) and exhibits characteristic temperature dependence as will be discussed later.

In Figs. 5(a) and (b) the angle-independent component \( a_0 \) (see Eq. (1)) and four-fold component \(-2a_4 \) are compared with the coefficients \( B_2 \) and \( C_2 \). Notice that the coefficient \( a_0 \) in Eq. (1) can be considered as the in-plane average \( H_c(\phi) \).
and thus provide the $H_{c2}-T$ phase diagram of Sr$_2$RuO$_4$. Indeed, Fig. 5(a) agrees well with the phase diagram reported in literature [18, 20]. The value $-2a_4$ should be equal to $\mu_0H_{c2}^{(2)} - \mu_0H_{c2}^{(100)}$ in the absence of current, characterizing the known four-fold $H_{c2}$ anisotropy. This quantity is nearly zero above 0.8 K but increases on cooling in the FOT region and reaches 0.03 T at the lowest temperature. Such behavior is consistent with the previous studies.

As shown in Fig. 5(d), the coefficient $C_2$ exhibits a finite value and decreases on cooling. The coefficient $D_2$ exhibits a smaller value than $C_2$ and shows only a weak temperature dependence (see SM [24]). These results indicate that the current-dependent components are cosine-like and increase on warming. It is consistent with the scenario where vortex flow gives current-dependent two-fold anisotropy of $H_{c2}$.

The coefficient $B_2$ shown in Fig. 5(c), characterizing current-independent two-fold behavior, is finite only below 0.8 K in the FOT region, for the orthogonal current. A similar tendency is clear for the diagonal current. Since other bulk measurements have not revealed such a current-independent two-fold component of $H_{c2}$, the behavior of $B_2$ is probably related to minute surface damage caused by the FIB processing. However, it is not sufficient to explain the $T$-dependence of $B_2$ because it does not change smoothly but appear only in the FOT region. This behavior can be explained if the resistivity suddenly becomes more sensitive to the inhomogeneity in the FOT region. One possible candidate for such sudden change in the resistivity property is the formation of the FFLO state, which is accompanied by a real-space modulation of the superconducting order parameter and is recently revealed by an NMR experiment [30]. If there is a surface damage, for example, the FFLO order-parameter modulation can be pinned to the surface damage and such pinning can induce additional two-fold behavior in $H_{c2}(\phi_H)$.

**SUMMARY AND CONCLUSION**

Here we summarize the main results of our study.

1. The angle-independent component $a_0$ in Eq. (1) can be considered as the in-plane average $H_{c2}$.

2. The quantity $-2a_4$ characterizes the known four-fold $H_{c2}$ anisotropy. It corresponds to $\Delta(\mu_0H_{c2})$ in the previous studies [19, 20] and exhibits about 80% of the value of $\Delta(\mu_0H_{c2})$ in the first-order transition region. In the second-order transition region, $-2a_4$ shows close to zero in the same way as $\Delta(\mu_0H_{c2})$. Thus, the four-fold anisotropy in $H_{c2}$ under in-plane current shows the same $T$-dependence as that of the four-fold anisotropy in $H_{c2}$ under zero current.

3. The coefficient $C_2$ shows the $\cos(2\phi_H - \phi_\ell)$ component of $H_{c2}^{(2\text{dep})}$. Since it takes positively finite values and increases with temperature, the current-induced two-fold anisotropy in $H_{c2}^{(2\text{dep})}$ is probably due to the vortex-flow effect.

4. The coefficient $B_2$ shows the two-fold $I$-independent sine component of $H_{c2}^{(2)}$. Such a two-fold component has not been observed in previous experiments [18–20]. This quan-

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**FIG. 5.** Relation among the first-order transition (FOT) region and $H_{c2}$ anisotropies under current. (a) $T$-dependence of $a_0$. Since $a_0$ corresponds to the in-plane average value of $H_{c2}$, this curve provide the $H_{c2}-T$ phase diagram of this sample. (b) $T$-dependence of the four-fold component $-2a_4$. The behavior of $-2a_4$ is consistent with the previous studies [19, 20]. In (a) and (b), the pink, orange, sky-blue and green points are for the data obtained with $I//[010]$, [100], [110] and [1|T0], respectively. (c) and (d): $T$-dependence of two-fold $I$-independent component and $I$-dependent component of $H_{c2}^{(2\text{dep})}$. Red squares and blue diamonds are the data obtained for orthogonal and diagonal current-directions, respectively. The two-fold $I$-independent sine component $B_2$ reaches about 0.005 T in the FOT region ($T \leq$ 0.8 K) and nearly zero in the second-order transition region ($T \geq$ 0.8 K). This result indicates that the sample in a high-field state becomes more sensitive to inhomogeneity effect, producing two-fold anisotropy. The $\cos(2\phi_H - \phi_\ell)$ component $C_2$ in (d) increase on warming. This observation is consistent with vortex flow effect.
tity revealed in our study becomes finite only in the first-order transition region. This is probably related to minute the crystal distortion and surface damage caused by FIB processing. However, it is not enough to explain why $B_2$ becomes finite only in the first-order transition region. As a possible scenario, we propose that the increased sensitivity to inhomogeneity is due to the realization of FFLO state in the first-order transition region.

In conclusion, we have established a clear approach to probe the electrical resistivity of two-dimensional superconductors, such as Sr$_2$RuO$_4$, with a varying current and magnetic field directions. The combination of the vector magnet and our FIB structured sample enables us to control the azimuthal field angle $\phi_H$, and switch in-plane current-direction $\phi_I$ along four crystal orientations. We have demonstrated that $H_{c2}$ in the $ab$ plane of Sr$_2$RuO$_4$ exhibits not only an ordinary four-fold anisotropy but also an additional two-fold anisotropy. This two-fold anisotropy increases on warming and probably expressed as $\cos(2\phi_H - \phi_I)$, suggesting that flux flow resistivity generated by the Lorentz effect leads to underestimation of $H_{c2}$ in the $ab$ plane. We find another two-fold term behaving as $\sin 2\phi_H$, independent of the current direction. This component, indicative of the sensitivity of the superconductivity to sample inhomogeneity, becomes noticeable only below 0.8 K, where field-induced phases such as the FFLO state are highly anticipated.

Our study is expected to provide hints toward clarification of the remaining issues on Sr$_2$RuO$_4$, such as the origin of the enhancement of $T_c$ under uniaxial strain or in eutectic crystals, and the relationship between the superconducting order parameters and the anisotropy of $H_{c2}$. In addition, our resistivity measurement system will be helpful for investigating interesting phenomena in other materials such as coupling between current and electronic nematicity in nematic superconductors [31] or in nematic electron liquid systems [32].

ACKNOWLEDGEMENT

The authors thank Y. Yanase, K. Machida, K. Ishida, S. Kitagawa, G. Mattoni, for valuable discussions. We also acknowledge technical supports from Kyoto Univ. LTM Center. This work is supported by Japan Society for the Promotion of Science (JSPS) Core-to-core program (No. JPJSCCA20170002) and by JSPS KAKENHI Nos. JP15H05852, JP15K21717, JP17H06136, and JP20H05158. This work is also supported by Research Grants Council of Hong Kong (CUHK 14301316) and CUHK Direct Grants (No. 4053299, 4053345).

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Supplemental Material for
Current-induced superconducting anisotropy of Sr$_2$RuO$_4$

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(Dated: November 12, 2021)

FIG. S1. AC susceptibility $\chi_{ac} \equiv \chi' - i\chi''$ of the Sr$_2$RuO$_4$ single crystal used for our device. Before the micro-fabrication, ac susceptibility measurement at zero DC field revealed that this thin crystal (batch: C432; 500 µm x 100 µm x 10 µm) exhibits superconducting transition at $T_c = 1.43$ K, which is defined as the peak of the imaginary part of $\chi_{ac}$. 
FIG. S2. Check of the alignment of magnetic field to the ab plane. The figure shows the polar-angle $\theta_H$ dependence of the resistance $R$ measured at 0.50 K under 1.29 T and 100-µA current along the [010] direction. The curves have vertical offsets. The red, orange and pink curves correspond to $\phi_H = 135^\circ, 45^\circ$ and $-45^\circ$, respectively. For all $\phi_H$, $R(\theta_H)$ curves show sharp and symmetric dips centered at $\theta_H = 90^\circ$ due to superconductivity. These data indicate that the misalignment of the magnetic field with respect to the ab plane ($\theta_H = 90^\circ$) is at most 0.1°.
FIG. S3. Raw magneto-resistance data. The figure represents resistive transition of our Sr$_2$RuO$_4$ device under currents. The yellow, brown, pink and black data points are obtained under $\phi_H = 0^\circ$, $45^\circ$, $90^\circ$ and $135^\circ$, respectively. (a) Data under currents along the [010] direction at 0.4 K. Similarly to the $R(H)$ curves in Fig. 2 of the main text, $H_{c2}$, indicated by the vertical arrows, exhibits two-fold anisotropy as evidenced by the differences between the curves of $\phi_H = 0^\circ$ and $90^\circ$, or those of $\phi_H = 45^\circ$ and $135^\circ$. (b) Data under currents along the [010] direction at 1.20 K, where superconducting transition is an ordinary second-order transition. (c) Data under currents along the [110] direction at 1.20 K.
FIG. S4. Representative $H_{c2}$ anisotropy of Sr$_2$RuO$_4$ under in-plane current. Similarly to Fig. 3 of the main text, (a) and (c) represent raw $H_{c2}$ data (circles) and fitting result (solid curve) under current, (b) and (d) represent two-fold components of $H_{c2}$ under current, extracted from the raw $H_{c2}$ data by subtracting the fitted four-fold component and the constant offset, (e) represents $I$-induced two-fold component of $H_{c2}$ obtained by taking the difference between the data in (b) and (d), and (f) represents $I$-independent two-fold component of $H_{c2}$ obtained by taking the summation of the data in (b) and (d), respectively. The blue, green, pink and yellow color indicate data under current parallel to the [110], [110], [010] and [100] directions, respectively. The blue and red color indicate data evaluated from the datasets for the orthogonal and diagonal current-directions, respectively. Column (1) $H_{c2}$ under currents along the diagonal directions at 0.30 K. Column (2) $H_{c2}$ under currents along the orthogonal directions at 1.20 K. Column (3) $H_{c2}$ under currents along the diagonal directions at 1.20 K.
FIG. S5. Examination of heating effect from the \( T \)-dependence of the coefficient \( a_0 \) considered as the in-plane average \( H_{c2} \). (a) Gray, sky-blue, yellow and red squares show \( a_0 \) measured with current strength \( I = 50, 100, 150 \) and \( 200 \mu A \) respectively. Gray curves shows the fitting curve obtained from the in-plane average \( H_{c2} \) measured with \( 50 \mu A \) (b) Red squares and gray curves show the same data as those in the panel (a). Green squares show the points with \( a_0 \) measured with \( I = 200 \mu A \) shifted onto the fitting curve. To estimate the difference between the actual sample temperature and the measured temperature, we assume that the change in \( a_0 \) is caused only by Joule heating and the gray curves exhibits the ideal \( H_{c2}-T \) phase diagram of the sample. This estimation indicates the temperature increase as evaluated by the shift indicated by the gray arrows is at most \( 0.2 \) K in the lowest temperature region and less than \( 0.1 \) K above \( 0.4 \) K. Therefore, the current-heating effect has no qualitative effects on the analyses and conclusions.
FIG. S6. $T$-dependence of the two-fold $I$-dependent component and $I$-independent component of $H^{(2)}_{\omega}$. Red squares and blue diamonds indicate values evaluated from the datasets for the orthogonal and diagonal current-directions, respectively. $B_2$ and $C_2$ shown in the panels (b) and (c) respectively are the same as $B_2$ and $C_2$ in Fig. 5 of the main text. (a) Two-fold $I$-independent cosine component. This coefficient exhibits nearly zero at all temperatures. (b) Two-fold $I$-independent sine component. This coefficient shows about 0.005 T in the first-order transition region ($T \leq 0.8$ K) and nearly zero in the second-order transition region ($T \geq 0.8$ K). This fact implies that the sample in a high-field state gets sensitive to inhomogeneity effect producing two-fold anisotropy. (c) Coefficient for the cos $2(\phi_H - \phi_I)$ component, which increases on warming. It is consistent with vortex flow effect. (d) Coefficient for the sin $2(\phi_H - \phi_I)$ component, exhibiting nearly zero in the whole temperature range.