Angular dependence of the upper critical field of Sr_2RuO_4

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One of the remaining issues concerning the spin-triplet superconductivity of Sr_2RuO_4 is the strong limit of the in-plane upper critical field H_{c2} at low temperatures. In this study, we clarified the dependence of H_{c2} on the angle θ between the magnetic field and the *ab* plane at various temperatures, by precisely and accurately controlling the magnetic field direction. We revealed that, although the temperature dependence of H_{c2} for $|\theta| \geq 5°$ is well explained by the orbital pair-breaking effect, $H_{c2}(T)$ for $|\theta| < 5°$ is clearly limited at low temperatures. We also revealed that the H_{c2} limit for $|\theta| < 5°$ is present not only at low temperatures but also at temperatures close to T_c. These features may provide additional hints for clarifying the origin of the H_{c2} limit. Interestingly, if the anisotropic ratio in Sr_2RuO_4 is assumed to depend on temperature, the observed angular dependence of H_{c2} is reproduced better at lower temperature with an effective-mass model for an anisotropic three-dimensional superconductor. We discuss the observed behavior of H_{c2} based on existing theories.

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I. INTRODUCTION

The layered perovskite superconductor Sr_2RuO_4 with the transition temperature T_c of 1.5 K has been extensively studied due to its unconventional pairing state.[1,2] Knight-shift measurements with nuclear magnetic resonance (NMR) (Refs. 3 and 4) and with spin-polarized neutron-scattering[5] have revealed the invariant spin susceptibility across T_c for $H \parallel ab$, which firmly indicates that the spin part of the Cooper-pair state is triplet. The orbital part is favorably interpreted as odd parity based on the measurements of the critical current I_c through $Pb/Sr_2RuO_4/Pb$ proximity junctions[6,7] and other experiments.[8,9] These results establish that Sr_2RuO_4 is an odd-parity spin-triplet superconductor. In addition, the μSR (Ref. 10) and Kerr effect[11] measurements indicate broken time-reversal symmetry in the superconducting state. The zero-field ground state consistent with all these results is expressed by the vector order parameter, the *d* vector, $\boldsymbol{d} = \Delta_0 \hat{z}(k_x \pm i k_y)$. However, recent Ru-NMR measurements under very low fields down to 20 mT revealed the invariant Knight shift for $H \parallel c$.[12,13] This means $\boldsymbol{d} \parallel ab(\perp H)$ with the following two possibilities.[12–14] The *d* vector can rotate freely in the *ab* plane,[15] or the *d* vector pointing along the *c* axis in zero field can flip perpendicular to the *c* axis by a small magnetic field along the *c* axis.[16] In either case, the spin of the Cooper pair can be polarized to any field directions at least above 20 mT.

Another unsolved issue in Sr_2RuO_4 is the origin of the strong limit of the upper critical field H_{c2}, which occurs when a magnetic field is applied parallel to the *ab* plane.[17] Similar H_{c2} limit is observed in another spin-triplet superconductor UPt_3 for $H \parallel c$,[18] as shown in the inset of Fig. 2(b). These H_{c2} limits are reminiscent of the Pauli effect, which results from the Zeeman energy of quasiparticles. However, in spin-triplet superconductors, the Pauli effect contributes to pair breaking only when $\boldsymbol{d} \parallel H$ because the spin of the triplet Cooper pairs can be polarized along the field direction when $\boldsymbol{d} \perp H$. As mentioned above, the *d* vector of Sr_2RuO_4 is likely to be perpendicular to the magnetic field possibly except at low fields. This suggests that the Pauli effect should not affect H_{c2}. We note that the *d* vector of UPt_3 for $H \parallel c$ was revealed to be perpendicular to the magnetic field ($\boldsymbol{d} \parallel a$) in phase C.[19] Therefore, the H_{c2} limit observed in UPt_3 cannot be attributed to the Pauli effect either. The origins of these H_{c2} limits have not been clarified yet.

In this paper, we report the dependence of H_{c2} of Sr_2RuO_4, determined from the ac susceptibility, on the magnetic field direction between the *ab* plane and *c* axis. Because of the large anisotropy of H_{c2} in Sr_2RuO_4, a small misalignment would lead to a large difference in the value of H_{c2}, especially when the field direction is nearly parallel to the *ab* plane. Therefore, in this study, we controlled the applied field direction more accurately and precisely than in the previous reports.[20,21] We evaluated the $H_{c2}(T)$ curves for different field directions and revealed that the H_{c2} limit is clearly observed only when the angle θ between the magnetic field and the *ab* plane is less than 5°. This H_{c2} limit was revealed to occur not only at low temperatures, but also at temperatures close to T_c. We also identified the angle θ dependence of H_{c2} at several fixed temperatures. We found that $H_{c2}(\theta)$ is fitted better at lower temperature with an effective-mass model for an anisotropic three-dimensional superconductor. In addition, we investigated the difference between $H_{c2}(\theta)$ for fields in the (100) plane and for fields in the (110) plane. The difference appears only at low temperatures below roughly 1 K for small θ. These results allow us to reexamine the origin of the H_{c2} limit based on existing theories.

II. EXPERIMENTAL

We used single crystals of Sr_2RuO_4 grown by a floating zone method.[22] In this paper, we focus on the result obtained from a single crystal with dimensions of approximately 1.0×0.5 mm^2 in the *ab* plane and 0.08 mm along the *c* axis. The directions of the tetragonal crystallographic axes of the sample were determined from x-ray Laue pictures. We shaped the sample so that the side surface of the sample was

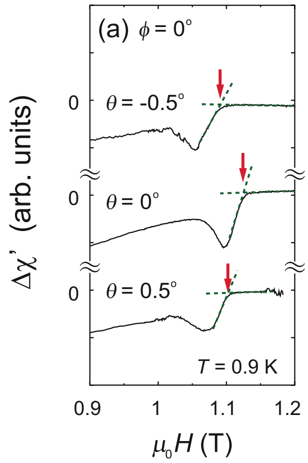

FIG. 1. (Color online) Field dependence of $\Delta\chi' = \chi'(0.9\ \mathrm{K}) - \chi'(2\ \mathrm{K})$ at several θ for (a) $\phi=0°$ and (b) $\phi=45°$. Here, ϕ denotes the azimuthal angle within the *ab* plane between the magnetic field and the [100] axis and θ denotes the angle between the magnetic field and the *ab* plane. H_{c2} is defined as the intersection of the linear extrapolations in $\Delta\chi'$. Thick and thin arrows represent H_{c2} and the anomaly due to the mosaic structure, respectively. The dip in $\Delta\chi'$ near H_{c2} is attributable to the ordinary peak effect (Ref. 21).

10° away from the (100) plane in order to avoid possible anisotropy effects due to surface superconductivity.[23] The crystal was annealed in oxygen at 1 atm and 1050 °C for a week to reduce the amount of oxygen deficiencies and lattice defects. A sharp superconducting transition was observed in the ac susceptibility measurements with the midpoint at $T_c = 1.503$ K.

We measured the ac magnetic susceptibility $\chi_{ac} = \chi' - i\chi''$ by a mutual-inductance technique using a lock-in amplifier with a frequency of 887 Hz. The sample was cooled down to 70 mK with a ^{3}He-^{4}He dilution refrigerator. The ac magnetic field of 20 μT-rms was applied nearly parallel to the *c* axis with a small coil. The dc magnetic field was applied using the "vector magnet" system,[24] with which we can control the field direction three dimensionally and precisely. The accuracy and precision of the field alignment with respect to the *ab* plane are better than 0.1 and 0.01°, respectively. Owing to the high sensitivity of the pick-up-coil parasitic background contributes to the χ_{ac} signal.[25] In order to obtain χ_{ac} contribution only from a superconductivity $\Delta\chi'$, we adopt $\Delta\chi'(T,H) = \chi'(T,H) - \chi'(2\ \mathrm{K},H)$. The small deviation of the normal-state values from zero indicates a good reliability of the background subtraction. We define H_{c2} as the intersection between the linear extrapolations of $\Delta\chi'(H)$ in the superconducting and normal states, as illustrated in Fig. 1 with dashed lines. The directions of the crystalline axes [100] and [001] with respect to the field direction were calibrated by making use of the anisotropy in H_{c2}.[20,21] Our highly accurate and precise measurements revealed that the present sample has a mosaic structure dominated by two domains sharing the [100] axis; the [001] axis of one domain is tilted nearly toward the [010] axis by 0.5° from the [001] axis of the other part.

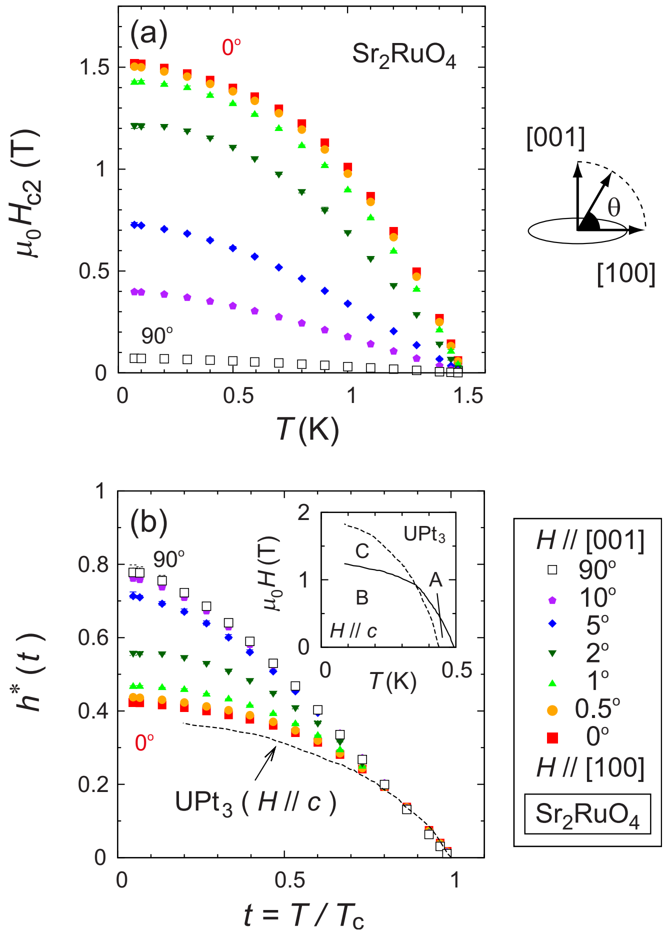

FIG. 2. (Color online) (a) Field-temperature (*H-T*) phase diagram of Sr_2RuO_4 at θ=0, 0.5, 1, 2, 5, 10, and 90° from top to bottom for $\phi=0°$. (b) Temperature dependence of h^* defined as Eq. (1). The inset represents the *H-T* phase diagram of UPt_3 for $H \parallel c$ (Ref. 18). The dashed curve in (b) is $h^*(t)$ of the boundary of the (B+C) phase in UPt_3 for $H \parallel c$ (the dashed curve in the inset).

III. RESULTS

Figure 2(a) is the field-temperature (*H-T*) phase diagram in various field directions at $\phi=0°$, where ϕ denotes the azimuthal angle within the *ab* plane between the magnetic field and the [100] axis. At $\phi=0°$, no anomaly due to the mosaic structure was seen in the raw χ_{ac} data, as shown in Fig. 1(a). Reflecting the large anisotropy of H_{c2} in Sr_2RuO_4, H_{c2} becomes rapidly small when the angle θ between the magnetic field and the *ab* plane increases from 0°. In the specific-heat measurements, the second superconducting transition was observed just below H_{c2} at low temperatures below 0.8 K.[17] Although such an additional transition was observed below 0.6 K in the ac susceptibility measurements, it was difficult to unambiguously identify it to be attributable to the second superconducting transition.[21] This is also the case for the present study. One possible reason for this difficulty is that ac susceptibility, mainly probing the vortex movements, may not be sensitive to the small change in the entropy detected by specific-heat measurements. Therefore, we do not focus on the feature of the additional transition in this paper.

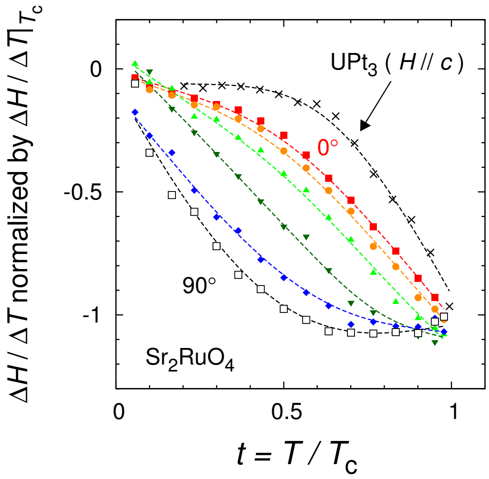

FIG. 3. (Color online) Temperature dependence of the slope of the H-T phase diagram of Sr_2RuO_4 at θ=90, 5, 2, 1, 0.5, and 0° from bottom to top for ϕ=0°. The slope of the H_{c2} curve for UPt_3 (Ref. 18), evaluated from the dashed curve in the inset of Fig. 2(b), is also plotted with crosses. The dashed curves are guides to the eyes.

To characterize the limit of $H_{c2}(T)$, we normalized H_{c2} by the initial slope at T_c,

$$h^*(t) = -\frac{H_{c2}(t)}{dH_{c2}/dt|_{t=1}} \quad (t \equiv T/T_c). \tag{1}$$

If H_{c2} is determined by the orbital pair-breaking effect, which originates from the kinetic energy of supercurrent around magnetic vortices, H_{c2} is described by the Werthamer-Helfand-Hohenberg (WHH) theory[26,27] and its extension to p-wave superconductors.[28,29] In these theories, it is expected that $h^*(t)$ increases linearly on cooling and is weakly suppressed at low temperatures with $h^*(t=0) \sim 0.7$. In Fig. 2(b), we plot $h^*(t)$ with different θ at ϕ=0°. The initial slope $dH_{c2}/dt|_{t=1}$ is defined from the linear fit to $H_{c2}(t)$ in the region $0.85 \leq t \leq 1$. For $|\theta| \geq 5°$, $h^*(t)$ behaves as expected from the WHH theory. In contrast, for $|\theta| \leq 2°$, $h^*(t)$ is strongly limited at low temperatures. This result indicates that the H_{c2} limit in Sr_2RuO_4 is prominent for $|\theta| < 5°$. To emphasize the H_{c2} limit in another spin-triplet superconductor UPt_3, we plot, in Fig. 2(b) with the dashed curve, $h^*(t)$ of the boundary of the (B+C) phase for $H \| c$. Although we chose the less limited one between the two $H_{c2}(T)$ curves in UPt_3 for $H \| c$, a strong limit of $h^*(t)$ is clearly seen.

If the orbital pair-breaking effect is mainly responsible for determining H_{c2}, the slope of the H-T phase diagram should be constant down to well below T_c. To identify the limit of H_{c2} near T_c, we evaluate the slope at temperature $(T_1+T_2)/2$ as $\Delta H_{c2}/\Delta T = [H_{c2}(T_1) - H_{c2}(T_2)]/(T_1 - T_2)$, where T_1 and T_2 are temperatures of adjacent data points. The results are shown in Fig. 3. For $|\theta| \geq 5°$, the slope is constant down to approximately 1 K and approaches zero at low temperatures, which is well explained by the orbital pair-breaking effect. However, for $|\theta| < 5°$, the slope near T_c is not temperature independent any more. This result suggests that the H_{c2} limit observed for $|\theta| < 5°$ is present not only at low temperatures, but also at temperatures close to T_c. The slope of the $H_{c2}(T)$ curve of UPt_3 for $H \| c$ [the dashed curve in Fig. 2(b)] also continues to vary up to T_c, as plotted in Fig. 3.

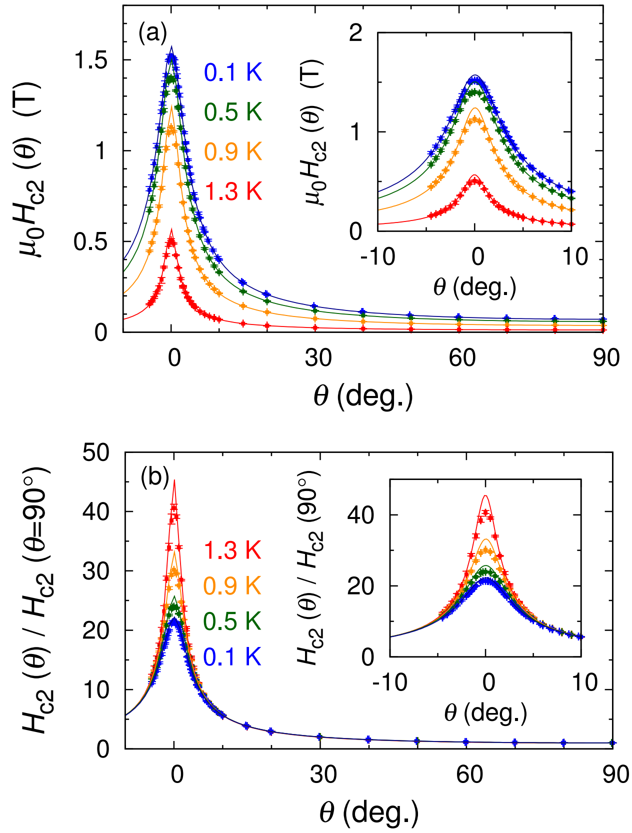

FIG. 4. (Color online) (a) Field-angle θ dependence of the upper critical field H_{c2} of Sr_2RuO_4 at various temperatures for ϕ=0°. (b) Angle θ dependence of H_{c2} normalized by $H_{c2}(\theta=90°)$. Solid curves are the fitting results using Eq. (3) in the range $2.5° \leq |\theta| \leq 90°$. Insets are enlarged views near θ=0°.

In order to characterize the nonlinear temperature dependence of H_{c2} in Sr_2RuO_4, we fitted $H_{c2}(t)$ for θ=0° by $a(1-t)^n$ with fitting parameters a and n. In any fitting range, n is obviously larger than n=0.5, which is expected for the two-dimensional (2D) superconductivity;[30] the fitting in the range $0.9 < t < 1$ yields n=0.9. In addition, the coherence length along the c axis ξ_c is estimated to be 3.2 nm using the Ginzburg-Landau (GL) equation

$$\xi_c = (\Phi_0 H_{c2\|c}/2\pi H_{c2\|ab}^2)^{1/2} \tag{2}$$

with $\mu_0 H_{c2\|ab}$=1.5 T and $\mu_0 H_{c2\|c}$=0.075 T. Here, Φ_0 is the flux quantum. This value of ξ_c is five times larger than the spacing of the conductive RuO_2 layers (0.62 nm).[2] Even if the WHH value $-0.7 dH_{c2}/dt|_{t=1}$=2.5 T is used for $H_{c2\|ab}$, ξ_c=1.9 nm is obtained. These facts indicate that the superconductivity of Sr_2RuO_4 cannot be classified as a 2D superconductivity.

Figure 4(a) represents the θ dependence of H_{c2} at various temperatures for ϕ=0°. The θ dependence of H_{c2} normalized by $H_{c2}(\theta=90°)$ is also plotted in Fig. 4(b). We found that,

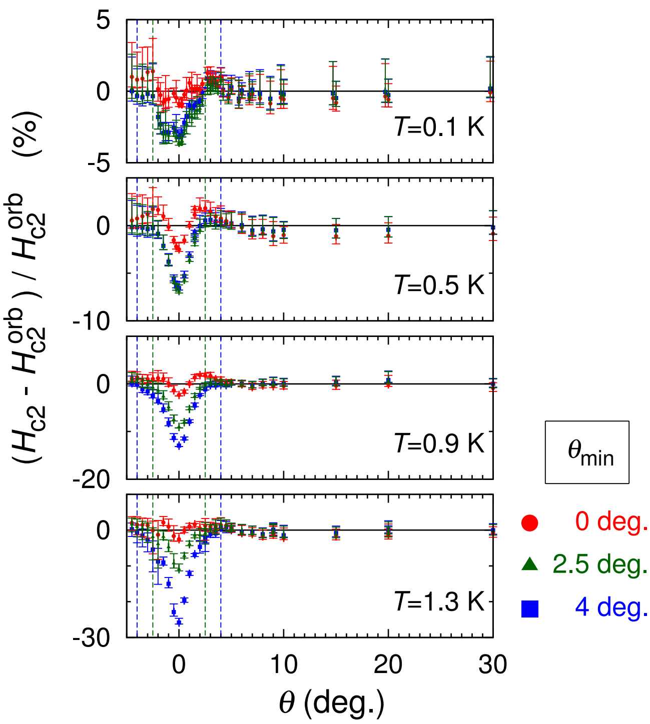

FIG. 5. (Color online) Error of the fitting of Eq. (3) to the $H_{c2}(\theta)$ data for $\phi=0°$ in the range $\theta_{min} \leq |\theta| \leq 90°$ for several θ_{min}. $\theta_{min}=2.5°$ is likely to give the most appropriate fitting range.

although $H_{c2}(\theta)/H_{c2}(90°)$ for $10° \leq |\theta| \leq 90°$ is nearly independent of temperature, it decreases on cooling for $|\theta| < 10°$.

We found that the observed $H_{c2}(\theta)$ is well explained by the GL theory for anisotropic three-dimensional (3D) superconductors,[31] if we allow the anisotropic ratio $\Gamma = H_{c2}(0°)/H_{c2}(90°)$ to depend on temperature. The angular dependence of H_{c2} is expressed as

$$H_{c2}^{orb}(T,\theta) = \frac{H_{c2\|ab}^{orb}(T)}{\sqrt{\Gamma(T)^2 \sin^2\theta + \cos^2\theta}}. \quad (3)$$

We fit Eq. (3) to the observed $H_{c2}(\theta)$ at temperature T_0 with two fitting parameters $H_{c2\|ab}^{orb}(T_0)$ and $\Gamma(T_0)$. We chose the fitting range as $\theta_{min} \leq |\theta| \leq 90°$ so that the range is as wide as possible while the fitting yields a good result in the whole chosen range. Figure 5 represents the error of the fitting for

TABLE I. The upper critical field and its anisotropy. Fitting parameters are obtained by the fit of $H_{c2}(\theta)$ for $\phi=0°$ using Eq. (3) with $\theta_{min}=2.5°$ at each temperature.

Experiment			Fitting parameters	
T (K)	$\mu_0 H_{c2\|ab}$ (T)	$H_{c2\|ab}/H_{c2\|c}$	$\mu_0 H_{c2\|ab}^{orb}$ (T)	Γ
0.1	1.517	21.4	1.574	22.1
0.5	1.399	23.7	1.504	25.5
0.9	1.130	30.5	1.243	33.2
1.3	0.496	41.3	0.568	46.1

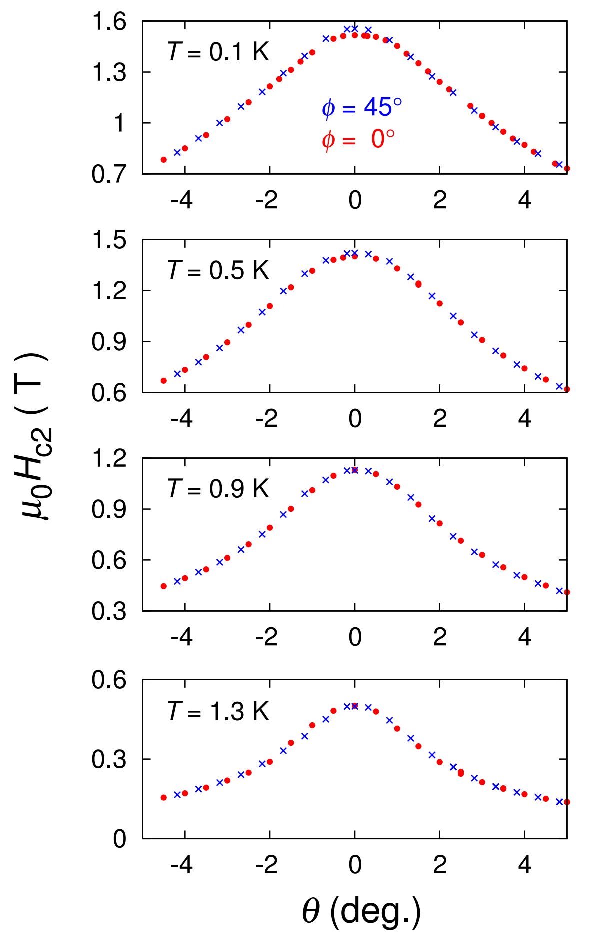

FIG. 6. (Color online) Comparison between $H_{c2}(\theta)$ at $\phi=0°$ (circles) and $\phi=45°$ (crosses).

different θ_{min} at each temperature. When $\theta_{min} \geq 2.5°$, $H_{c2}(\theta)$ is well fitted by Eq. (3) in the chosen fitting range. By contrast, for $\theta_{min} < 2.5°$, $H_{c2}(\theta)$ exhibits systematic deviation from Eq. (3) around $|\theta| \sim 2°$. Thus, we conclude that $\theta_{min}=2.5°$ is the most appropriate. The fitting results are plotted in Fig. 4 with the solid curves and the obtained fitting parameters are listed in Table I. Interestingly, the observed $H_{c2}(\theta)$ is fitted by Eq. (3) better at lower temperatures, as being clear in the inset of Fig. 4(b). This tendency is also clear when the fit ratio Γ is compared with the experimental ratio $H_{c2}(0°)/H_{c2}(90°)$. We should mention that the thin-film model[32] applied to a 2D superconductor,[33] in which $H_{c2}(\theta)$ exhibits a cusp at $\theta=0°$, cannot account for our data. While we carefully examined the θ dependence of H_{c2}, a kink in $H_{c2}(\theta)$ around $\theta=2°$ revealed by the specific-heat measurements at 0.1 K (Ref. 17) was not detected in the present study. We note that a kink in $H_{c2}(\theta)$ was not detected in the thermal-conductivity measurements at 0.32 K, either [Fig. 4(a) in Ref. 17]. On the basis of the presently available results, we cannot clarify why the kink in $H_{c2}(\theta)$ was observed only in the specific-heat measurement at 0.1 K.

In Fig. 6, we compare the θ dependence of H_{c2} at angles between $\phi=0°$ and $\phi=45°$. As indicated by Fig. 1(b), two onset features appear in the field dependence of $\Delta\chi'$ at $\phi=45°$, reflecting the fact that the present sample consists

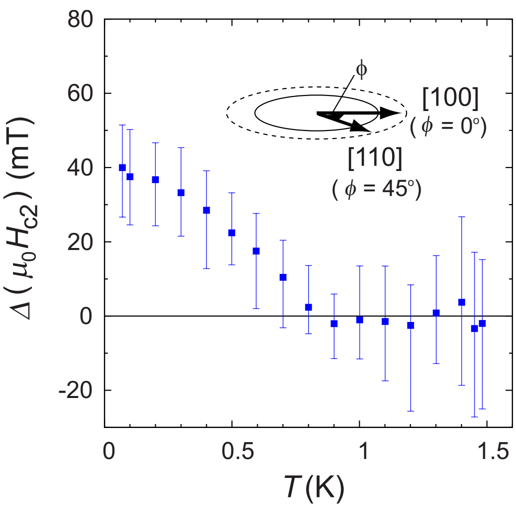

FIG. 7. (Color online) Temperature dependence of the in-plane H_{c2} anisotropy between ϕ=0° and ϕ=45° at θ=0°. $\Delta(\mu_0 H_{c2})$ is defined as $\mu_0 H_{c2}(\phi=45°)-\mu_0 H_{c2}(\phi=0°)$.

mainly of two domains. Since it is possible to separate the contribution from each of these domains, we plot in Fig. 6 $H_{c2}(\theta)$ for the major domain. From Fig. 6, we found that $H_{c2}(T,\theta)$ at ϕ=45° is both qualitatively and quantitatively similar to $H_{c2}(T,\theta)$ at ϕ=0°. Small difference in $H_{c2}(\theta)$ at angles between ϕ=0° and ϕ=45° is observed only when the magnetic field is applied nearly parallel to the *ab* plane. Figure 7 represents the temperature dependence of the in-plane H_{c2} anisotropy between ϕ=0° and ϕ=45°. Here, we define $\Delta(\mu_0 H_{c2})$ as $\mu_0 H_{c2}(\phi=45°)-\mu_0 H_{c2}(\phi=0°)$. Although the temperature T^* at which $\Delta(\mu_0 H_{c2})$ starts to increase on cooling depends on samples (0.9 K $\leq T^* \leq$ 1.2 K), $\Delta(\mu_0 H_{c2})$ of 40 mT at low temperatures is nearly the same among different samples with best T_c.[20]

On the basis of the phenomenological theory proposed by Gorkov,[34] superconductivity with a two-component order parameter, $k_x \pm ik_y$, should be accompanied by a substantial fourfold anisotropy in the in-plane H_{c2}. However, as presented in Fig. 7, no in-plane H_{c2} anisotropy is observable above about 1 K; the anisotropy grows on cooling, but reaches at most 3% at low temperatures. This lack of the large in-plane H_{c2} anisotropy is attributable to the multiband effect.[35–37] Because the directions of the gap minima are 45° different between the active (γ) and passive (α and β) bands,[38] the H_{c2} anisotropy reflecting the gap structure on different Fermi-surface sheets can be cancelled.[35,36]

IV. DISCUSSION

Let us discuss the origin of the H_{c2} limit in Sr_2RuO_4. For both 2D and 3D superconductors in which the main pair-breaking effect is due to the ordinary orbital effect, such a limit is not expected. Thus, in order to explain the H_{c2} limit, we need an additional pair-breaking mechanism.

One of the possible additional pair-breaking effects in Sr_2RuO_4 is an unusual orbital pair-breaking effect. For example, in a nearly 2D superconductor $(TMET\text{-}STF)_2BF_4$,[39] it is proposed that $H_{c2}(T)$ for $H \parallel a$ is limited due to the limit of the coherence length by the layer spacing, which leads to the decrease of the anisotropy ratio of H_{c2} on cooling. For Sr_2RuO_4, ξ_c estimated using Eq. (2), for which the ordinary orbital pair-breaking effect is assumed, is limited to be approximately 3.2 nm below 1 K. In fact, if we strictly apply Eq. (2), ξ_c takes a minimum at 0.8 K and even increases by about 3% at low temperatures. However, we cannot find a clear answer to this limit because the limited value of ξ_c, 3.2 nm, is five times larger than the layer spacing. Therefore, the origin of the apparent limit of the coherence length in Sr_2RuO_4 seems different from that in $(TMET\text{-}STF)_2BF_4$.

Recently, Machida and Ichioka proposed the Pauli effect as an additional pair-breaking effect leading to the H_{c2} limit in Sr_2RuO_4.[40] Using a model with a single-band spherical Fermi surface and by assuming the Pauli effect, they reproduced the observed $H_{c2}(\theta)$ at 0.1 K (Ref. 17) as well as field dependences of the specific heat[41] and magnetization.[42] Interestingly, we found that the Machida-Ichioka model well reproduces our results of $H_{c2}(T,\theta)$, too. Nevertheless, this would not lead to the conclusion that the H_{c2} limit in Sr_2RuO_4 is attributable to the Pauli effect because the Machida-Ichioka model overlooks some key experimental as well as theoretical facts. First, Machida-Ichioka model does not include the multiband effect. Their single-band model explains the field dependence of the specific heat at low temperatures. However, Sr_2RuO_4 has three cylindrical Fermi surfaces, α, β, and γ.[2,43] Although the active band γ is dominant in the superconductivity in high fields, the passive bands α and β also contribute to the superconductivity in low fields.[44] The contribution from the α and β bands is essential to explain the plateaulike dependence quantitatively.[44] In fact, inclusion of the multiband effect is needed to explain the T^2 dependence of the specific heat at low temperatures in zero field.[45–47] Second, as mentioned in Sec. I, the Pauli effect contradicts the results of the Knight-shift experiments.[3–5] Although they proposed the possibility that the spin part of the Knight shift was too small to be detected in the NMR experiments, the spin part at the Ru site is in reality as large as 4%.[4] In addition, the superconductivity was distinctly observed in $1/T_1$ through Ru NMR in the identical setup.[13] These facts exclude the possibility of the Pauli mechanism. Therefore, an alternative mechanism needs to be introduced to explain both the Knight-shift behavior and the H_{c2} limit.

V. SUMMARY

We have clarified the temperature and field-angle dependence of H_{c2} of Sr_2RuO_4. Our experiments were performed with an accurate and precise control of the applied magnetic field to avoid errors due to the misalignment. We revealed that the H_{c2} limit is clearly observed for $|\theta| < 5°$ and it occurs not only at low temperatures but also at temperatures close to T_c. We also found that, by assuming a temperature-dependent anisotropic ratio, the GL theory for an anisotropic 3D superconductor can explain the angular dependence of H_{c2} well, particularly at lower temperatures. The observed behavior of $H_{c2}(\theta)$ is qualitatively the same between ϕ=0° and ϕ=45°. Only a small in-plane H_{c2} anisotropy was observed at low temperatures, which disappears rapidly as the magnetic field direction leaves from the *ab* plane. Until now, the origin of the effective pair-breaking effect, which is compatible with both the invariance to the Knight shift and the limiting behavior of H_{c2}, remains unclear.

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