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Anomalous Enhancement of Upper Critical Field in Sr_2RuO_4 Thin Films

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We report large enhancement of upper critical field H_{c2} observed in superconducting Sr_2RuO_4 thin films. Through dimensional crossover approaching two dimensions, H_{c2} except the in-plane field direction is dramatically enhanced compared to bulks, following a definite relation distinct from bulk one between H_{c2} and the transition temperature. The anomalous enhancement of H_{c2} is highly suggestive of important changes of the superconducting properties, possibly accompanied with rotation of the triplet d-vector. Our findings will become a crucial step to further explore exotic properties by employing Sr_2RuO_4 thin films.

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Superconductors with a multicomponent order parameter, represented by spin-triplet superconductors, have attracted great interest as a ground of rich physics originating in the internal degrees of freedom. Among them, a layered-perovskite superconductor Sr_2RuO_4 has been a leading candidate possibly having chiral p-wave symmetry[1–3], which is one of topological superconducting states supporting Majorana modes at edges and vortices[4,5]. For further investigation and possible applications of the unique properties, the use of Sr_2RuO_4 thin films has been increasingly demanded in recent years[4,5].

In general, bulk superconducting state or pairing symmetry can be altered in the thin film form, affected by dimensionality change, inversion symmetry breaking, and/or epitaxial strain[6]. Spin-triplet superconducting states are characterized by d-vector, which represents the pair amplitude for the spin component perpendicular to the corresponding basis. Particularly in the case of Sr_2RuO_4, it has been theoretically suggested that the d-vector can flip from perpendicular (chiral p-wave) to parallel to the RuO_2 ab plane in the reduced dimensions, while the system still can host the Majorana modes[7]. Also, in helium-3 superfluid phases, changes of the p-wave order parameter have been experimentally demonstrated by mesoscopically confining it in a two-dimensional (2D) cavity[8]. In this context, it is indispensable to examine fundamental superconducting properties of Sr_2RuO_4 thin films. While growth of the superconducting films had been extremely challenging over the past decades since the discovery of Sr_2RuO_4[9,10], the reproducible and controllable growth has been recently achieved by refining molecular beam epitaxy techniques[11,12].

Upper critical field H_{c2} is one of the fundamental superconducting parameters related to superconducting symmetry, and thus has been intensively investigated in the study of Sr_2RuO_4 bulks[13–27]. While its behavior is generally consistent, some features have been interpreted as incompatible with the simple $p_x \pm ip_y$ model[5]. In particular, H_{c2} observed for the in-plane field direction is much more suppressed than expected at low temperatures, also accompanied with the first-order superconducting transition[13,14]. This suppression implies that H_{c2} for $H \parallel a$ might be affected by the paramagnetic pair breaking induced by the Zeeman splitting, called the Pauli limit[5].

Here we report detailed dependences of H_{c2} in Sr_2RuO_4 thin films, by measuring low-temperature magnetotransport systematically changing the field angle. The superconducting films are grown on a lattice-matched cubic substrate, yielding extremely limited defects in the films[11]. In addition to dimensional crossover confirmed in the field angle and temperature dependences, H_{c2} in the films is largely enhanced over a wide range of field angles except the in-plane direction, up to about four times the bulk value. This anomalous enhancement indicates that the triplet d-vector in thin films may be aligned on the ab plane, consistent with the recent theoretical prediction[7].

Superconducting single crystalline Sr_2RuO_4 films as displayed in Figs. 1(a) and (b) were epitaxially grown on cubic $(LaAlO_3)_{0.3}(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$ (LSAT) (001) substrates by oxide molecular beam epitaxy, following the same procedures detailed in Ref.[11]. Sr and Ru elemental fluxes were simultaneously supplied from a conventional Knudsen cell and an electron beam evaporator, respectively. The deposition was performed flowing distilled 100% ozone with a pressure of 1×10^{-6} Torr and heating the substrate at 900 °C. The film thickness is typically 50 nm along the c axis and the channel area of each sample is approximately 500 μm × 200 μm in the ab plane. Four-point measurements of the longitudinal resistivity were performed using low-frequency lock-in techniques with an excitation current of 3 μA along the a axis. Two samples were cooled down to 60 mK in a ^{3}He-^{4}He dilution refrigerator equipped with a superconducting magnet. As shown in Figs. 1(c) and (d), a superconducting transition with $T_c \sim 1.3$ K (onset) is confirmed for a typical sample. While the present films do not yet reach the high standard quality of Sr_2RuO_4 bulk single crystals[28], the transition temperature and its sharpness are now qualitatively comparable to the first reported bulk single crystal[1]. For field rotation in the ac plane, the samples were set on a single-axis rotating stage mounted on the mixing chamber.

Figures 1(e) and (f) show field dependence of the in-plane resistivity in a 50 nm thick Sr_2RuO_4 film, taken for $H \parallel a$ and $H \parallel c$ geometries at various temperatures down to 60 mK. Unlike the Ru eutectic phase[15,16] or uniaxially strained phase[17], hysteresis between the upward and downward sweeps is not detected in the resistivity. With increasing field, the resistivity changes from zero to a normal-state value due to the suppression of superconductivity through H_{c2}. Reflecting anisotropic superconductivity of this compound, the superconducting state is maintained up to higher fields for $H \parallel a$ than for $H \parallel c$.

Detailed field angle dependence of H_{c2} approaches 2D behavior with reducing the system thickness. Figure 2 compares field angle dependence between a Sr_2RuO_4 bulk[13] and the film, where the out-of-plane field angle θ is measured from the a axis. In the bulk, the angle dependence except for a very low angle region is well described by the following anisotropic 3D Ginzburg-Landau (GL) model[13,14].

$$\left(\frac{H_{c2}(\theta)\sin\theta}{H_{c2||c}}\right)^2 + \left(\frac{H_{c2}(\theta)\cos\theta}{H_{c2||a}}\right)^2 = 1 \tag{1}$$

H_{c2} is assumed to be dominated by the diamagnetic pair breaking process originating from the screening currents, known as the orbital limit. The coherence length along the c axis ξ_c, calculated from the GL expression $\xi_c = \sqrt{\Phi_0 H_{c2||c}/2\pi {H_{c2||a}}^2}$, is 3.2 nm, which is much larger than the lattice spacing of the RuO_2 layers. In this regard, superconductivity in the Sr_2RuO_4 bulk is not classified into ideal 2D systems[13]. On the other hand, the angle dependence in the 2D limit is explained by the Tinkham model.

$$\left|\frac{H_{c2}(\theta)\sin\theta}{H_{c2||c}}\right| + \left(\frac{H_{c2}(\theta)\cos\theta}{H_{c2||a}}\right)^2 = 1 \tag{2}$$

As shown in Figs. 2(c) and (d), angle dependence observed in the Sr_2RuO_4 film is fitted better by the 2D model. Assuming that both $H_{c2\|a}$ and $H_{c2\|c}$ are determined by the orbital limit as described by the GL equations, the effective superconducting thickness d is estimated at 23 nm from $d = \sqrt{6\Phi_0 H_{c2||c}/\pi {H_{c2||a}}^2}$. Considering that the film thickness is 50 nm, the film can be understood to be located in a dimensional crossover region.

In a very low angle region, H_{c2} seems suppressed compared to the 2D model. One possible origin of the deviation is the 2D-3D crossover. In such an intermediate superconducting state, the following empirical model interpolating Eqs. 1 and 2 has been proposed to explain the transitional angle dependence[29].

$$\alpha\left|\frac{H_{c2}(\theta)\sin\theta}{H_{c2||c}}\right| + (1-\alpha)\left(\frac{H_{c2}(\theta)\sin\theta}{H_{c2||c}}\right)^2 + \left(\frac{H_{c2}(\theta)\cos\theta}{H_{c2||a}}\right)^2 = 1 \tag{3}$$

The curve fitting is improved by adopting this model with α ranging about from 0.8 to 0.9 (for details see Supplemental Material[30]), also suggesting that the system is located in the crossover region. H_{c2} around $H \parallel a$ may be affected also by the presence of the Pauli limit, as discussed later.

Fig. 3(a) summarizes the H-T phase diagram obtained for the Sr_2RuO_4 film. Surprisingly, H_{c2} for $H \parallel c$ shows linear temperature dependence down to the lowest temperature without suppression as in the Werthamer-Helfand-Hohenberg (WHH) theory[31], as also clearly confirmed in the raw data in Fig. 1(f). The linear dependence without any suppression at low temperatures may be related to d-vector flipping from perpendicular to parallel to the ab plane in thin films, where the Pauli limit is no longer effective for the out-of-plane direction. For $H \parallel a$, H_{c2} follows the WHH-like curve but is rather weakly suppressed at low temperatures, in comparison to the bulk, as clearly seen in Fig. 3(b). Such a deviation from clear square-root temperature dependence expected in the 2D GL model has been also confirmed in other crossover systems showing the transitional field angle dependence with $0 < \alpha < 1$[32]. h^*, H_{c2} normalized by the initial slope at T_c, is saturated at about 0.64 for $H \parallel a$, which is even higher than the value of 0.42 measured for the bulk[13].

While the superconducting state approaches 2D like in the Sr_2RuO_4 thin film, the anisotropy ratio $\Gamma = H_{c2\|a}/H_{c2\|c}$ itself is reduced to 10 near T_c and 6 at the lowest temperature. This primarily results from increase in $H_{c2\|c}$, about four times over the bulk. As confirmed in Fig. 2(a), H_{c2} is anomalously enhanced over a wide range of field angles centered at $H \parallel c$. Figure 4(a) plots the correlation between $H_{c2\|c}$ and T_c for Sr_2RuO_4 bulks and films including previously reported other superconducting samples[9,11]. Almost independent of the sample quality, the bulk and film

$H_{c2\|c}$ follow each universal curve, which is roughly proportional to T_c^2 as expected for the orbital-limiting H_{c2}. In the case of dirty samples, ξ decreases with decrease of the mean free path l. This results in the extrinsic enhancement of H_{c2}, and this trend can be confirmed for MgB_2 and $YBa_2Cu_3O_7$ as positive correlation in the l-ξ plot in Fig. 4(b). In the case of clean samples, on the other hand, ξ increases with decrease of l, accompanied by the decrease of T_c or superconducting gap Δ_0. This trend appears as negative correlation in the l-ξ plot. The Sr_2RuO_4 films and also bulks independently show the clean-limit trend, excluding the extrinsic effects as a possible origin of increase of $H_{c2\|c}$.

By assuming that the GL in-plane coherence length $\xi_{ab} = \sqrt{\Phi_0/2\pi H_{c2\|c}}$ is equal to the Pippard one $\xi_{ab,0} = \hbar v_{F,ab}/\pi\Delta_0$ at the lowest temperature and using the superconducting gap relation $2\Delta_0 = ak_B T_c$, the following relation can be derived.

$$\frac{H_{c2\|c}}{{T_c}^2} = \frac{\pi\Phi_0}{8}\left(\frac{ak_B}{\hbar v_{F,ab}}\right)^2 \tag{4}$$

In the right hand side, material dependent parameters are only the coupling ratio a and the in-plane Fermi velocity $v_{F,ab}$. For example, if we assume the BCS limit $a = 3.5$ and take an experimental value of $v_{F,ab} = 9.3 \times 10^4$ m/s averaged on the active γ band[35,36], the dashed curve in Fig. 4(a) is obtained in rough agreement with but somewhat below the bulk trend, although a detailed analysis is surely dependent on the momentum-dependent gap structure[18] as well as the multi-band effect[19]. In the case of thin films, on the other hand, other intrinsic origins should cause the further enhancement of $H_{c2\|c}$ from the bulk trend. In terms of the epitaxial strain effect, a change in the in-plane lattice parameter compared to bulks is as small as -0.07% at room temperature[11], which can be further reduced to $+0.03\%$ at low temperatures[33,34]. In addition, angle-resolved photoemission spectroscopy on strained Sr_2RuO_4 films grown on various substrates has demonstrated that the in-plane effective mass shows weak monotonic dependence on the strain value (less than 5% for the 1% in-plane lattice change) for all the three bands[35], indicating that $v_{F,ab}$ is not a principal factor determining the enhancement. The uniaxial strain effect on bulks and films[12,17] is also excluded, as the present tiny strain is biaxial. Instead, an increase in the coupling ratio a (almost double) is one plausible origin. The enhancement of $H_{c2\|c}$ and the rather two-dimensional-like field-angle dependence are commonly observed in the films, regardless of the definition of T_c nor the film quality, as shown in Supplemental Material[30]. Because all the other possible origins, such as film quality, epitaxial strain, and quantum confinement, are carefully excluded, the enhancement of $H_{c2\|c}$ or a is most likely related to the observed dimensional crossover. Electrons may couple more strongly in the real space through the dimensional crossover, resulting in the shorter ξ_{ab}. Its microscopic mechanism will need to be further elucidated from theoretical aspects, while it may be also consistent with the recent theoretical prediction on two-dimensional Sr_2RuO_4 films, as discussed below.

While H_{c2} is largely enhanced centered at $H \parallel c$, it remains relatively low for $H \parallel a$. One origin of this difference is a change in the out-of-plane electronic structure by quantum confinement in films. However, an increase in the out-of-plane Fermi velocity $v_{F,c}$, which may account for the elongation of ξ_c, is less likely in terms of the mass enhancement due to the confinement. Another possible origin of this relative suppression is the Pauli limit. The presence of the Pauli limit for $H \parallel a$ is not generally consistent with the d-vector direction ($d \parallel c$) in the 2D $p_x \pm ip_y$ state[5]. Therefore, the suppression of $H_{c2\|a}$ suggests a change of the pairing symmetry, possibly accompanied with the d-vector flipping ($d \parallel ab$) suggested for thin films[7]. This is also consistent with the disappearance of the suppression newly observed in the temperature dependence of $H_{c2\|c}$ (Fig. 3(b)), indicating the absence of the Pauli limit for $H \parallel c$ in thin films.

In summary, we have revealed changes of the Sr_2RuO_4 superconducting state induced by confining it into thin films. Through the dimensional crossover, H_{c2} is intrinsically enhanced centered at $H \parallel c$ compared to bulks, while it remains suppressed for $H \parallel a$. The anomalous enhancement of H_{c2} suggests important changes of the spin-triplet superconducting state in the reduced dimensions. Taken together, these findings are compatible with the triplet state with the d-vector flipped parallel to the RuO_2 plane, which still could support the Majorana modes at edges and vortices[7]. Our study will provide the significant basis for further investigating superconducting properties of Sr_2RuO_4 thin films and applying its exotic states to junction devices.

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Figures

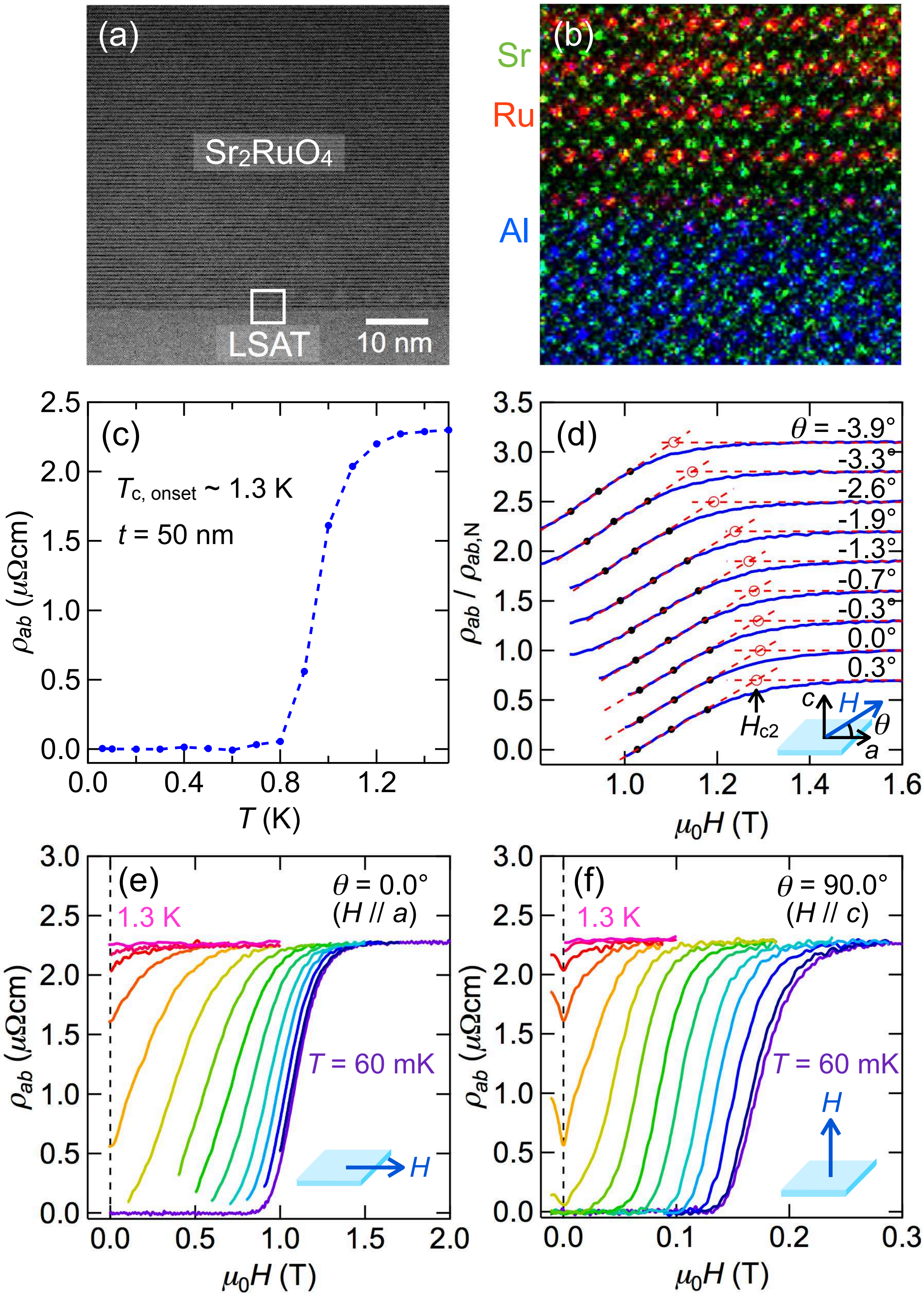
(a)
Sr2RuO4
LSAT
10 nm
(b)
Sr
Ru
Al
(c)
Tc, onset ~ 1.3 K
t = 50 nm
ρab (μΩcm)
T (K)
(d)
θ = -3.9°
-3.3°
-2.6°
-1.9°
-1.3°
-0.7°
-0.3°
0.0°
0.3°
Hc2
ρab / ρab,N
μ0H (T)
(e)
θ = 0.0°
(H // a)
1.3 K
T = 60 mK
H
ρab (μΩcm)
μ0H (T)
(f)
θ = 90.0°
(H // c)
1.3 K
T = 60 mK
H
ρab (μΩcm)
μ0H (T)

FIG. 1: Characterization of a superconducting Sr_2RuO_4 thin film. (a) Cross-sectional transmission electron microscope image and (b) its magnification in the boxed area, colored by energy dispersive x-ray spectrometry for Sr K, Ru L, and Al K edges. (c) Temperature dependence of the in-plane resistivity ρ_{ab}, taken for the Sr_2RuO_4 film with the transition temperature of $T_c \sim 1.3$ K (onset) and the film thickness of $t = 50$ nm. (d) Field dependence of ρ_{ab} measured around $\theta = 0^\circ$ at $T = 0.1$ K. Here, θ denotes the angle between the magnetic field and the a axis within the ac plane. The data are normalized by the normal-state in-plane resistivity $\rho_{ab,\mathrm{N}}$. An open circle represents H_{c2} defined as the intersection between two dashed lines extrapolated from normal ($\rho_{ab,\mathrm{N}}$) and superconducting (0.3–0.7$\rho_{ab,\mathrm{N}}$) regions. The points with resistivity of 0.3$\rho_{ab,\mathrm{N}}$, 0.5$\rho_{ab,\mathrm{N}}$, and 0.7$\rho_{ab,\mathrm{N}}$ are denoted by a filled circle. (e) and (f) In-plane ($\theta = 0^\circ$) and out-of-plane ($\theta = 90^\circ$) field dependence of ρ_{ab} at the lowest temperature of $T = 60$ mK and from 0.1 to 1.3 K at intervals of 0.1 K.

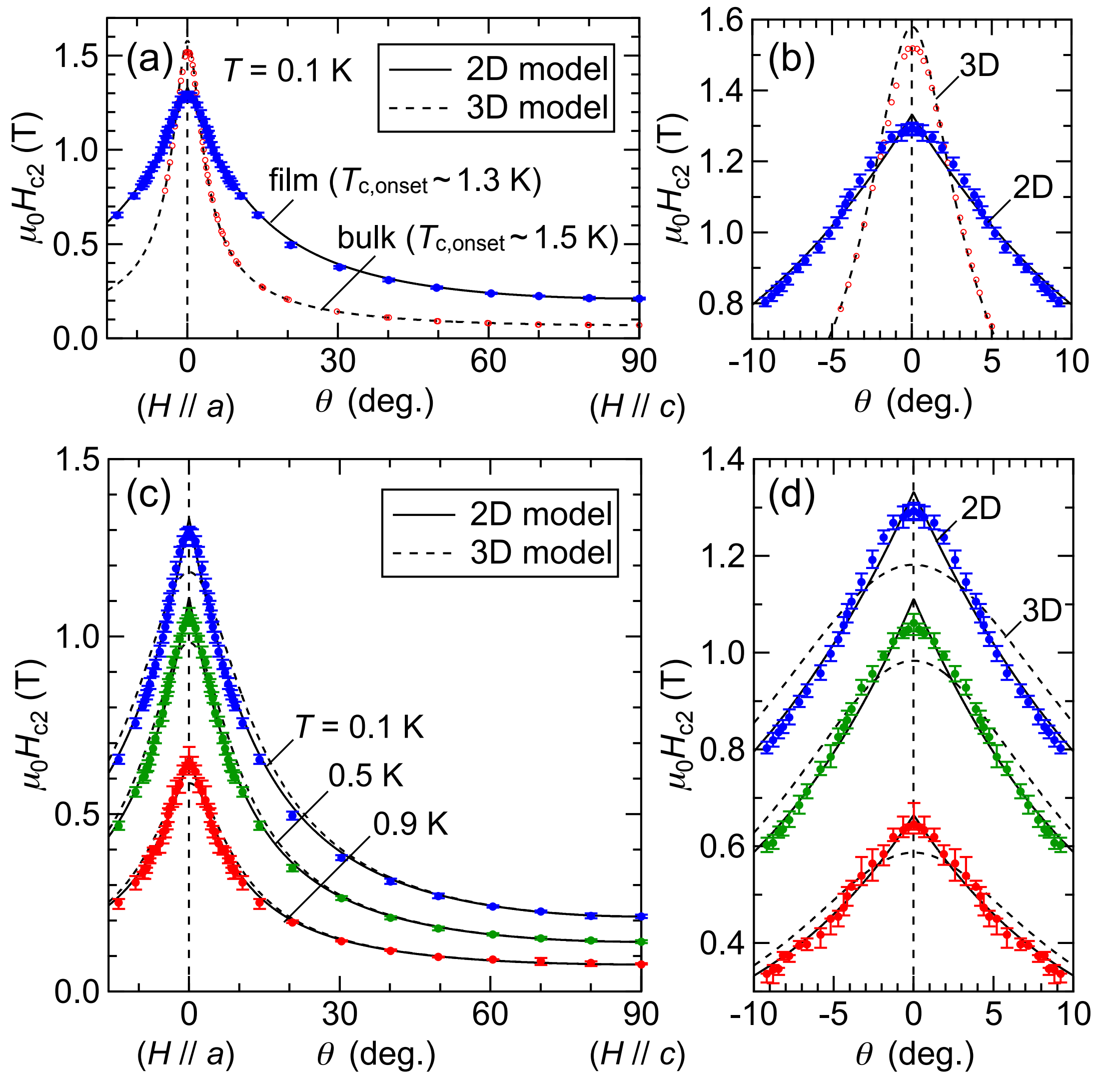

FIG. 2: Dimensional crossover of the Sr_2RuO_4 superconducting state. (a) Field angle dependence of H_{c2} in the Sr_2RuO_4 film at $T = 0.1$ K, compared to bulk one previously reported in Ref.[13]. Dashed and solid curves are fitting results using the three-dimensional (3D) Ginzburg-Landau (GL) anisotropic mass model (Eq. 1) and the two-dimensional (2D) Tinkham model (Eq. 2), respectively. An enlarged view centered at $\theta = 0^\circ$ is shown in (b). (c) and (d) Field angle dependence in the film at different temperatures fitted by the 2D and 3D models and its magnification around $\theta = 0^\circ$. The field angle dependence in the film is described better by the 2D model.

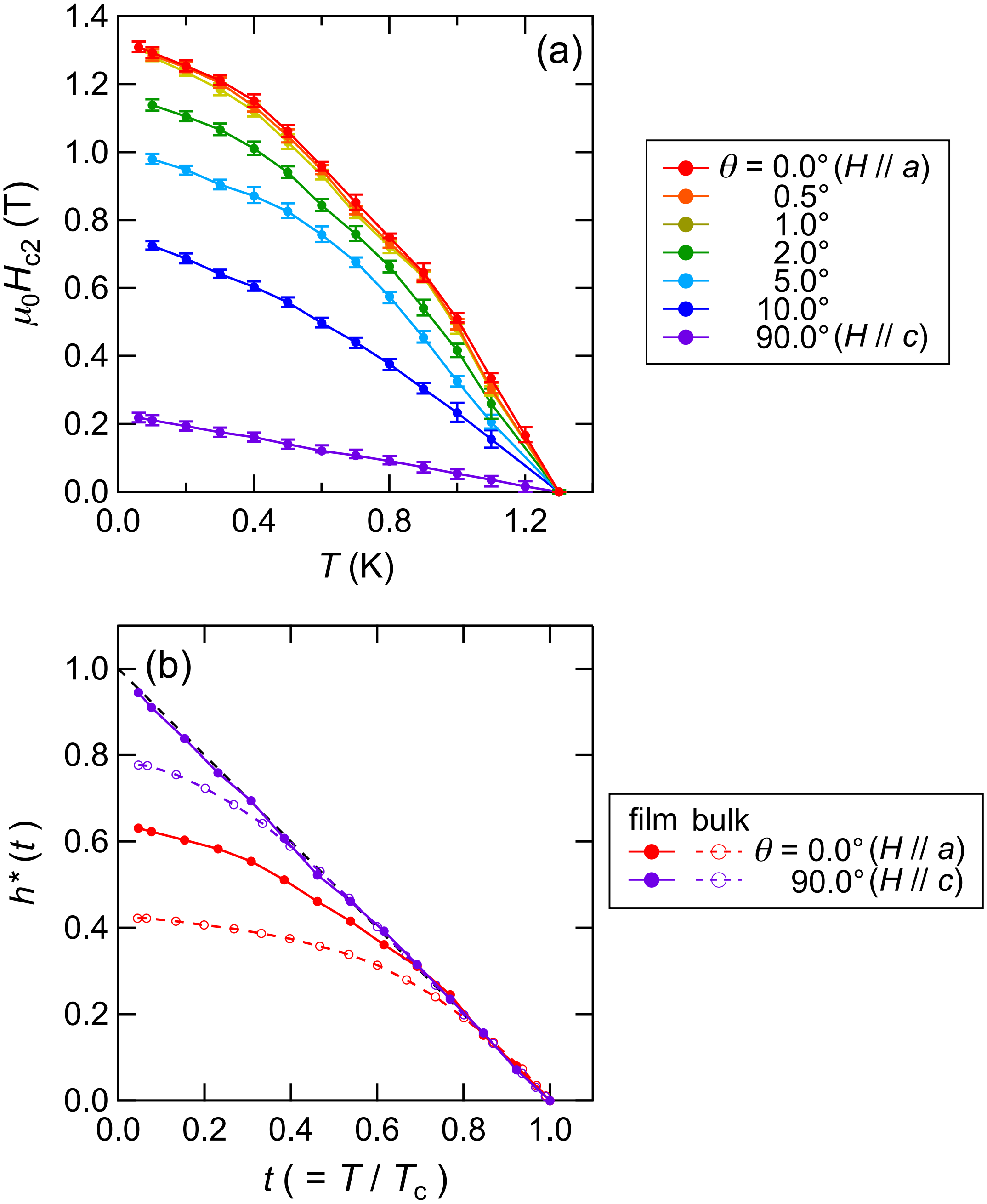

FIG. 3: Superconducting phase diagram in the crossover region. (a) H-T phase diagram of superconductivity in the Sr_2RuO_4 film at various field angles between $\theta = 0^\circ$ and 90°. (b) Temperature dependence of the normalized upper critical field $h^*(t)$ defined as $h^*(t) = -H_{c2}(t)/(\mathrm{d}H_{c2}/\mathrm{d}t)|_{t=1}$ ($t = T/T_c$), compared to the bulk one[13].

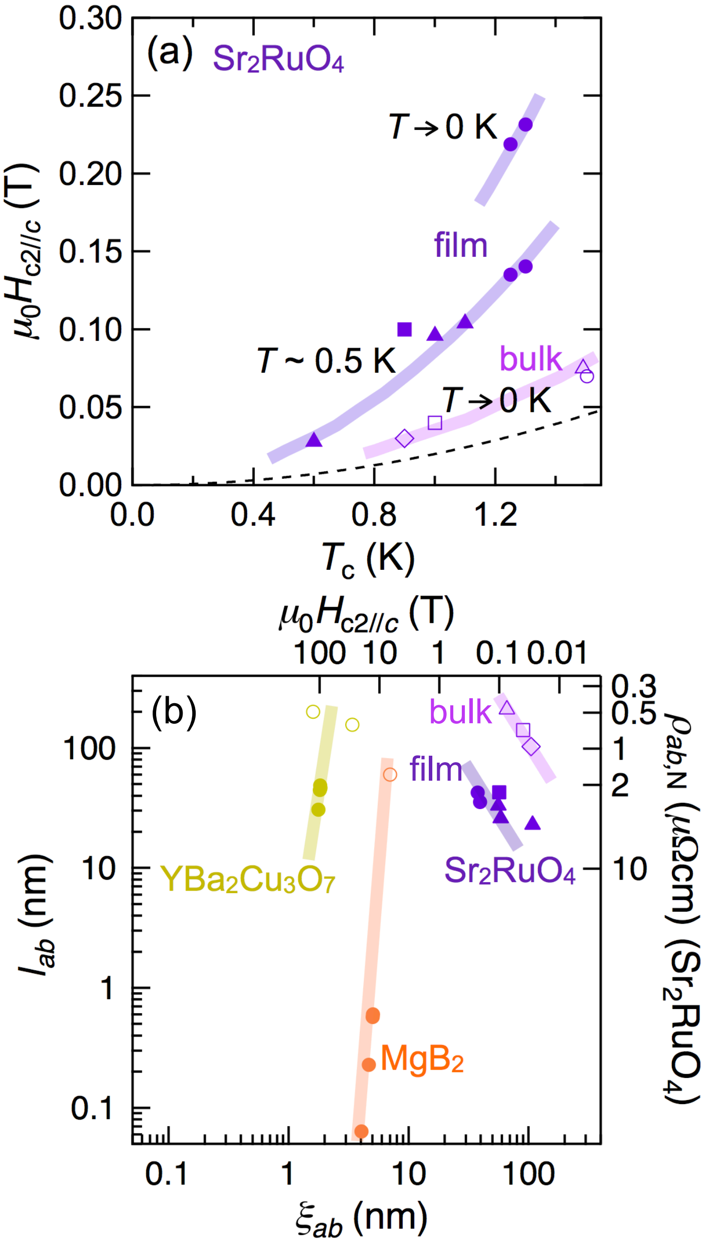

FIG. 4: Enhancement of upper critical field in thin films. (a) $H_{c2\|c}$ plotted as a function of T_c, including data previously reported for superconducting Sr_2RuO_4 bulks and films. As represented by the zero-temperature values deduced in this study, $H_{c2\|c}$ are systematically enhanced in thin films (● (present study), ▲[11], ■[9]), in comparison to bulk ones (○[13], △[22], □[23], ◇[24]). The dashed curve is calculated following Eq. 4. (b) Mean free path l_{ab} vs coherence length ξ_{ab} summarized for the Sr_2RuO_4 bulks and films. l_{ab} is estimated from the common-l approximation $l_{ab} = hc/2e^2\rho_{ab,\mathrm{N}} \sum_i k_{\mathrm{F},i}$, with the interlayer spacing $c/2$ and the i-th Fermi wave number $k_{\mathrm{F},i}$[22,37]. The corresponding $\rho_{ab,\mathrm{N}}$ and $H_{c2||c}$ are labeled on the right and top axes, respectively. For reference, data in MgB_2[38] and $YBa_2Cu_3O_7$[39,40] are also presented for bulks and films as denoted by open and closed symbols.