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Unusual temperature dependence of the in-plane critical field in Sr$_2$RuO$_4$

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Abstract. We have studied the upper critical field $H_{c2}$ of the spin-triplet superconductor Sr$_2$RuO$_4$. For $H \parallel c$, $H_{c2}$ increases linearly with decreasing temperature down to well below $T_c = 1.5$ K, consistent with the orbital depairing effect. However, for $H \parallel ab$, the increase of $H_{c2}$ is strongly suppressed below 1 K. The origin of this in-plane $H_{c2}$ limit has been actively discussed, but its mechanism is still unclear. In the present study, we performed ac susceptibility measurements in the applied fields with higher accuracy than in previous studies to obtain the field-temperature phase diagrams for $H \parallel c$ and $H \parallel ab$. We revealed that $H_{c2\parallel c}(T)$ does not exhibit a linear temperature dependence even above 1 K, which indicates that the $H_{c2}$ suppression is operative even close to $T_c$. In addition, we found that the $H_{c2}$ anisotropy ratio $H_{c2\parallel ab}/H_{c2\parallel c}$ decreases monotonically from 60 to 20 with decreasing temperature.

1. Introduction
Superconducting properties of Sr$_2$RuO$_4$ [1, 2] and UPt$_3$ [3] have been actively studied because of their unconventional pairing states. Both are now believed to be spin-triplet superconductors from the Knight shift measurements [4, 5, 6]. In fact nearly all the observations reported so far are consistent with the spin-triplet scenario [2]. However, there are a few puzzling phenomena. One of such puzzles is the upper critical field $H_{c2}$ limitation at low temperatures. In UPt$_3$ for $H \parallel c$, the increase of $H_{c2}$ is strongly suppressed with decreasing temperature [7]. Interestingly, Sr$_2$RuO$_4$ has rather similar $H_{c2}$ behavior for $H \parallel ab$ [8]. The origin of these $H_{c2}$ limits has not been clarified for a long time. In order to conclude on the pairing symmetry of Sr$_2$RuO$_4$ and UPt$_3$, this puzzle must be solved. Recently, K. Machida and M. Ichioda proposed that the Pauli paramagnetic effect is essential to understand the in-plane $H_{c2}$ limit of Sr$_2$RuO$_4$, based on a spherical Fermi surface model [9]. However, the Pauli paramagnetic effect occurs only for either the singlet pairing state or triplet pairing states with the $d$ vector locked in the basal plane. Therefore the proposed theory is not compatible with various experiments suggesting $d(k) = \hat{z} \Delta_0(k_x \hat{i} + ik_y \hat{j})$ in Sr$_2$RuO$_4$ [2, 4, 10, 11]. In order to identify the origin of the in-plane $H_{c2}$ limit of Sr$_2$RuO$_4$, detailed features of $H_{c2}$ need to be clarified.

Although there have been a number of reports on $H_{c2}$ of Sr$_2$RuO$_4$ [12, 13], the alignment of the applied magnetic field to the crystalline axes was not perfect in most experiments. Because of a large anisotropy in $H_{c2}$ of Sr$_2$RuO$_4$, a small misalignment would lead to a large difference in the value of $H_{c2}$ when the field direction is close to the $ab$ plane. Therefore, in order to identify the precise features of $H_{c2\parallel ab}$, a very accurate alignment of the applied magnetic field parallel to the $ab$ plane is essential. In this study, we measured the ac susceptibility with a very accurate alignment of the applied field and studied $H_{c2}$ of Sr$_2$RuO$_4$ quite precisely.
2. Experimental
We used a single crystal of Sr$_2$RuO$_4$ grown by a floating zone method, with dimensions of approximately 1.0 × 0.5 mm$^2$ in the $ab$ plane and 0.08 mm along the $c$ axis. The sample was annealed in oxygen at 1 atm and 1050 °C for a week in order to reduce the amount of oxygen deficiencies and lattice defects. The directions of the tetragonal crystallographic axes of the sample were determined from x-ray Laue pictures. The side surface of the sample was rotated from the (100) plane by about 10 degrees to avoid possible anisotropy effects due to surface superconductivity [14]. The ac susceptibility measurements revealed a sharp superconducting transition with the midpoint at $T_c=1.503$ K, which is one of the best $T_c$ of Sr$_2$RuO$_4$ reported [15].

We measured ac magnetic susceptibility $\chi_{ac} = \chi' + i\chi''$ by a mutual-inductance technique using a lock-in amplifier at 887 Hz. The sample was cooled down to 70 mK with a $^3$He-$^4$He dilution refrigerator. The ac magnetic field of 20 $\mu$T-rms was applied nearly parallel to the $c$ axis with a small coil. The dc magnetic field was applied using the “Vector Magnet” system [16], with which we can control the field direction three dimensionally and precisely. We determined the crystalline [100] and [001] axes from the anisotropy of $H_{c2}$. The accuracy and precision of the field alignment with respect to the $ab$ plane are better than 0.1 degree and 0.01 degree, respectively.

3. Results and Discussion
Figure 1 shows the dc field dependence of $\chi'$ for $H || [100]$ at 0.5 K and 2 K. Owing to the high sensitivity of the pick-up-coil, parasitic background contributes to the $\chi_{ac}$ signal. In order to obtain $\chi_{ac}$ contribution only from a superconductivity $\Delta\chi'$, we adopt $\Delta\chi'(T, H) = \chi'(T, H) - \chi'(2 K, H)$. As an example, the field dependence of $\Delta\chi'$ at 0.5 K is shown in the inset of Fig. 1.

![Figure 1](image1.png)

**Figure 1.** Field dependence of $\chi'$ at 0.5 K and 2 K for $H || [100]$. The inset represents $\Delta\chi'(0.5 K) - \chi'(2 K)$.

![Figure 2](image2.png)

**Figure 2.** Example of the definition of $H_{c2}$. $H_{c2}$ is defined as the intersection of the linear extrapolations in $\Delta\chi'$.

Here we define $H_{c2}$ as the intersection of the linear extrapolation of the most rapidly changing part of $\Delta\chi'$ and that of the normal state part, which is represented in Fig. 2. Using this definition of $H_{c2}$, we obtain the $H$-$T$ phase diagrams for $H || [100]$ and $H || [001]$, shown in Fig. 3.

In the vicinity of $T_c$, $H_{c2}$ parallel to the [001] axis increases linearly with decreasing temperature, as expected from the ordinary Ginzburg-Landau theory. From the Werthamer-Helfand-Hohenberg (WHH) theory [17, 18] and its extension to the $p$-wave superconductor [19], the reduced $H_{c2}$ is estimated using
the following equation:

\[ h^\ast(t) = - \frac{H_{c2}(t)}{dH_{c2}/dT|_{T=1}} \quad (t = T/T_c). \] (1)

For \( H \parallel [001] \) in our experiments, \( h^\ast(0) \) is estimated to be approximately 0.78, which well coincides with the theoretical expectation \( h^\ast(0)=0.727 \) in the clean limit [17], as well as the prediction for a \( p \)-wave superconductor [19]. In contrast, \( h^\ast(0) \) is estimated to be approximately 0.42 for \( H \parallel [100] \), which is much lower than the theoretical expectations. Here, the initial slope \( dH_{c2}/dT|_{T=1} \) was experimentally estimated in the temperature region between 1.3 K and \( T_c \).

In order to identify the characteristic temperature at which \( H_{c2} \) starts to deviate from a linear temperature dependence, we also plot the temperature dependence of the slope of the \( H-T \) phase diagram in Fig. 4. We evaluated the slope at the temperature \((T_1 + T_2)/2\) as \( \Delta H/\Delta T = [H_{c2}(T_1) - H_{c2}(T_2)]/(T_1 - T_2) \). Here, \( T_1 \) and \( T_2 \) are temperatures of adjacent data points. As represented in Fig. 4 as a dotted line, the slope for \( H \parallel [001] \) is almost constant from \( T_c \) down to 0.7 K and approaches zero at low temperatures, which is well explained by the WHH and the equivalent theories. In contrast, the slope for \( H \parallel [100] \) starts to vary toward zero at \( T_c \), which suggests that \( H_{c2} \) suppression is operative not only at low temperatures, but also even at temperatures close to \( T_c \).

Figure 5 shows temperature dependence of the anisotropy ratio \( H_{c2}[100]/H_{c2}[001] \). The anisotropy ratio is approximately 20 at low temperatures, which is consistent with the previous reports [8]. We found that the anisotropy ratio monotonically increases with increasing temperature and reaches 60 near \( T_c \). In other words, the anisotropy ratio is highest near \( T_c \) and it is suppressed at low temperatures. Until now, the origin of this suppression remains unresolved.

**4. Conclusion**

We studied the temperature dependence of the upper critical field \( H_{c2} \) of \( \text{Sr}_2\text{RuO}_4 \) from ac susceptibility measurements with a very accurate alignment of the applied magnetic field. We obtained the \( H-T \) phase diagrams for both \( H \parallel [001] \) and \( H \parallel [100] \) and revealed that the in-plane \( H_{c2} \) is suppressed even at temperatures close to \( T_c \). In contrast, \( H_{c2} \) parallel to the [001] axis exhibits ordinary behavior in the whole temperature region. We also found that the anisotropy ratio \( H_{c2}[100]/H_{c2}[001] \) is 20 at low temperatures and rises to approximately 60 near \( T_c \).
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**Figure 5.** Temperature dependence of the anisotropy ratio \( H_{c2} \parallel [100]/H_{c2} \parallel [001] \).