Detailed study of the ac susceptibility of Sr_2RuO_4 in oriented magnetic fields

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We have investigated the ac susceptibility of the spin-triplet superconductor Sr_2RuO_4 as a function of magnetic field in various directions at temperatures down to 60 mK. We have focused on the in-plane field configuration (polar angle $\theta \simeq 90°$), which is a prerequisite for inducing multiple superconducting phases in Sr_2RuO_4. We have found that the previous attribution of a pronounced feature in the ac susceptibility to the second superconducting transition itself is not in accord with recent measurements of the thermal conductivity or of the specific heat. We propose that the pronounced feature is a consequence of additional involvement of vortex pinning originating from the second superconducting transition.

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I. INTRODUCTION

Sr_2RuO_4 is a layered perovskite superconductor without copper.[1] Despite its superconducting transition temperature T_c being rather low (ideally 1.5 K),[2,3] Sr_2RuO_4 has been of great interest because of its unconventional spin-triplet pairing. Soon after the discovery of its superconductivity, the possibility of spin-triplet pairing was pointed out on theoretical grounds.[4,5] In fact, recent experiments have revealed its unconventional nature. In particular, the observation of spontaneous magnetic moments accompanying the superconducting state indicates broken time-reversal symmetry.[6] Besides this, NMR measurements have demonstrated that the Knight shift is unaffected by the superconducting transition, providing a definitive indication of spin-triplet pairing with the spin of Cooper pairs lying within the ab plane.[7]

One of the most interesting aspects of spin-triplet superconductivity is that multiple superconducting phases could be induced owing to the Cooper pairs possessing an internal degree of freedom. In fact, this is exemplified in UPt_3,[8] which is another spin-triplet superconductor. Although the details of the superconducting wave function of Sr_2RuO_4 are still controversial,[9–13] simple consideration based on existing experimental results will allow one to understand that the superconducting symmetry is probably represented by the degenerate two-component order parameter $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0(k_x + ik_y)$.[6,7,14,15] Agterberg[16] theoretically suggests that under such circumstances, this degeneracy in energy will be lifted in a magnetic field H parallel to the ab plane, leading to another superconducting phase with $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0 k_{x'}$ being induced above a certain field H_2, where $H // x'$. Agterberg also predicted that the appearance of this phase will be accompanied by an enhancement of the in-plane fourfold anisotropy of the upper critical field.[16]

The occurrence of a second superconducting phase in Sr_2RuO_4 was first suggested by measurements of the ac susceptibility and the specific heat, which were reported in our previous paper.[17] We have attributed a clear kink in the ac susceptibility to the second superconducting transition and interpreted anomalous behavior in the electronic specific heat as entropy release due to the second superconducting transition. The main conclusions of the previous[17] paper may be summarized in the following way. The second superconducting transition occurs at a field slightly lower than the upper critical field H_{c2} only when the magnetic field is applied accurately parallel to the ab plane. Concomitantly, the in-plane anisotropy of H_{c2} is significantly enhanced. Also a slight misalignment of the angle between the magnetic field and the ab plane causes both the second superconducting transition and the enhancement of the in-plane anisotropy to be suppressed. Agterberg's theoretical prediction[16] receives partial support from these experimental facts. Recent measurements of the specific heat[9] and of the thermal conductivity[18] have also detected a steep change attributable to the second superconducting transition, resulting in a field-temperature (H-T) phase diagram similar to the one deduced from the ac susceptibility measurements.[17]

Nevertheless, there are significant discrepancies between theory and experiment in the H-T phase diagram. For example, according to previous studies of the ac susceptibility,[17] the specific heat[9] and the thermal conductivity,[18] the phase boundary between the two superconducting phases seems to merge with the upper critical field line at a bicritical point close to $H = 1.2$ T, $T = 0.8$ K. On the contrary, there should not be a bicritical point theoretically;[16] the merging point is expected to be at $H = 0$, $T = T_c$.

In this paper, we have extended to 60 mK the ac susceptibility measurements on Sr_2RuO_4 in magnetic fields along various directions, subsequent to the study by Mao *et al.*;[17] the measurements in Ref. 17 were carried out down to 0.35 K. We will compare in detail the signs of the second superconducting transition obtained by various experimental probes.

II. EXPERIMENT

In our previous study of the ac susceptibility and the specific heat of Sr_2RuO_4,[17] we investigated three single crystals with different shapes chosen from different batches. In this paper, of those three samples, we concentrate on the sample referred to as sample B in Ref. 17 for detailed studies. The sample is a single crystal of Sr_2RuO_4 which was grown by a floating-zone method with an infrared image furnace.[19] The

sample was polished into a rectangle such that a side surface of the sample and the *a* axis make an angle of about 25°. The size was $1.9\times1.3\times0.5$ mm^3, with the shortest dimension along the *c* axis. The sample was annealed in oxygen at 1050 °C for three weeks in order to reduce the amount of defect. A measurement of the ac susceptibility shows a sharp superconducting transition at $T_c=1.46$ K (midpoint). An x-ray rocking curve of the sample shows the characteristics of a single crystal of high quality; the diffraction peak width [full width at half maximum (FWHM)] being comparable to that of a Si crystal (with FWHM of 0.06°) in the diffractometer. The directions of the tetragonal crystallographic axes of the sample were determined by x-ray Laue pictures.

Low temperatures down to 60 mK were obtained by means of a dilution refrigerator. Temperatures were measured using a RuO_2 resistor. Magnetic fields of up to 2 T, generated by a superconducting solenoid, were applied to the sample.

Measurements of the ac susceptibility were done by a mutual-inductance method with an alternating field of 50 μT at frequencies of 700–1000 Hz; the measurement frequencies were carefully chosen to ensure that frequency-dependent artifact was not involved. The ac modulation field was applied along the *c* axis. The sample was mounted in a double-axis rotator that enables both the polar and azimuthal angles to be changed independently with a precision of 0.01°. (For referring to the direction of the applied magnetic field with respect to the crystallographic axes, we shall introduce the polar angle θ, for which $\theta=0°$ corresponds to the [001] direction, and the azimuthal angle ϕ, for which $\phi=0°$ with $\theta=90°$ corresponds to the [100] direction.) It is to be noted that the tetragonal symmetry of the crystal structure is conserved down to temperatures as low as 110 mK.[20]

III. RESULTS AND DISCUSSION

Figure 1(a) shows the ac susceptibility of Sr_2RuO_4 as a function of magnetic field parallel to the [110] direction, with an accuracy of $\Delta\theta\leqslant0.02°$, at several temperatures. As in Ref. 17 three prominent features, labeled P_1, P_2, and P_3, are seen. Their definitions are illustrated in Figs. 1(a) and 1(b); we follow the definitions used in our previous study.[17] (i.e., we define H_{c2} as the intersection of the linear extrapolation of the most rapidly changing part of the real part of the ac susceptibility ($\chi=\chi'+i\chi''$) and that of the normal state.[21,22]) P_1 and P_3 correspond to the upper critical field H_{c2} and the second peak H_p due to vortex synchronisation,[23] respectively. These two features are seen at all the temperatures. On the other hand, the feature P_2, which was interpreted as a manifestation of a second superconducting transition,[17] appears only below ~0.7 K. These observations confirm the results of Mao *et al.*[17] that cover temperatures down to 0.35 K; we have shown that the feature P_2 persists at least to 60 mK. Along the line of the discussion in the Agterberg theory,[16] the feature P_2 has been considered to represent a second-order transition from a state with a nodeless gap to a state with lines of nodes (when the applied field is increased). In contrast, as shown in Fig. 1(b), when the applied field is along the [100], only the two features P_1 and P_3 are clearly seen, and P_2 is not observed in the ac susceptibility even at a temperature as low as 60 mK. This is also consistent with our previous results.[17]

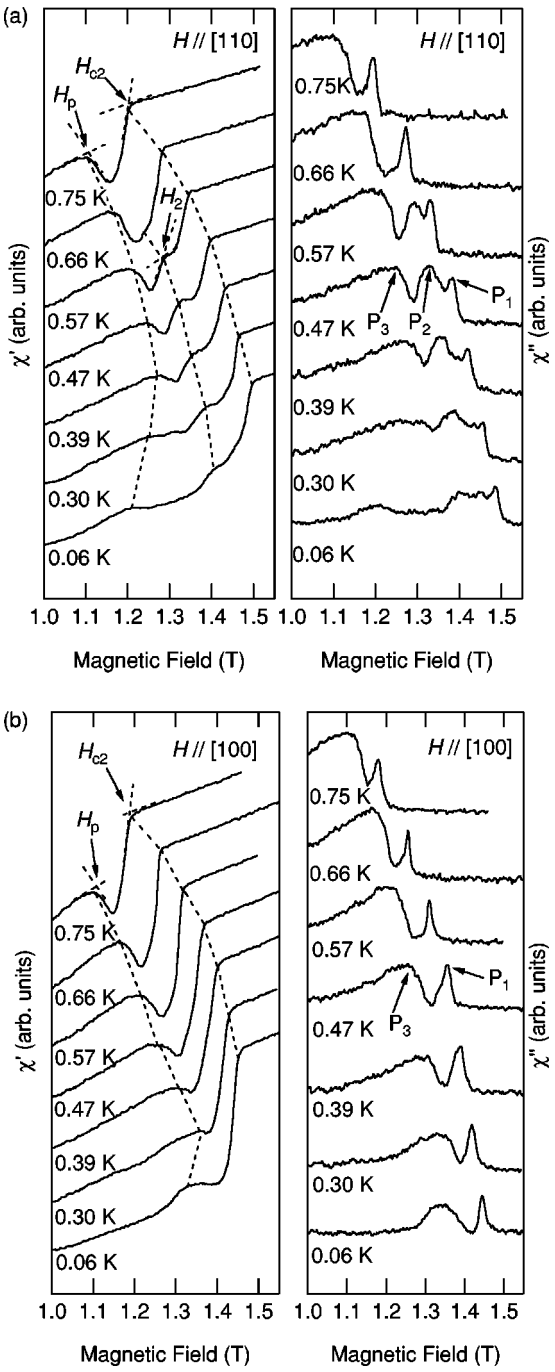

FIG. 1. Ac susceptibility ($\chi=\chi'+i\chi''$) at various temperatures (a) for *H*// [110] and (b) for *H*// [100]. Whilst P_1, P_2, and P_3 are all observed for *H*// [110], only P_1 and P_3 are seen for *H*// [100]. The ac-field amplitude used is 50 μT. Traces have been offset for clarity.

In order to see how these features change in field position, the characteristic fields H_{c2}, H_2, and H_p are plotted against the azimuthal angle ϕ (for $\theta=90°$, $T=60$ mK) in Fig. 2. All of the three features exhibit clear fourfold anisotropy, apart from that P_2 is not observed over $\sim20°$ around the [100]

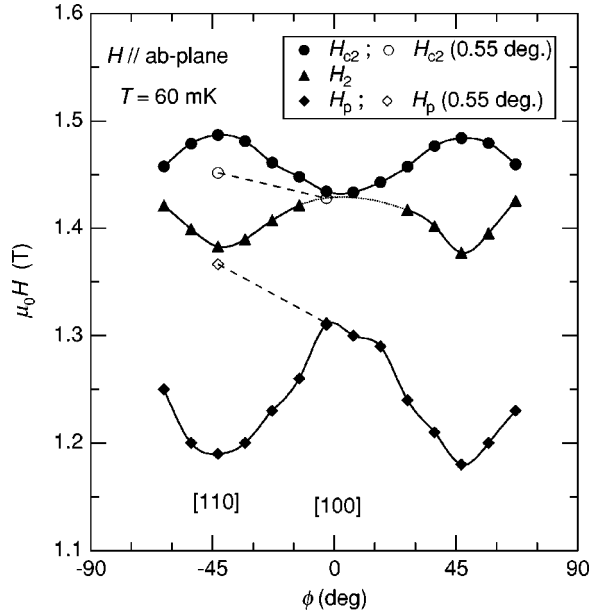

FIG. 2. Azimuthal-angle ϕ dependence of H_{c2}, H_2, and H_p with the magnetic field parallel to the ab plane ($T=60$ mK). H_{c2} is the upper critical field, H_2 is related to the second superconducting transition and H_p is the second peak due to vortex synchronization. All of these characteristic fields show clear fourfold symmetry. Smooth curves (solid and dotted lines) through each set of the data points are employed as guides to the eye. The open symbols represent H_{c2} and H_p with a slight misalignment of $\Delta\theta=0.55°$. Straight lines (dashed lines) are employed between these symbols.

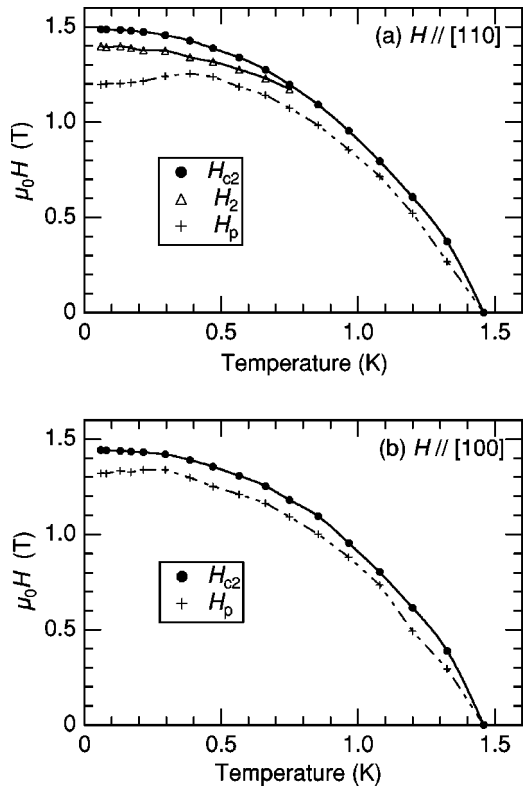

FIG. 3. Field-temperature phase diagrams (a) for H// [110] and (b) for H// [100]. Whilst H_p appears for both configurations, H_2 appears only for H// [110].

direction. The azimuthal-angle dependences of H_2 and H_p are both out of phase with that of H_{c2}. H_2 appears to merge into H_{c2} and is not seen in the vicinity of $\phi=0°$. The in-plane anisotropy of the upper critical field is 3.5% at 60 mK.

Figure 3 shows the H-T phase diagrams for the H// [100] case and for the H// [110] case as contrasting examples. Whereas the Agterberg theory[16] suggests that the second superconducting transition occurs in both cases, the feature P_2 disappears for H// [100]. It should be noted that hysteresis was hardly observed between the upward and downward field sweeps; no obvious evidence for a first-order transition was seen.

Contrary to the results of the ac susceptibility, a very recent study of the specific heat,[24] made subsequent to the work in Ref. 9 has obtained definitive evidence for the second superconducting transition in magnetic fields along the [100] direction; they have observed a clear split at the superconducting transition in the temperature dependence of the specific heat. This observation shows a good agreement with a recent study of the thermal conductivity.[18] Besides, the latter study suggests that the azimuthal-angle dependence of H_{c2} and the second superconducting transition field show fourfold symmetry,[18] but are in phase with H_{c2}, in apparent disagreement with the present ac susceptibility measurements. The clear split in the specific heat[24] provides *thermodynamic* evidence for a phase transition, and the transition point should not depend on the probe used. Therefore, these significant discrepancies inevitably cast doubt upon the attribution of P_2 seen in the ac susceptibility to the second superconducting transition itself.

In connection to Fig. 3, it is worth mentioning that the temperature dependence of H_p at low temperatures below $\sim$0.4 K is unusual. H_p drops with decreasing temperature; the H_p line looks repelled by the H_2 line. This occurs immediately below the second superconducting phase in the H-T phase diagram. (e.g., H-T phase diagram shown in Fig. 8 of Ref. 18) A similar tendency can be seen in a close region of the phase diagram for H// [100] despite the absence of P_2. Even though P_2 does not necessarily represent the second superconducting transition itself, the tendency suggests that there is a strong correlation between the second superconducting transition and the feature P_2.

Also the thermal conductivity[18] and the specific heat[9] indicate that the second superconducting transition is induced in fields parallel to the ab plane and that even a slight misalignment of the field suppresses the second superconducting transition. In Fig. 4, the H-T phase diagrams for H// [110] and H// [100] but with an intentional misalignment of $\Delta\theta=0.55°$ are presented, where the feature P_2 is suppressed. Concomitantly, the unusual temperature dependence of H_p seen in Fig. 3 is also suppressed. This fact supports that there is another correlation between the second superconducting transition and the feature P_2 seen in the ac susceptibility. These two correlations are, in principle, common to both the [110] and [100] directions, which is consistent with that the second superconducting transition occurs for both H// [110] and H// [100] as evidenced by the specific heat and the thermal conductivity.

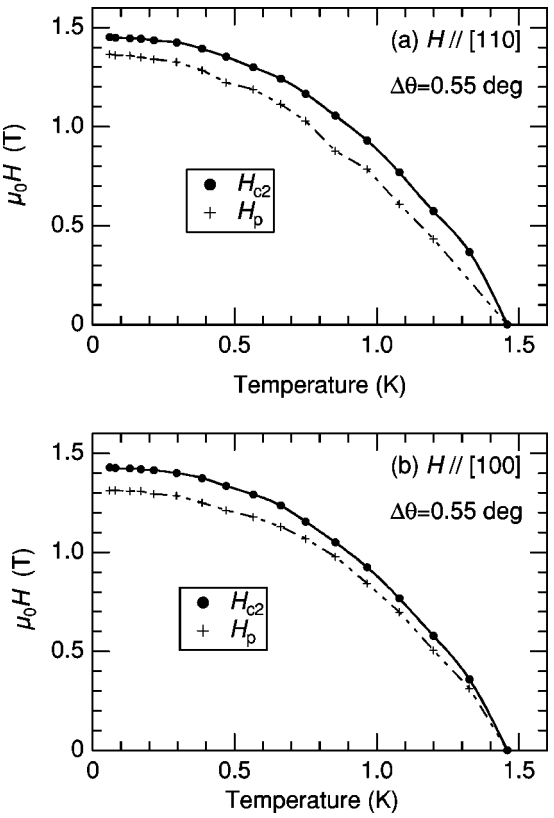

FIG. 4. Field-temperature phase diagrams (a) for H// [110] and (b) for H// [100] in misaligned fields ($\Delta\theta = 0.55°$).

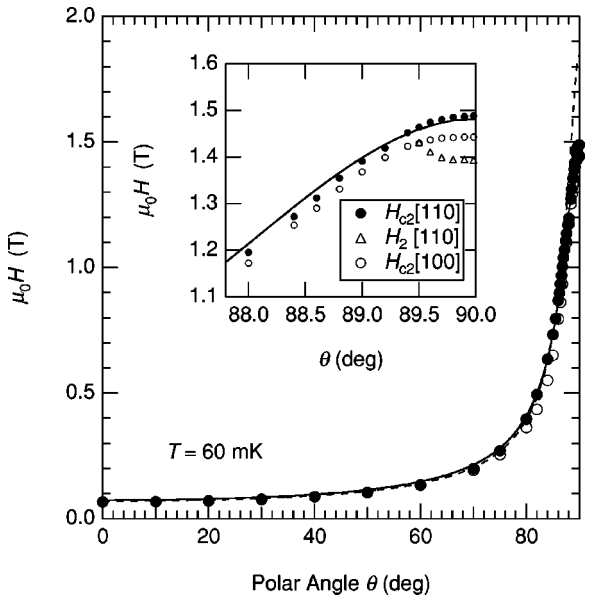

FIG. 5. Polar-angle θ dependence of H_{c2} and H_2 with the field inclined towards the [110] and [100] directions ($T = 60$ mK). The inset shows a blow-up of the main panel. The solid line and the dashed line represent fits of the Ginzburg-Landau anisotropic effective-mass approximation [Eq. (2)] to the data for $0° \leqslant \theta \leqslant 90°$ and for $0° \leqslant \theta \leqslant 85°$, respectively.

Figure 2 additionally shows H_{c2} and H_p when the field is along the [110] and [100] directions but with a misalignment of $\Delta\theta = 0.55°$ (open symbols). The effects of this slight misalignment are rather strong: (1) The in-plane anisotropy of the upper critical field is reduced to 1.6%. (2) H_2 disappears. (3) H_p seems to have a fourfold symmetry, being *in phase* with H_{c2}.

As Mao *et al.*[17] has suggested, the very accurate alignment of the magnetic field to the ab plane is essential for inducing the second superconducting transition. The enhanced anisotropy of $H_{c2}(\phi)$ and the second superconducting transition both simultaneously appear and disappear when the polar angle θ is varied across 90°. As the enhanced anisotropy of $H_{c2}(\phi)$ is theoretically expected to accompany the second superconducting transition,[16] the observed correlation between these supports the application of the Agterberg theory at least in a qualitative fashion.

Next we discuss another aspect that seems to be closely related to the second superconducting transition; the upper critical field H_{c2} immediately above the second superconducting phase is considerably lower than expected values. There are at least two ways of describing this aspect. One is via the dependence of H_{c2} on the polar angle θ. The other is via the shape of the H-T phase diagram (i.e., comparison with a linear extrapolation of the gradient near T_c).

First, we show in Fig. 5 the dependences of H_{c2} and H_2 on the polar angle θ, with the field inclined towards the [110] and [100] directions. The inset of Fig. 5 illustrates a blow-up of the main panel. Clearly, a misalignment of $\sim 0.5°$ is large enough to suppress the feature P_2. Although the measurement temperature 60 mK is considerably lower than the transition temperature 1.46 K, we apply the Ginzburg-Landau anisotropic effective-mass approximation:[25]

$$H_{c2}(\theta) = \frac{H_{c2}(\theta = 0)}{\sqrt{\cos^2\theta + \Gamma^{-2}\sin^2\theta}}. \tag{1}$$

Here Γ is the square root of the ratio between the effective masses for interplane and in plane motion, or the ratio between the upper critical fields for in plane and interplane. Two kinds of fitted curves for H// [110] are shown together with the experimental data in Fig. 5. The solid curve represents a fit of Eq. (1) to the data for $0° \leqslant \theta \leqslant 90°$ whilst the dashed curve represents a fit for $0° \leqslant \theta \leqslant 85°$. The solid curve as a whole seems to reasonably reproduce the experimental data. The obtained value for Γ is 20.1 for H// [110]. The same fitting to the data for H// [100] yields $\Gamma = 22.0$. These values are in agreement with a previous study,[26] in which Γ is estimated to be 20. However, it is seen that the solid curve slightly but systematically deviates from the experimental data at low values of θ. On the other hand, the dashed curve (obtained from $0° \leqslant \theta \leqslant 85°$) in Fig. 5 fits better to low θ data at the expense of the high θ region. In fact, the fitting for $0° \leqslant \theta \leqslant 85°$ yields $H_{c2}(90°) = 1.85$ T; the experimentally obtained value for $H_{c2}(90°)$ being 1.48 T. These facts imply the suppression of H_{c2} in a certain range of θ close to 90°. Since the second superconducting transition occurs for θ close to 90° exclusively, this suggests correlation between the second superconducting transition and the suppression of H_{c2}.

Second, we use a formula such as the Werthamer-Helfand-Hohenberg (WHH) formula[27] to demonstrate the suppression of H_{c2}. Although the WHH formula is intended for orbital depairing in a weak-coupling BCS-type superconductor in the dirty limit, we, for reference, apply the formula

$$\mu_0 H_{c2}(0) = -0.693 \mu_0 \left. \frac{dH_{c2}}{dT} \right|_{T=T_c} T_c, \qquad (2)$$

to the H-T phase diagrams shown in Figs. 3(a), 3(b), 4(a) and 4(b). Lebed and Hayashi[28] also obtained a WHH-like formula for a quasi-two-dimensional p-wave superconductor with an isotropic gap in fields parallel to the ab plane. The resultant formula in Ref. 28 is identical to Eq. (2), but with the coefficient being -0.75 instead of -0.693. In reality, the application of the WHH formula to the present H-T phase diagrams leads to the upper critical field being rather overestimated by a factor of 2–2.5. In other words, the upper critical field at very low temperatures appears to be suppressed compared to that expected from the orbital depairing. This occurs immediately above the second superconducting phase. This also seemingly suggests that the emergence of the second superconducting transition is closely related to the suppression of low-temperature upper critical fields. However, the H-T phase diagram for $\Delta\theta = 0.55°$, at first sight, seems to be a counter example. Possible implications of this will be discussed later.

In contrast, the H-T (H// ab-plane) phase diagram established in Ref. 23 by Yoshida *et al.* from ac susceptibility measurements is well explained by the WHH formula. Also the H-T (H// c-axis) phase diagram obtained by Mackenzie *et al.* from resistive measurements[29] fits the same formula very well. As these two works both use samples with T_c of $\sim$0.9 K, the discrepancy between the present work and Refs. 23,29 may be due to the difference in sample quality. However, it is still unclear whether the second superconducting phase can be induced in samples with T_c of $\sim$0.9 K. In Ref. 23, the ac susceptibility data taken in magnetic fields parallel to the ab plane do not show any signs of the second superconducting transition such as P_2. The possible reasons we suggest are too weak ac-field amplitude (they used ac amplitudes of $3-7$ μT), too large misalignment (they estimated the actual misalignment to be less than 1.5°), and/or the relatively poor sample quality ($T_c \simeq 0.9$ K).

We now discuss differences in results from various experimental probes, in reference to the second superconducting transition. The greatest difference is the absence of P_2 in the ac susceptibility for H// [100] whilst the specific heat measurements have observed a clear split at the superconducting transition in fields along the [100] direction.[24] As previously mentioned, this implies that the feature P_2 in the ac susceptibility does not necessarily represent the second superconducting transition itself albeit P_2 is closely related to the second superconducting transition. A possible interpretation for the feature P_2 will be that P_2 is a consequence of another peak effect due to the second superconducting phase.

In order to support this interpretation, we show the dependence of the ac susceptibility on the ac-field amplitude. Figure 6 shows the real and imaginary parts of the ac susceptibility with different ac amplitudes.[30] The application of an ac-field above $\sim$100 μT hampers reliable thermometry. The applied dc field direction and the temperature have been chosen to be the [110] direction and 0.43 K, respectively, so that the feature P_2 is rather clear. The feature P_2 strongly depends on the ac-field amplitude, which is particularly clear in the imaginary part, whilst the upper critical field H_{c2} hardly depends on the ac amplitude. P_2 can be seen only above an ac-field amplitude of $\sim$25 μT and shifts to lower magnetic fields with increasing ac amplitude. It is reported that in the imaginary part of the ac susceptibility of $2H$-$NbSe_2$, the peak position is sensitive to the amplitude of the ac field whilst that the upper critical field is hardly affected by the ac amplitude.[31] Therefore, the behavior of P_2 observed in Fig. 6 strongly suggests that the feature P_2 arises from flux pinning rather than from a phase transition itself.

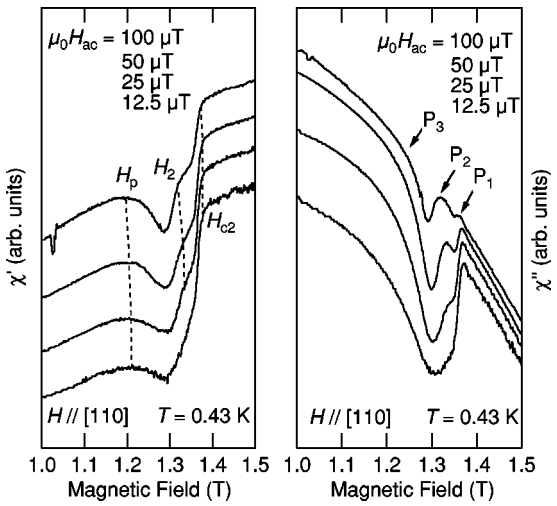

FIG. 6. Dependence of the ac susceptibility ($\chi = \chi' + i\chi''$) on the ac-field amplitude. The data were taken in dc magnetic fields parallel to the [110] direction at 0.43 K. The variation of the appearance of P_2 with ac-field strength indicates that P_2 involves flux pinning. Traces have been offset for clarity.

The feature P_2 in the real part of the ac susceptibility may be regarded as a hump added to a smoothly varying background. This is strongly suggested by Fig. 6. Therefore, the peak feature around H_2 can be interpreted as an increase in the real part of the ac susceptibility, i.e., a decrease in pinning, due to the second superconducting transition. This stands in contrast to the usual peak effect such as P_3 we observe since the usual peak effect is due to an increase in pinning and is observed as a dip in the real part of the ac susceptibility.

Let us here make a remark on the nature of possible phase boundaries in the H-T plane. As the feature P_2 seems to mostly occur below the second superconducting transition field,[32] the feature P_2 should be mediated by fluctuation, suggestive of the second superconducting transition being second order. Taking into consideration the existence of the bicritical point around $H = 1.2$ T, $T = 0.8$ K, the H_{c2} branch above the second superconducting transition line possibly represents a first-order transition.

Another significant difference amongst various probes lies

in the size of the misalignment that suppresses the signs of the second superconducting transition. The minimum of such a misalignment is $\Delta\theta = 0.5°$ for the ac susceptibility and 3° for the thermal conductivity.[18] This is very likely to be due to the difference in probe. For example, the ac susceptibility measurements involve vortex motion and thus should be considerably more complicated than the thermal conductivity measurements. (When the out-of-plane component of the field becomes large enough for staircase vortices to form, the ac susceptibility will be largely affected. This roughly corresponds to $\Delta\theta = 0.5°$.[26]) It is not straightforward to discuss from existing experimental information how much misalignment of $\Delta\theta$ is large enough to suppress the second superconducting transition. If we closely relate the suppression of the low-temperature H_{c2} to the second superconducting transition, $\Delta\theta = 0.5°$ is found to be too small to suppress the second superconducting transition. Also the result of the fitting of Eq. (1) for the range $0° \leqslant \theta \leqslant 85°$ shown in Fig. 5 perhaps suggests that H_{c2} is suppressed over quite a wide range of $\Delta\theta$.

IV. SUMMARY

In summary, we have investigated the ac susceptibility of the spin-triplet superconductor Sr_2RuO_4 down to 60 mK, placing particular importance on multiple superconducting phases. Whilst the present results reproduce the previous results of the ac susceptibility[17] very well, there are several discrepancies between the present results and those of recent specific heat and thermal conductivity measurements.[9,17,18,24] The feature P_2 seen in the ac susceptibility is very likely to be closely related to the second superconducting transition; however, the previous attribution of P_2 to the second superconducting transition itself clearly disagrees with the recent studies of the thermal conductivity and the specific heat.[18,24] We propose that the feature previously attributed to the second superconducting transition[17] is a consequence of additional involvement of vortex pinning originated from the second superconducting transition. Vortex motion, prominent in the ac susceptibility, appears to be predominantly effective in overshadowing the features of the second superconducting transition.

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[21] The characteristic fields H_{c2}, H_2, and H_p have been defined using the real part of the ac susceptibility. These field positions do not always exactly coincide with the peaks in the imaginary part, but show reasonable agreement.

[22] Throughout the paper, we do not take into accout the demagnetization factor. The applied field almost fully penetrates at H_{c2} whilst we will later discuss the dependence of H_{c2} on the angle between the ab plane and the applied field. When we discuss the characteristic fields H_{c2}, H_2, and H_p, all of which are close to H_{c2}, the field is parallel to the platelike sample. The demagnetization effect is the least important in this field configuration.

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