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Interface superconductivity in the eutectic Sr_2RuO_4-Ru: 3-K phase of Sr_2RuO_4

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Enhanced superconductivity in the eutectic system Sr_2RuO_4-Ru is referred to as the 3-K phase of the spin-triplet superconductor Sr_2RuO_4 because of its enhanced superconducting transition temperature T_c of $\sim$3 K. We have investigated the field-temperature (H-T) phase diagram of the 3-K phase for fields parallel and perpendicular to the *ab* plane of Sr_2RuO_4, using out-of-plane resistivity measurements. We have found an upturn curvature in the $H_{c2}(T)$ curve for $H \| c$, and a rather gradual temperature dependence of H_{c2} close to T_c for both $H \| ab$ and $H \| c$. We have also investigated the dependence of H_{c2} on the angle between the field and the *ab* plane at several temperatures. Fitting the Ginzburg-Landau effective-mass model apparently fails to reproduce the angle dependence, particularly near $H \| c$ and at low temperatures. We propose that all of these characteristic features can be explained, at least in a qualitative fashion, on the basis of a theory by Sigrist and Monien that assumes surface superconductivity with a two-component order parameter occurring at the interface between Sr_2RuO_4 and Ru inclusions. This provides evidence of the chiral state postulated for the 1.5-K phase by several experiments.

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I. INTRODUCTION

Sr_2RuO_4 is the first layered perovskite superconductor without copper;[1] it is isostructural to the cuprate high-temperature superconductor $La_{2-x}Ba_xCuO_4$. This is one of the reasons that Sr_2RuO_4 has attracted great attention[2] despite its superconducting transition temperature T_c being rather low (ideally 1.5 K).[3,4] In fact, a number of theoretical and experimental studies have revealed its unconventional nature. More importantly, it is now well established that Sr_2RuO_4 is a spin-triplet superconductor, in contrast to the spin-singlet *d*-wave pairing in high-T_c cuprates.[2] This was first confirmed by ^{17}O-NMR measurements.[5] The Knight shift is unaffected by the superconducting transition, strongly suggestive of spin-triplet pairing with the spin of Cooper pairs lying within the *ab* plane.[5] Also, the observation of spontaneous magnetic moments accompanying the superconducting state indicates broken time-reversal symmetry,[6] suggesting a two-component order parameter with a relative phase of $\pi/2$. Subsequent experiments of Ru-NMR (Ref. 7) and polarized neutron scattering,[8] both of which measure the Knight shift, support the ^{17}O-NMR measurements.[5] A detailed small-angle neutron scattering study has revealed the vortex field distribution, which cannot be understood without the two-component order parameter.[9] These results constrain the basic form of the vector order parameter to be $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0(k_x + ik_y)$, which is called a chiral state.

Since the Fermi surface consists of three cylindrical (quasi-two-dimensional) sheets,[10,11] the above vector order parameter leads to an isotropic gap $[\Delta(\boldsymbol{k}) = \Delta_0\sqrt{k_x^2+k_y^2}]$. However, a number of experimental results[12–18] have revealed the power-law temperature dependence of various thermodynamic quantities and thus strongly suggest lines of nodes in the superconducting gap. This fact postulates modifications to be made to the basic form $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0(k_x + ik_y)$. In fact, several theories have been proposed to reconcile the discrepancy between those experimental facts and the vector order parameter $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0(k_x + ik_y)$.[19,20] These theories take into account the orbital dependent superconductivity[21] and propose very strong in-plane anisotropy[20] or horizontal lines of nodes[19] in the superconducting gap to explain the power-law temperature dependence in thermodynamic data.

Amongst several remarkable features related to Sr_2RuO_4, an enhancement of T_c in the Sr_2RuO_4-Ru eutectic system, is rather surprising. This enhancement was found during the course of the optimization of sample growth. While the ideal T_c of Sr_2RuO_4 turned out to be 1.5 K,[3,4] the ac susceptibility of certain batches was known to exhibit rather weak diamagnetism at a considerably higher onset temperature of about 3 K. A clear resistance drop, below which the resistance does not necessarily fall to zero, was also observed at a very close

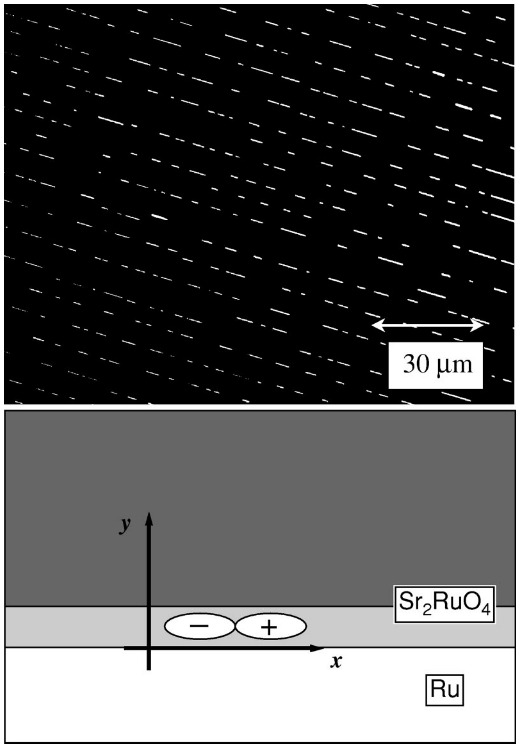

FIG. 1. Top: Optical microscopy picture of a polished surface parallel to the RuO_2 plane (bright region: Ru, dark region: Sr_2RuO_4). Bottom: Schematic of the interface between Sr_2RuO_4 and a Ru inclusion modeled in Sigrist and Monien's theory. The interface within the Sr_2RuO_4 part has a thin layer where a *p*-wave state nucleates at an enhanced transition temperature of ~3 K; its wave function has lobes and nodes parallel and perpendicular to the interface, respectively.

temperature. As discussed in Ref. 22, Maeno *et al.* established that these observations are indeed due to superconductivity and, as a result of careful investigations into the material origin, that it reproducibly occurs in the Sr_2RuO_4-Ru eutectic.

The eutectic system, a two-phase composite structure of a single-crystalline Sr_2RuO_4 matrix and lamellar microdomains of ruthenium metal embedded in it,[22] is obtained by the same method as Sr_2RuO_4 but with excess of Ru and/or at a faster growth speed.[23] The top panel of Fig. 1 shows an optical microscopy picture of a polished surface parallel to the RuO_2 plane. Typical dimensions of lamellae are 1 μm in thickness and 1–30 μm in length and width; the separation between adjacent lamellae is of the order of 10 μm. Although the appearance of the Ru inclusions may depend on growth conditions, the density of Ru inclusions should be uniquely determined by the composition at the eutectic point of Sr_2RuO_4 and Ru. In the top panel of Fig. 1, lamellae apparently line up along a certain direction, but empirically, there is no particular preferred orientation relative to the crystallographical axes. The direction often varies even within a small piece of single crystal of Sr_2RuO_4.

Such a eutectic system shows a broad superconducting transition with an onset of about 3 K. On further cooling, this transition is followed by the original superconducting transition of Sr_2RuO_4 at 1.5 K. The higher-T_c superconductivity is called the 3-K phase and the original lower-T_c superconductivity is referred to as the 1.5-K phase. The manifestations of 3-K phase superconductivity in resistance and ac susceptibility suggest that the superconductivity is inhomogeneous and filamentary. In addition, 3-K phase superconductivity is considered to be essentially sustained in Sr_2RuO_4. Because the upper critical field of the 3-K phase is the highest (lowest) when the applied field is parallel (perpendicular) to the *ab* plane of Sr_2RuO_4.[22] (That is, the anisotropy of the upper critical field reflects the crystallographical directions of Sr_2RuO_4.)

In addition to the enhancement of T_c to ~3 K, the field-temperature phase diagram of the 3-K phase has intriguing properties. Earlier work of resistive measurements[24] has revealed clear hysteresis of the upper critical field H_{c2} in magnetic fields parallel to the *ab* plane at low temperatures: Two distinctly different H_{c2}'s are obtained when the applied magnetic field (or the temperature) is swept upwards and downwards. Also the $H_{c2}(T)$ curve for $H\|c$ looks rather unusual, being nearly a straight line or possibly concave upwards.[22,24]

Neither theoretically nor experimentally has very much been known about the 3-K phase thus far. However, recent tunneling measurements on *c*-axis junctions of the Sr_2RuO_4-Ru eutectic have observed zero bias conductance peaks,[25] which is a hallmark of unconventional superconductivity.[26] Therefore, the superconductivity in the 3-K phase is also unconventional and probably originates from the triplet pairing of Sr_2RuO_4. Also Sigrist and Monien (SM)[27] have recently proposed a phenomenological theory that assumes surface spin-triplet superconductivity at the Sr_2RuO_4-Ru interface although the theory does not consider the mechanism of the enhanced superconductivity.

In the present work, we have studied the field-temperature phase diagram to higher precision than the previous work in Ref. 24 for a further discussion. We have also investigated the dependence of the upper critical field on the angle between the applied field and the *ab* plane. We will discuss these results with the help of SM's theory.[27] In addition, we have measured the specific heat to obtain thermodynamic evidence for nonbulk superconductivity, which supports an assumption of their theory.

II. EXPERIMENT

The eutectic samples of Sr_2RuO_4-Ru used in this study were grown by a floating zone method. The Ru inclusions were lamellate with typical dimensions of 1 μm×10 μm ×30 μm. A surface parallel to the *ab* plane of the sample used for resistive measurements is shown in the top panel of Fig. 1. There were no particular directions along which lamellae preferably line up throughout the whole sample. Details of the crystal growth are described in Ref. 22. We have measured the resistivity as a function of magnetic field or temperature to determine $H_{c2}(T)$ and $T_c(H)$. The dimensions of the sample, cut from a crystalline rod, were 0.96 ×1.04 mm^2 in the *ab* plane and 0.58 mm along the *c* axis. We employed a lock-in technique at 137 Hz with a current of 0.5 mA along the *c* axis. Low temperatures down to 60 mK

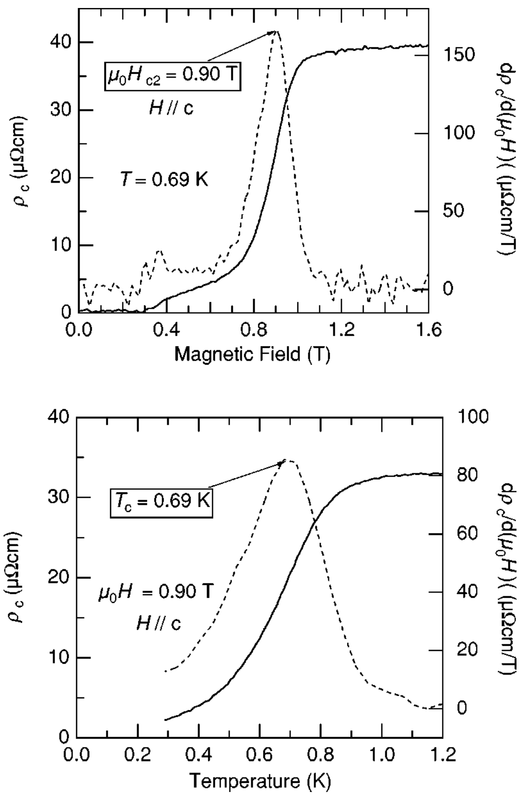

FIG. 2. Typical traces of the resistance of the 3-K phase (solid lines) and their derivatives with respect to magnetic field or temperature (dashed lines). These illustrate that the transition points (inflection points) from field sweep and temperature sweep show a good agreement.

were reached by means of a ^{3}He cryostat or a dilution refrigerator. Magnetic fields of up to 5 T were generated by a superconducting solenoid. The sample was mounted in a rotator that enabled the angle between the *ab* plane and the applied magnetic field to be changed. We have also measured the specific heat by a relaxation method (Quantum Design, model PPMS) down to 0.4 K. The sample for the specific heat measurement was cut from the same crystalline rod as used for the resistive measurements, and weighed at about 11 mg.

III. RESULTS AND DISCUSSION

A. *H-T* phase diagram

Figure 2 shows typical traces of the resistance as a function of magnetic field or temperature. The transition point has been defined as the inflection point associated with the superconducting transition to the 3-K phase. Figure 2 also demonstrates that the transition points determined from $H_{c2}(T)$ and $T_c(H)$ show a good agreement.

Figure 3 shows the resultant field-temperature (H-T) phase diagram of the 3-K phase of Sr_2RuO_4 for fields parallel to the *ab* plane and the *c* axis. This phase diagram contains a considerably larger number of transition points than those in Ref. 24, which makes possible a more detailed discussion. We, however, note that both phase diagrams appear to be very similar and consistent.

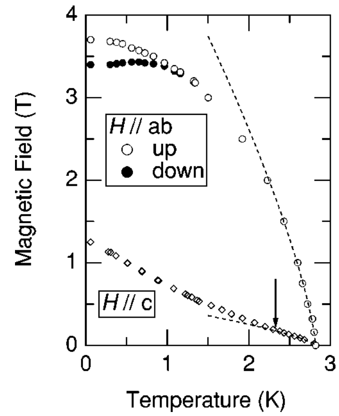

FIG. 3. Field-temperature phase diagram of the 3-K phase. The transition points have been determined as the inflection point associated with the superconducting transition to the 3-K phase. The H_{c2} curve for $H\|c$ shows an upward curvature with an inflection of 2.32 K, as indicated by an arrow. The dashed curves represent fits of $(1-T/T_c)^n$ dependence to data close to T_c. ($n=0.75$ and 0.72 for $H\|ab$ and $H\|c$, respectively.)

As seen also in the phase diagram of Ref. 24, there are two branches, corresponding to up and down sweeps, below $\sim$1.2 K for $H\|ab$. This is a consequence of the hysteresis of H_{c2} mentioned in the Introduction. As discussed in Ref. 24, two possibilities may be envisaged for the hysteresis. One is that the magnetic field effectively applied to the region responsible for the 3-K phase superconductivity is hysteretic. The second one is that the hysteresis of H_{c2} is instrinsic (i.e., due to a first order transition). Obviously, the latter case is even more interesting as the superconducting transition (type II) in magnetic fields is normally second order.[28] Possible interpretations for the latter case will include a first order transition due to spin depairing.[29] However, we point out that this is irrelevant to the spin-triplet state we suggest (chiral state).

The present study has revealed that the lower branch (down sweep of field or temperature) of the $H_{c2}(T)$ curve for $H\|ab$ is nearly flat, which seems to be rather unusual. This finding may contribute to a further understanding of the hysteretic behavior of H_{c2}. In addition to the hysteresis, we note two prominent features confirmed only in the H-T phase diagram obtained in the present study. (1) The temperature dependence of H_{c2} in the vicinity of T_c is rather gradual. (2) An upward curvature is seen below an inflection point of 2.32 K in the $H_{c2}(T)$ line for $H\|c$.

We will below propose that these two features may be explained, at least in a qualitative fashion, by SM's recent theory.[27] As stated in their original paper, they do not intend to consider the mechanism of the enhancement of T_c in the eutectic system, but they have constructed a phenomenological theory.

The theory includes the following reasonable assumptions. First, 3-K superconductivity occurs at interfaces between Sr_2RuO_4 and Ru inclusions. (For simplicity, they treat the interface as a single flat plane, as depicted in the lower panel of Fig. 1.) Second, the superconducting order parameter is represented by a two-component order parameter with

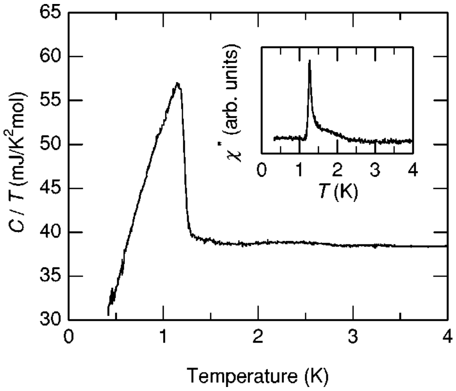

FIG. 4. Specific heat divided by temperature C_p/T of the eutectic system Sr_2RuO_4-Ru. While a clear peak associated with the 1.5-K superconducting transition is seen, a signature of 3-K superconductivity is barely evidenced. Inset: imaginary part of the ac susceptibility. A broad feature associated with the superconducting transition to the 3-K phase is seen.

a relative phase of $\pi/2$, similar to Sr_2RuO_4. They used a Ginzburg-Landau (GL) free energy for tetragonal symmetry[30] with the two-component order parameter $\eta = \eta_x + i\eta_y$, corresponding to $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}\Delta_0(\eta_x k_x + i\eta_y k_y)$.[27] The GL free energy also involves a δ-function potential enhancing the T_c at the interface between Sr_2RuO_4 and Ru.

The above assumptions receive support from existing experimental results such as a weak diamagnetism in the ac susceptibility, an imperfect resistance drop mentioned in the Introduction and the observation of zero bias conductance peaks.[25] We have also measured the specific heat for the 3-K phase in the present study. Figure 4 shows the specific heat divided by temperature. A sharp peak is seen at about 1.2 K, which is attributed to the original superconducting transition in Sr_2RuO_4. However, a signature of the transition to the 3-K phase is barely observed in the specific heat. This thermodynamically supports the first assumption above. In contrast, the imaginary part of the ac susceptibility, displayed in the inset to Fig. 2, shows a broad transition to the 3-K phase well above the sharp 1.5-K original transition. It should be noted that a small hump is seen in the specific heat between 2 and 3 K, which is very close to the transition temperature of the 3-K phase. The attribution of this small hump to the superconducting transition of the 3-K phase leads to its volume fraction being estimated to be $\sim 1.5\%$.[31]

Based on the above formulation, SM have considered the upper critical field in fields within the flat interface depicted in the bottom panel of Fig. 1. In both cases of $H \| ab$ and $H \| c$, H_{c2} is proportional to $(1-T/T_c)^{0.5}$ in the vicinity of T_c, which is common to surface superconductivity in a field applied parallel to the surface.[32] Examples include superconductivity at twin boundaries.[32]

Fitting the functional form $H_{c2}(T) = A(1-T/T_c)^n$ to the gradual temperature dependence of H_{c2} in the vicinity of T_c yields $n = 0.75$ and $n = 0.72$ for $H \| ab$ and for $H \| c$, respectively, where A and n are the adjustable parameters.[33] These exponents have been obtained from fitting the data between T_c and approximately $0.9T_c$. As all of these exponents are in contrast to the standard $(1-T/T_c)$ dependence, we suggest that it supports surface superconductivity in the 3-K phase.

On the other hand, fitting $(1-T/T_c)^n$ dependence to the H-T phase diagram of the 1.5-K phase based on specific heat measurements[34,35] yields $n = 0.90$ and $n = 1.0$, for $H \| ab$ and $H \| c$, respectively.[36] Also a phase diagram from resistive measurements on the 1.5-K phase, albeit the number of data points are rather few, the temperature dependence appears to be linear close to T_c.[37] While the exponents of about 0.7 obtained for the 3-K phase somewhat deviate from the predicted value $n = 0.5$, those values are considerably smaller than $n = 1$.

Although we claim that the H-T phase diagram obtained probably supports surface superconductivity, a possible criticism is that the exponents obtained being around 0.7 is *not* in good enough agreement with the theoretical value of 0.5. This discrepancy should originate from the above discussion along the line of SM's theory[27] being somewhat crude for comparison with experiment. Matsumoto and Sigrist[38] have very recently improved the calculations in SM's paper[27] and have obtained an exponent of about 0.7 for the temperature range used for our fitting; this exponent is very close to our results.

The formalism and assumptions Matsumoto and Sigrist have used are in principle based on those of SM's theory. Their important improvement is that Matsumoto and Sigrist use a more realistic wave function of the order parameter in magnetic fields than SM's calculations.[38] Whereas SM used an exponentially decaying wave function as in Ref. 32, Matsumoto and Sigrist have pointed out that this functional form is appropriate only when the field is very low. In fact, Matsumoto and Sigrist's numerical results indicate that the exponent tends to 0.5 with approaching $T = T_c$ or $H = 0$. Matsumoto and Sigirst have taken into consideration the harmonic potential due to the applied magnetic field, leading to a contraction of the wave function. They have obtained higher T_c's and consequently exponents of around 0.65 and around 0.75 for $H \| ab$ and for $H \| c$, respectively.[38]

While we intend to discuss the exponent in the vicinity of H_{c2}, the range of temperature over which the fit has been applied inevitably has a finite width. Our exponents quoted above are from a temperature range of approximately $0.9T_c < T < T_c$. The exponent from fitting seems not to significantly depend on the temperature range over which the fitting was done, unless the lower temperature limiting the fitting range is too low[39] or the number of the data used for the fit are too few.[40]

SM's theory also provides a qualitative explanation for the anomalous behavior of the $H_{c2}(T)$ curve for $H \| c$ (Ref. 27) (i.e., upward curvature at low temperatures and high fields). They predict that only one of the two components of a superconducting order parameter, such as k_x or k_y is stabilized at T_c in zero applied field and that the other component with a relative phase of $\pi/2$ arises at a slightly lower temperature.[41] However, the application of a magnetic field *not* perpendicular to the c axis will induce simultaneously the two components with a relative phase of $\pi/2$ at T_c. [Since the triplet state represented by the order parameter $k_x + i\varepsilon k_y (0 < \varepsilon \leqslant 1)$ has an orbital magnetic moment along

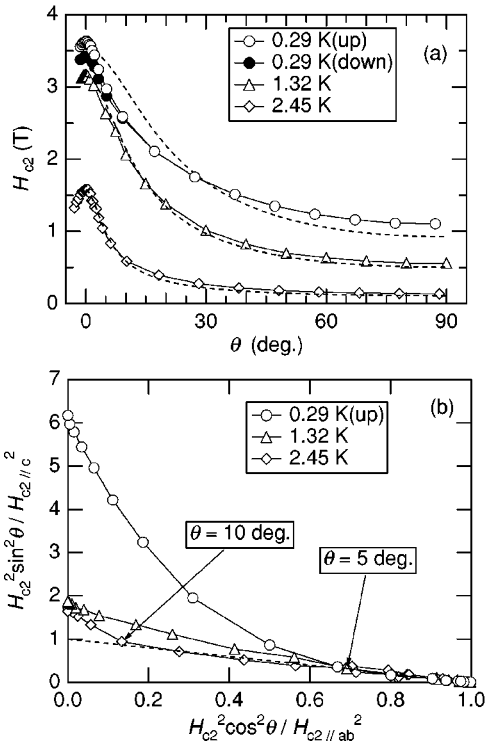

FIG. 5. (a) Angle θ dependence of the upper critical field H_{c2} at 0.29, 1.32, and 2.45 K. The dashed curves represent fits of the GL effective mass model for $0° \leq \theta \leq 90°$. For 0.29 K, there are two branches reflecting hysteresis of H_{c2}; the open (solid) circles correspond to up (down) sweep of field. (b) The same data (but without down-sweep branch at 0.29 K) and fits of the GL effective mass model for $0° \leq \theta \leq 5°$ (for 0.29 and 1.32 K) and $0° \leq \theta \leq 10°$ (for 2.45 K). The data are plotted as $(H_{c2}\cos\theta/H_{c2\|ab})^2$ vs $(H_{c2}\sin\theta/H_{c2\|c})^2$, so that all of the fits are represented by the dashed straight line.

the c axis, the state is energetically stabilized by a finite magnetic-field component parallel to the c axis.] As a consequence of both components being stabilized, the coupling between the two components results in an enhancement of H_{c2}. In addition, the coupling becomes stronger at lower temperatures, leading to an upward curvature in the $H_{c2}(T)$ curve for $H\|c$.

In addition to the mechanism described in the last paragraph, Matsumoto and Sigrist suggest that there is another mechanism for the low-temperature high-field enhancement of H_{c2} for $H\|c$.[38] They have recently raised that the region of the enhanced superconductivity (3-K phase) has a finite width in an actual eutectic system although SM adopted the GL free energy with a δ-function potential enhancing the T_c at the interface. At sufficiently high fields, the spacial extension of the wave function of the order parameter will be confined within the region where the enhanced superconductivity nucleates, leading to an additional enhancement of H_{c2}.

B. Angle dependence of the upper critical field

Figure 5(a) shows the angle θ dependence of the upper critical field H_{c2} at 0.29, 1.32, and 2.45 K. (θ is the angle between the ab plane and the c axis; $\theta=0$ corresponds to $H\|ab$.) Only at 0.29 K of these three temperatures, does H_{c2} show hysteresis close to $H\|ab$. For 0.29 K, the hysteresis of H_{c2} persists to $|\theta| \approx 10°$. [For 60 mK, the hysteresis is observed for $|\theta| \lesssim 20°$. The angle range for which the hysteresis can be seen decreases with increasing temperature. As mentioned in Sec. III A, the hysteresis of H_{c2} disappears at $\sim$1.2 K even for $\theta \approx 0°$ ($H\|ab$).] While the lower branch (down sweep of field) for 0.29 K is plotted with solid circles in Fig. 5(a), the upper branch (up sweep of field) is used for the fitting below in this subsection. This is because whether the up sweep or down sweep is used hardly affects the discussion below. Also shown in Fig. 5(a) are fits of the GL effective mass model

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

Here Γ is the square root of the ratio of the effective mass for interplane motion to that for in-plane motion (i.e. $\Gamma = H_{c2\|ab}/H_{c2\|c}$).[42] We have taken $H_{c2\|ab}$ and $H_{c2\|c}$ to be the adjustable parameters for the fitting. The resultant values of ($H_{c2\|ab}$, $H_{c2\|c}$) shown in Fig. 5(a) are (3.52 T, 0.92 T), (3.14 T, 0.50 T), and (1.57 T, 0.11 T) for 0.29 K (up sweep), 1.32 K and 2.45 K, respectively. [Those for 0.29 K (down sweep) is (3.33, 0.97 T); the curve is not shown.]

Although this model is known to best fit for temperatures close to T_c, it reproduces as a whole the θ dependence of H_{c2} for the 1.5-K phase (pure Sr_2RuO_4) even at 60 mK.[43] It should be noted here that a region close to $H\|ab$ (e.g., $\Delta\theta \lesssim 5°$) for the 1.5-K phase is exceptional due to the unusual suppression of the upper critical field; this is probably related to (or caused by) a double superconducting transition.[34,43] In contrast, Fig. 5(a) exhibits that the model apparently fails to reproduce experimental results of the 3-K phase in a very wide angle range. This discrepancy between the data and the model is particularly evident for low-temperature data.

SM's theory[27] can be extended to the case of the applied field pointing to arbitrary directions within the Sr_2RuO_4-Ru interface plane.[27,44] A discussion with a minor simplification yields an analytic functional form identical to the GL effective mass model [Eq. (1)].[42] However, this analytic expression is valid for the present system only when the temperature T is close to T_c and/or the coupling of the two components is small (i.e., H is nearly parallel to the ab plane). In fact, in the framework of the GL formalism, SM resorted numerical means to investigate the behavior of $H_{c2}(T)$ for $H\|c$ at low temperatures.

Consequently, fitting Eq. (1) to data for a certain angle range close to $H\|ab$ will show a reasonable agreement. In this context, comparison of the data with the effective-mass model will reveal how the discrepancy becomes evident and thus will enable the enhancement of H_{c2} due to the coupling of the two components to be discussed. As Fig. 5(a) indicates that fitting Eq. (1) to the whole data for $0° \leq \theta \leq 90°$ does not yield satisfactory results, we have fitted Eq. (1) to the data for $0° \leq \theta \leq 5°$ (at 0.29 and 1.32 K) and $0° \leq \theta \leq 10°$ (at 2.45 K), yielding ($H_{c2\|ab}$, $H_{c2\|c}$) of (3.62 T, 0.45 T), (3.16 T, 0.41

T), and (1.58 T, 0.11 T) for 0.29 K (up sweep), 1.32 K and 2.45 K, respectively. [Those for 0.29 K (down sweep) are (3.41 T, 0.46 T).]

In Fig. 5(b), the same data as in Fig. 5(a) are plotted as $[H_{c2}(\theta)\cos\theta/H_{c2\parallel ab}]^2$ vs $[H_{c2}(\theta)\sin\theta/H_{c2\parallel c}]^2$; the results of the fitting described in the last paragraph are used for $H_{c2\parallel ab}$ and $H_{c2\parallel c}$ at each temperature. This plot allows one to see the deviation from the effective mass model more clearly. Since Eq. (1) may be rewritten as

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

the functional form of Eq. (1) is represented by a straight line connecting (0,1) and (1,0) in this plot. Figure 5(b) indeed illustrates that Eq. (1) fits well the data at each temperature for a limited angle range close to $H\parallel ab$. The data start to deviate from the functional form of Eq. (1) [i.e., the dashed straight line in Fig. 5(b)] at about $\theta=5°$ (for 0.29 and 1.32 K) and $\theta=10°$ (for 2.45 K). (Note that the angle θ at which the deviation becomes evident is irrespective of the choice of values for $H_{c2\parallel ab}$ or $H_{c2\parallel c}$.)

The deviation is obviously large at low temperatures and large angles (close to $H\parallel c$). In other words, H_{c2} is enhanced at low temperatures and large angles. Similarly, $H_{c2\parallel c}$ from the fitting for the whole data ($0°\leqslant\theta\leqslant 90°$) is larger than that from the limited range ($0°\leqslant\theta\leqslant 5°$ or $10°$). This tendency is in very good agreement with SM's theory.[27] They suggest that the coupling between the two components of the order parameter, which enhances H_{c2}, becomes stronger with decreasing temperature and increasing magnetic field component parallel to the c axis.

Before finishing this subsection, we here make a remark on the angle dependence of H_{c2} from another viewpoint. The deviation from the GL effective model Eq. (1) becomes larger with decreasing temperature and $H_{c2}(\theta)$ becomes peaked in the vicinity of $H\parallel ab$ at low temperatures. The latter behavior is somewhat reminiscent of the two-dimensional thin film model[45]

$$\left(\frac{H_{c2}(\theta)\cos\theta}{H_{c2\parallel ab}}\right)^2+\frac{H_{c2}(\theta)|\sin\theta|}{H_{c2\parallel c}}=1. \tag{3}$$

In fact, Eq. (3) shows a peaked feature close to $H\parallel ab$ while Eq. (2) does not. The thin film model[45] assumes $d\ll\xi_{ab}$ and leads to $H_{c2\parallel c}=\Phi_0/2\pi\xi_{ab}^2$ and $H_{c2\parallel ab}=\sqrt{3}\Phi_0/\pi d\xi_{ab}$, where ξ_{ab} is the coherence length parallel to the *ab* plane, d is the layer spacing, and $\Phi_0=2.07\times10^{-15}$ T/m^2 is the fluxoid. Nevertheless, the use of these formulas for the 3-K phase at 60 mK results in $\xi_{ab}=16.2$ nm and $d=1.90$ nm, which does not satisfy a prerequisite of the model $d\ll\xi_{ab}$. Also, $d=1.90$ nm is substantially larger than the layer spacing of Sr_2RuO_4, 0.637 nm. These facts suggest the application of the thin film model to the 3-K phase is inappropriate.

IV. CONCLUSION

In summary, we have investigated the field-temperature phase diagram of the 3-K phase of Sr_2RuO_4 in detail using resistivity measurements. We have found a rather gradual temperature dependence of the upper critical field H_{c2} close to T_c and an enhancement of H_{c2} for $H\parallel c$ at low temperatures. We have also investigated the dependence of H_{c2} on the angle between the field and the *ab* plane at several temperatures. Fitting of the GL effective-mass model apparently fails to reproduce the angle dependence. All of these experimental results, with the help of the theory of SM may be interpreted in a consistent manner with other existing experimental facts. Taken together with the phenomenological theory by SM, these observations support that the 3-K phase is surface spin-triplet superconductivity with a two-component order parameter occurring at Sr_2RuO_4-Ru interfaces. This, although indirectly, supports the basic form of $\boldsymbol{d}(\boldsymbol{k})=\mathbf{z}\Delta_0(k_x+ik_y)$ for bulk Sr_2RuO_4 as well.

Note added in proof. Recently, Matsumoto *et al.* have written a paper[46] which includes part of the private communication with Matsumoto and Sigrist in Ref. 38.

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