First-Order Superconducting Transition of Sr_2RuO_4

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By means of the magnetocaloric effect, we examine the nature of the superconducting-normal (S-N) transition of Sr_2RuO_4, a most promising candidate for a spin-triplet superconductor. We provide thermodynamic evidence that the S-N transition of this oxide is of first order below approximately 0.8 K and only for magnetic field directions very close to the conducting plane, in clear contrast to the ordinary type-II superconductors exhibiting second-order S-N transitions. The entropy release across the transition at 0.2 K is 10% of the normal-state entropy. Our result urges an introduction of a new mechanism to break superconductivity by magnetic field.

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The order of a phase transition provides one of the most fundamental pieces of information of the long-range ordered state accompanied by the phase transition. In the case of superconductivity, the order of the superconducting-normal (S-N) transition in magnetic fields reflects how the superconductivity interacts with the magnetic field and how it is destabilized. For example, for a type I superconductor, the in-field S-N transition is a first-order transition (FOT) [1], because of an abrupt disappearance of the superconducting (SC) order parameter caused by the excess energy for magnetic-flux exclusion. For a type II superconductor, in contrast, the in-field S-N transition is ordinarily a second-order transition (SOT) [1]. In this case, penetration of quantized vortices with accompanying kinetic energy due to orbital currents leads to a continuous suppression of the SC order parameter up to the upper critical field H_{c2}. This type of pair breaking is called the orbital effect.

A well-known exception for type II superconductivity is the case where the superconductivity is destroyed by the Zeeman spin splitting [2]. When the spin susceptibility in the SC state, χ_{sc}, is lower than that in the normal state, χ_n, the SC state acquires higher energy $\Delta E_Z \sim (1/2)(\chi_n - \chi_{sc})\mu_0 H^2$ with respect to the normal state, due to the difference of polarizability of the electron spin. This destroys superconductivity at the Pauli limiting field $\mu_0 H_P \sim [2\mu_0 E_{cond}/(\chi_n - \chi_{sc})]^{1/2}$, where ΔE_Z reaches the SC condensation energy E_{cond}. Such a pair-breaking effect is called the Pauli effect. It is theoretically predicted that a strong Pauli effect leads to a first-order S-N transition at temperatures sufficiently lower than the critical temperature T_c [3]. This prediction has been confirmed in a few spin-singlet superconductors [4–6].

The type II superconductor Sr_2RuO_4 ($T_c = 1.5$ K) is one of the most promising candidates for spin-triplet superconductors [7–9]. Due to its unconventional superconducting phenomena originating from the orbital and spin degrees of freedom as well as from the nontrivial topological aspect of the SC wave function, this oxide continues to attract substantial attention [10–14]. The spin-triplet state has been directly confirmed by extensive spin susceptibility measurements by means of the nuclear magnetic resonance (NMR) using several atomic sites [15–17] and the polarized neutron scattering [18]: Both experiments have revealed $\chi_{sc} = \chi_n$ in the entire temperature-field region investigated. This means that $H_P \propto (\chi_n - \chi_{sc})^{-1/2}$ is infinite and the Pauli effect is irrelevant in this material.

Interestingly, several properties of the S-N transition of Sr_2RuO_4 have not been understood for more than 10 years within the existing scenarios for the spin-triplet pairing. For example, $H_{c2}(T)$ is more suppressed than the expected behavior for the orbital effect, when the field is parallel to the conducting *ab* plane [19–21]. In addition, several quantities such as the specific heat C [20], thermal conductivity κ [20], magnetization M [22], exhibit sudden recovery to the normal-state values near H_{c2} for $H \parallel ab$ and at low temperatures.

To resolve the origin of such unusual behavior, we performed measurements of the magnetocaloric effect (MCE) of Sr_2RuO_4. The MCE is a change of the sample temperature T in response to a variation of the external magnetic field H; we measure T while sweeping H at a constant rate. The thermal equation of the MCE is written as [23]

$$\left(\frac{\partial S}{\partial H}\right)_T = -\frac{C}{T}\left(\frac{dT}{dH}\right) - \frac{k(T - T_{bath})}{T\dot{H}} - \frac{1}{T}\frac{d'Q_{loss}}{dH}, \quad (1)$$

where S is the entropy, C is the heat capacity of the sample, k is the thermal conductance between the sample and thermal bath, $\dot{H}$ is the sweep rate of the magnetic field, T_{bath} is the temperature of the thermal bath, and $d'Q_{loss}$ is the dissipative loss of the sample. When k is small so that the second term is negligible, the equation reduces to the relation for the conventional adiabatic MCE. In the other limit where the thermal coupling between the sample and bath is strong, the first term in turn becomes negligible, leading to the "strong-coupling limit" relation [23] $(\partial S/\partial H)_T \simeq -(k\Delta t/\dot{H}) - T^{-1}(d'Q_{loss}/dH)$ with $\Delta t \equiv (T - T_{bath})/T$ [24]. In this limit, the measured Δt is linearly dependent on $(\partial S/\partial H)_T$. Thus, it is expected that T and Δt exhibit peaklike anomalies at a FOT and steplike anomalies at a SOT. Because of this qualitative difference, the strong-coupling MCE is suitable to distinguish a FOT and a SOT. We found that our calorimeter indeed works almost in this strong-coupling limit, with the first term in Eq. (1) amounting to at most 10% of the second term. We however didn't neglect the first term in the evaluation of the entropy discussed below.

For the present study, we used single crystals of Sr_2RuO_4 grown by the floating-zone method [25]: Sample 1 weighing 0.684 mg with $T_c = 1.45$ K and sample 2 weighing 0.184 mg with $T_c = 1.50$ K. The value of T_c of sample #2 is equal to the ideal T_c of Sr_2RuO_4 in the clean limit [26], indicating its extreme cleanness. The MCE was measured using a hand-made sensitive calorimeter. Magnetic field was applied using a vector magnet system [27]. Details of the experimental method is described in the Supplemental Material [24].

We first present the MCE for $H \parallel ab$ $(H \sim [100])$ and $T \sim 0.2$ K measured at $\mu_0\dot{H} = \pm 1.02$ mT/sec in Figs. 1(a) and 1(b). Obviously, $T(H)$ exhibits peak-like behavior near H_{c2}, rather than a single steplike behavior. This feature becomes clearer in the background-subtracted

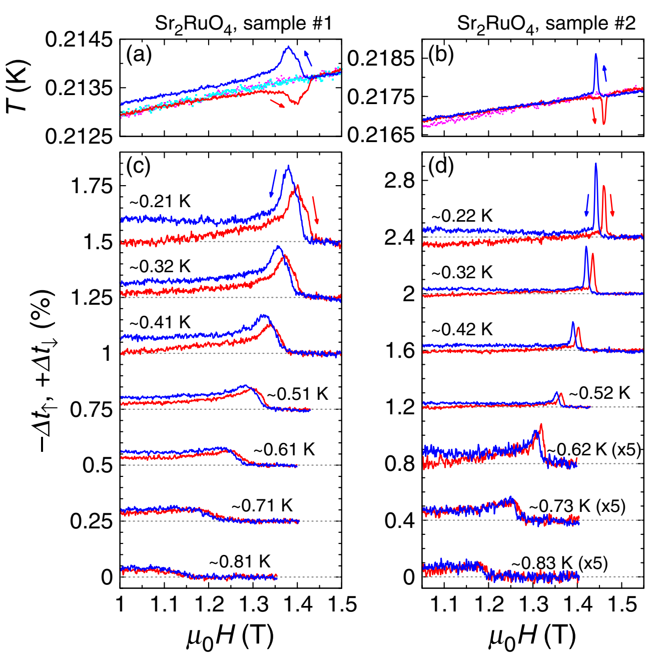

FIG. 1 (color online). (a), (b) Representative raw data of the MCE of Sr_2RuO_4 for $T \sim 0.2$ K at $H \parallel ab$ $(H \sim [100])$. The dotted curves indicate $\tilde{t}_{bg} T_0$ for the up sweep (pink or darker gray) and the down sweep (cyan or lighter gray), which corresponds to the background contribution [24]. (c), (d) Relative temperature change $-\Delta t_\uparrow$ (red or lighter gray) and $\Delta t_\downarrow$ (blue or darker gray) due to the MCE for the same field condition at different temperatures. For clarity, each pair of curves is shifted vertically by 0.25% and 0.4% for panels (c) and (d), respectively. The high-temperature ($T > 0.6$ K) data in (d) are multiplied by 5.

$\Delta t_\uparrow(H)$ (up-sweep) and $\Delta t_\downarrow(H)$ (down-sweep) curves shown in Figs. 1(c) and 1(d) [24]. The observed peak provides indication of a FOT in Sr_2RuO_4. Note that a slight asymmetry in the MCE signal (i.e., $|\Delta t_\uparrow(H)| < |\Delta t_\downarrow(H)|$) is attributed to the energy dissipation mainly due to vortex motion causing a heating in both the field up-sweep and down-sweep measurements [28]. More importantly, H_{c2} is clearly different between the up-sweep and down-sweep curves. The difference between the up-sweep onset $H_{c2\uparrow}$ and the down-sweep onset $H_{c2\downarrow}$ is approximately $\mu_0\Delta H_{c2} \equiv \mu_0(H_{c2\uparrow} - H_{c2\downarrow}) = 20$ mT for sample #1 and 15 mT for sample #2. This difference corresponds to 15–20 sec for $\mu_0\dot{H} = 1.02$ mT/sec. The difference cannot be attributed to an extrinsic delay of the temperature measurement, since the delay time of our apparatus is much shorter than 15–20 sec [29]. We have also confirmed that a finite ΔH_{c2} is observed for lower sweep rates such as $\mu_0\dot{H} = \pm 0.2$ mT/sec. Therefore, this difference of H_{c2} is indeed intrinsic, and provides definitive evidence that the S-N transition is a FOT accompanied by supercooling (or possibly superheating). Note that the very sharp peak in $\Delta t(H)$ at H_{c2} for sample #2 demonstrates the cleanness and homogeneity of this sample.

Next, we focus on the variation of the MCE with temperature and field angle. As represented in Figs. 1(c) and 1(d), both the peak in $\Delta t(H)$ and the supercooling becomes less pronounced as temperature increases. Around 0.8 K, these features totally disappear and the S-N transition becomes a SOT as expected for ordinary type II superconductors. In Fig. 2, we present several MCE curves for fields tilted away from the ab plane toward the c axis by the amount which we define as θ. When the field is tilted only by ~2 degrees, the FOT features disappear.

From the MCE data for $H \parallel ab$, we deduce the entropy using Eq. (1) [24]. Figure 3(a) again characterizes the FOT with a huge peak in $(\partial S/\partial H)_T$ and supercooling or

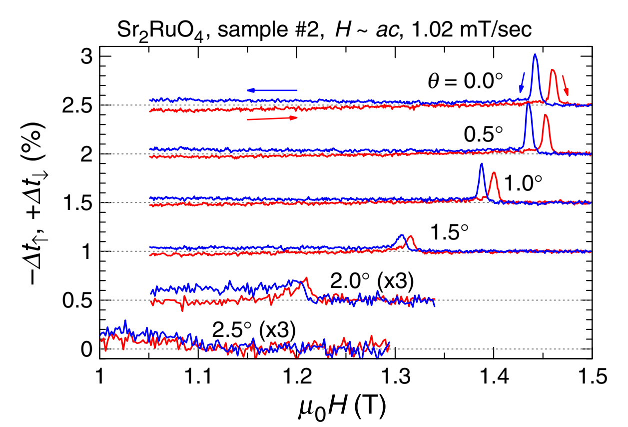

FIG. 2 (color online). Field angle θ variation of the MCE of sample 2 at $T \sim 0.2$ K. The red (lighter gray) and blue (darker gray) curves indicate $-\Delta t_\uparrow(H)$ and $\Delta t_\downarrow(H)$, respectively. Each curve is shifted vertically by 0.5% and the data for $\theta \geq 2.0°$ are multiplied by 3 for clarity.

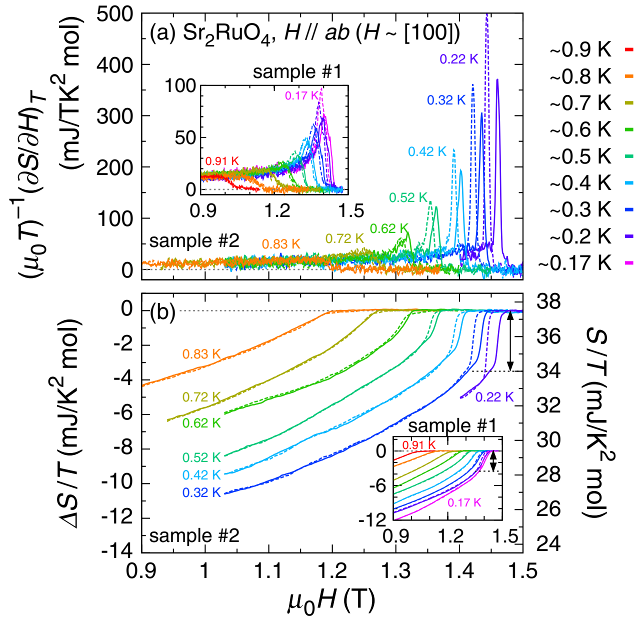

FIG. 3 (color online). (a) Field dependence of $(\mu_0 T)^{-1} \times (\partial S/\partial H)_T$ of Sr_2RuO_4 deduced from the MCE. (b) Field dependence of $\Delta S/T$. In both (a) and (b), the main panels present data for sample 2 and the insets for sample 1; the solid and broken curves present up- and down-sweep data, respectively. The right axis of (b) indicates S/T obtained by assuming $S_n/T = 37.5$ mJ/K^2 mol [30]. The double-headed arrow in (b) illustrates the jump $\delta S/T = -3.5 \pm 1$ mJ/K^2 mol at the transition.

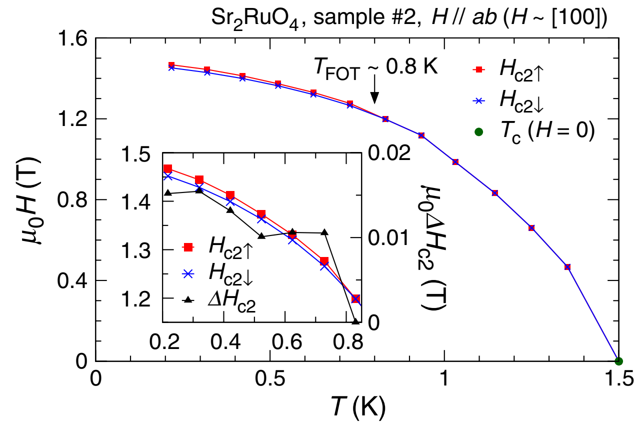

FIG. 4 (color online). Superconducting phase diagram of Sr_2RuO_4 for $H \parallel ab$ ($H \sim [100]$) deduced from the MCE for sample 2. The red squares and the blue crosses indicate the onset H_{c2} for the up- and down-sweeps, respectively. The inset presents H_{c2} and ΔH_{c2} (triangles) in the low-temperature region.

superheating. In Fig. 3(b), we present $\Delta S \equiv S - S_n = \int_{H_{c2}}^{H} (\partial S/\partial H)_T dH$ divided by temperature. Here, S_n is the entropy in the normal state. The total entropy S can be calculated with the assumption $S_n/T = \gamma_e$, where $\gamma_e = 37.5$ mJ/K^2 mol is the electronic specific heat coefficient [30]. The jump in S/T across the FOT is approximately $\delta S/T = -3.5 \pm 1$ mJ/K^2 mol at the lowest measured temperatures. This value of $\delta S/T$ amounts to approximately 10% of S_n/T, and the latent heat $L = T\delta S$ at 0.2 K is 0.14 ± 0.04 mJ/mol. We can check the consistency of this value using the Clausius-Clapeyron equation $\mu_0 dH_{c2}/dT = -\delta S/\delta M$, where δM is the jump in M across the FOT. Using the values $\mu_0 dH_{c2}/dT \sim -0.20 \pm 0.05$ T/K estimated from our $H_{c2\uparrow}$ data for sample #2 and $\delta M \sim -0.014$ emu/g from the magnetization study [31], we obtain $\delta S/T = -4.7 \pm 1.2$ mJ/K^2 mol for 0.2 K. This value reasonably agrees with the value from our MCE experiment. In addition, S at lower fields also exhibits agreement with other thermodynamic studies [20,22], as explained in the Supplemental Material [24].

We summarize the present observations in the phase diagrams presented in Figs. 4 and 5. The region for which the FOT emerges is limited to temperatures below $T_{\rm FOT} \sim 0.8$ K for $\theta = 0°$ and field angles within $|\theta| < 2°$ for $T \sim 0.2$ K. Interestingly, the FOT region is included in a wider region in which the behavior of H_{c2} cannot be described solely by the conventional orbital effect [21]: $H_{c2}(T)$ substantially deviates from the linear behavior and $H_{c2}(\theta)$ cannot be fitted with the effective mass model (Fig. 5). These facts indicate that the ordinary orbital effect cannot be an origin of the FOT.

Let us compare the present results with previous observations. The rapid recoveries of κ/T [20] and M [22] near H_{c2} for $H \parallel ab$ have been observed in the region where the S-N transition is revealed to be of first order. Thus, it now turns out that these recoveries are actually consequences of the FOT. However, supercooling (or superheating) at the S-N transition in Sr_2RuO_4 has never been reported in previous studies. This is probably because the supercooled metastable normal state easily nucleates into the SC state. Thus, a fast and continuous sweep is helpful to observe the supercooling, rather than point-by-point measurements.

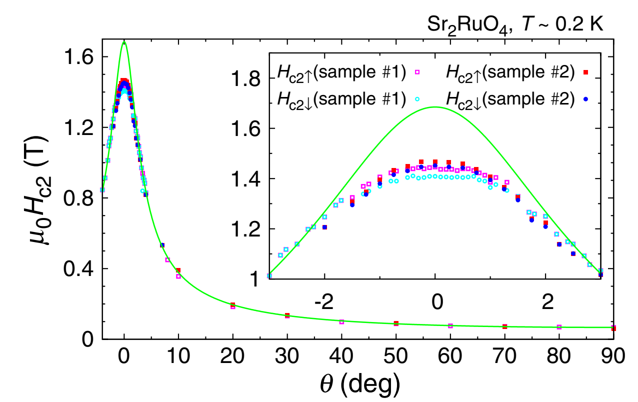

FIG. 5 (color online). Field-angle dependence of $H_{c2\uparrow}$ (squares) and $H_{c2\downarrow}$ (circles) at $T \sim 0.2$ K. The green curve indicates the fitting to the data of sample #1 in the range $|\theta| > 2.0°$ [21] with the effective mass model $H_{c2}(\theta) = H_{c2}(90°)/(\sin^2\theta + \cos^2\theta/\Gamma^2)^{1/2}$, where Γ is the anisotropy parameter and is obtained to be 25 from the fitting.

The smallness and cleanness of the present samples have also assisted the observation, because the number of nucleation centers (e.g., surface defects, lattice imperfections) is reduced for small and clean samples. In contrast to the previous studies on the bulk SC phase, a hysteresis in the in-field S-N transition was observed for the interfacial 3 K phase superconductivity in the Sr_2RuO_4-Ru eutectic [32,33]. Possible relation between this hysteresis and the present observation is worth examining. We note that the second transition revealed by the C/T measurement [20] at H_2, which is 20–30 mT below H_{c2}, cannot be attributed to the onset of the FOT. Although we have not so far obtained convincing MCE data supporting the H_2 anomaly, we need higher experimental resolution to clarify this issue.

In the rest of this Letter, we discuss the origin of the FOT. As we have explained, the FOT should originate from a pair-breaking effect beyond the conventional orbital effect. Naively, a possible candidate of such a pair-breaking effect is the Pauli effect [34]. However, in the case of Sr_2RuO_4, NMR and neutron studies have revealed $\chi_{sc} = \chi_n$ [15–18]. In particular, the observed negative hyperfine coupling provides strong evidence that NMR correctly detects the spin susceptibility [9]. In addition, $\chi_{sc} = \chi_n$ has been confirmed for different nuclei at several atomic sites. Other experiments also indirectly support the spin-triplet scenario [10–12,14]. Therefore, the Pauli effect should be absent in Sr_2RuO_4 and the FOT cannot be attributed to the Pauli effect either. Thus, an unknown pair-breaking effect, or in other words, a nontrivial interaction between superconductivity and magnetic field, must be taken into account.

For a spin-triplet superconductor with $\chi_{sc} = \chi_n$, it is naively expected that the $|\downarrow\downarrow\rangle$ condensate can be mutually converted to the $|\uparrow\uparrow\rangle$ condensate by magnetic field without destroying the SC state. However, in contrast to the expectation, the present experiment shows that the triplet SC state in Sr_2RuO_4 is no more stable at high fields where the Zeeman splitting is no longer a perturbation. Indeed, $\mu_0 H_{c2}$ for $T \to 0$ nearly matches the field $\mu_0 \tilde{H}_P = (2\mu_0 E_{cond}/\chi_{sc})^{1/2} \sim 1.4$ T, where *the Zeeman spin energy in the SC state* $(1/2)\chi_{sc}\mu_0 H^2$ is equal to E_{cond} [35]. This fact suggests that the Zeeman splitting between $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ condensates, which has not been considered in the existing theories on the SC phase diagram of Sr_2RuO_4 [34,36–39], is a source of the nontrivial coupling between magnetic field and triplet superconductivity.

Let us propose possible mechanisms of the nontrivial interaction. We can categorise them into microscopic and macro- or mesoscopic mechanisms. The microscopic mechanisms include the pinning of the electron spin direction at certain k points predicted by the band calculation [40] and confirmed by the angle-resolved photoemission spectroscopy [41]. Such a pinning of the spin direction may lead to a constraint in the spin polarization due to the Zeeman effect. The closeness of the Fermi energy to the van Hove singularity [42] is also worth considering, because a slight modification of the chemical potential due to the Zeeman effect might disturb the pairing glue.

The macro- or mesoscopic mechanisms include possible interactions among the Cooper-pair orbital angular momentum $\boldsymbol{L}$, the Cooper-pair spin $\boldsymbol{S}$, the vortex vorticity, and the magnetic field. Indeed, a pair-breaking effect due to $\boldsymbol{L}$ was proposed in Ref. [43], although this theory cannot be directly applied as long as the orbital motion is assumed to be purely two dimensional. As another macro- or mesoscopic mechanism, the kinematic polarization discussed in the context of the stability of the half-quantum vortex is instructive [14,44]. It was proposed that a velocity mismatch between $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ condensates around a half-quantum vortex results in a shift of the chemical potential of these two condensates due to difference in their kinetic energies and leads to an additional spin polarization coupling to the magnetic field. By an analogy to this theory, we expect that consideration of kinematics of the condensates in high fields may provide a route to unveil the nontrivial coupling between the Cooper pair and magnetic field.

In summary, our MCE study of Sr_2RuO_4 revealed definitive evidence for a first-order S-N transition in the low-temperature region for fields nearly parallel to the *ab* plane. The FOT, not attributable to conventional mechanisms, indicates a nontrivial interaction between spin-triplet superconductivity and magnetic field. This new information on the bulk superconductivity serves as a basis for investigations of the nontrivial topological nature of the SC wave function associated with the "Majorana-like" edge modes. We also anticipate that the abrupt growth of the order parameter across the FOT, accompanied by vortex formation and nontrivial symmetry breaking, should provide a new playground for investigation of novel vortex dynamics, which might be related to quantum turbulence and/or to the Kibble-Zurek mechanism.

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