Stabilization of superconductivity by metamagnetism in an easy-axis magnetic field on UTe₂

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Influence of metamagnetism on superconductivity is studied for the field along an easy *a*-axis by AC magnetic susceptibility, magnetization, and magnetocaloric effect (MCE) in UTe₂. The improvement of crystal quality leads to an upward curvature of $T_c(B)$ above 6 T, that results in a largely enhanced upper critical field of 12 T. The entropy analysis also shows a metamagnetic crossover around 6 T. The sharp negative peak in MCE below T_c indicates that the crossover becomes a transition of first-order character in the superconducting state. Our results shows that metamagnetism induces a transition inside the superconducting phase and stabilizes the superconductivity for the field along the easy axis.

The interplay between magnetism and superconductivity is one of the most intriguing subjects in condensed matter physics. In the best-studied superconductor classes, such as high- T_c cuprate, iron-pnictides, heavy fermion systems, superconductivity appears on the verge of antiferromagnetism and the quantum-critical fluctuations are suggested to provide the attractive interactions of superconducting (SC) paring [1–3].

А recently discovered superconductor, UTe_2 $(T_c=1.6 \text{ K})$ [4], which was proposed to be near ferromagnetic quantum critical point, attracts much attention because of the unusual SC properties and the possibility of spin-triplet paring [4–14]. The SC transition temperature T_c for magnetic field along b-axis is initially suppressed, but remarkably, enhanced above $\sim 15 \,\mathrm{T}$ [9, 15, 16]. For certain field angles between the b and c-axes, superconductivity is once completely suppressed, but reappears at a very high field of 40 T [6, 17]. Such reinforcement and re-entrance are reminiscent of some other uranium ferromagnetic superconductors, like URhGe and UCoGe [18]. However, unlike these superconductors, UTe₂ does not exhibit a static ferromagnetic (FM) order. Therefore, the compound was suggested to be located at a ferromagnetic quantum critical point. This is supported by divergencies of magnetic susceptibility and muon spin relaxation rate at low temperatures [19]. However, we note that such divergence of magnetic susceptibility is absent in higher-quality single crystals recently grown through the molten-salt flux (MSF) method [20]. This result is consistent with the temperature-independent Knight shift of the ¹²⁵Te nuclear magnetic resonance at low temperatures [21]. Instead of FM fluctuations, magnetic excitations at the incommensulate Q-vector were observed in neutron scattering experiments [22, 23]. Thus, the properties of magnetic fluctuations, which are responsible for the SC paring, remain unknown [24]. The

paring symmetry is extensively studied to understand the mechanism of supercondeuctivity [13, 14, 25–29]. Furthermore, significant efforts are made to improve the crystal quality [20, 30–32]. In the best samples, T_c is significantly enhanced to over 2 K from the originally reported value [20, 31–33].

In strong magnetic fields, metamagnetism might play an important role in stabilizing the superconductivity in UTe_2 . For the magnetic field along the *b*-axis, a sharp metamagnetic (MM) transition is observed at 35 T [15, 34]. Due to MM fluctuations, an enhancement of electron mass with the field is observed and proposed to be the driving force for the SC reinforcement [9, 15, 16]. Meanwhile, for magnetic field along the *a*-axis only a weak metamagnetism is observed in the magnetization around 7 T [34]. Anomalies are also detected in the transport measurements at similar fields, which are suggested to be associated with a Fermi-surface instability [35]. Interestingly, as pointed out in the literature [34, 35], this weak metamagnetism around 7 T for $B \parallel a$ -axis occurs with the same critical value of the magnetization (0.4 μ_B) for the sharp MM transition at 35 T for $B \parallel b$ -axis.

In this Letter, we study the influence of metamagnetism on the superconductivity for B||a-axis. From AC magnetic susceptibility, magnetization, and magnetocaloric effect measurements, we construct an SC phase diagram and map entropy in the temperature-magnetic field parameter space. A significant improvement of a sample quality achieved by the MSF method [20] leads to a largely enhanced upper critical field for B||a-axis, over the MM field of 7 T. This allows us to investigate the interplay between the metamagnetism and superconductivity for B||a-axis. For the first time, we will show herein that the metamagnetism induces a transition inside the SC phase and causes the enhancement of the upper critical field (Fig. 1).

The AC magnetic susceptibility was measured at temperatures down to $75\,\mathrm{mK}$ and magnetic fields up to $15\,\mathrm{T}$ in a He³-He⁴ dilution refrigerator with an SC magnet. Magnetization was measured for the field up to 9 T using a vibration-sample magnetometer (VSM) in a commercial Physical Property Measurement System of Quantum Design. Magnetization above 9 T was measured by VSM in the High-Field Laboratory for Superconducting Materials at the Institute for Materials Research at Tohoku University. We measured M(B) at various temperatures to obtain a magnetization landscape (see Supplemental Material [36]). From the slope along the *T*-axis we obtain the contour data of the field derivative of entropy, using a thermodynamic relation, $(\partial M/\partial T)_B = (\partial S/\partial B)_T$. The entropy increment from zero field can be obtained by integrating $\partial S/\partial B$ over the magnetic field. The magnetocaloric effect, $(\partial T/\partial B)_S$, was measured using the alternating-field method [37]. High quality single crystals grown by the MSF method were used. The T_c for the same badge is $2.0 \,\mathrm{K}$.

Figure 1 shows the SC phase diagram for the magnetic field along the three principal axes. The unusual upward curvature of the $T_c(B)$ for $B \parallel b$ -axis (green triangles) signals the field-induced reinforcement of the superconductivity in higher fields, connected to a sharp MM transition at 35 T. Notably, the $T_c(B)$ for $B \parallel a$ -axis (blue circles) also exhibits an upward curvature above $\sim 6 \,\mathrm{T}$ and the deviation from the calculation for the strong coupling regime (red dotted line). For the calculation, spin triplet paring is mimicked by omitting the Pauli limiting effect. This upward curvature is in a sharp contrast to the downward curvature for a sample with a lower T_c (pastel blue diamonds) [4]. Consequently, the upper critical field B_{c2} for B||a-axis is largely enhanced to almost twice the value of the lower- T_c sample grown from chemical vapor transport (CVT) method, even though the $T_c(0)$ is enhanced only by 25%. Below we show that the MM crossover induces a transition inside SC phase and is responsible for the enhancement of B_{c2} .

For an ideal paramagnet the entropy decreases with the field at any temperature, because the magnetic field aligns magnetic moments (see Supplemental Material [36]). In heavy fermion compounds, there is a crossover across $T_{\rm K}$. At high temperatures above $T_{\rm K}$ the paramagnetic behavior is expected, while below $T_{\rm K}$ the entropy depends on the subtle field variations of the density of states, because in Fermi liquid (FL) $S=\gamma T$, where γ is Sommerfeld coefficient (see Supplemental Material [36]).

The obtained entropy increment for UTe₂ for B||aaxis exhibits a typical paramagnetic behavior with the decreasing entropy with the field above 15 K, while below 15 K field dependence of entropy becomes weaker (see Supplemental Material [36]). However, a closer inspection reveals an anomalous enhancement around 6-9 T only at low temperatures below 6 K. Figure 2(a) displays

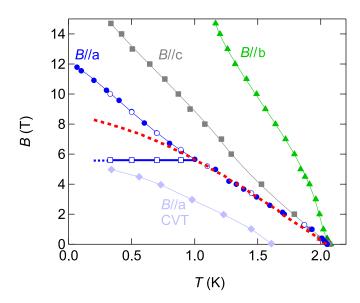


FIG. 1. Superconduting phase diagram of the high-quality single crystals of UTe₂, grown by the molten-salt flux method [20], for the magnetic field along the three principal axes. B_{c2} is determined by the AC magnetic susceptibility (solid green, grey, and blue symbols, see Supplemental Material [36]) and the magnetocaloric effect (blue open symbols, Fig. 3(d)). The blue open squares are the transition observed inside the SC state for B||a (Fig. 3(d)). The solid lines are guide for the eyes. For comparison, B_{c2} of samples grown by chemical vapor transport (CVT) from the initial report in [4] is plotted (pastel blue diamonds). The red dotted line is the calculation for superconductors in the strong coupling regime [38], where Pauli-limiting effect is omitted, in order to mimic spin-triplet paring.

the contour plot of $\partial S/\partial B$ with the red, blue, and white for positive, negative, and 0 values, respectively. An anomalous positive $\partial S/\partial B$ (red) region is observed in the paramagnetic background with a large negative $\partial S/\partial B$ values (blue). The white line in the lower field side corresponds to $\partial S/\partial B=0$ from negative to positive, therefore an entropy minimum. On the other hand the white line in the higher field side corresponds to an entropy maximum (Fig. 2(b)). The magnetic susceptibility $\partial M/\partial B$ shows a maximum around this magnetic field (Fig. 2(c)), in agreement with the result of previous study on magnetization [34]. The weak peak in the magnetic susceptibility $\partial M/\partial B$ is a clear signature of an MM crossover. We mention that the peak in $\partial M/\partial B$ is at a slightly lower field than the maximum position of entropy, although they are usually at the same position because $\partial M/\partial T = \partial S/\partial B$ changes the sign at an MM field [39, 41– 43] (Fig. 3(a,b)). The discrepancy is probably caused by the large decreasing background contribution in the $\partial M/\partial B$ superimposed by the relatively small peak. By contrast, S is a better probe to detect the crossover, because the peak to background ratio is larger than that for $\partial M/\partial B$.

We extend the detection of entropy anomaly to lower

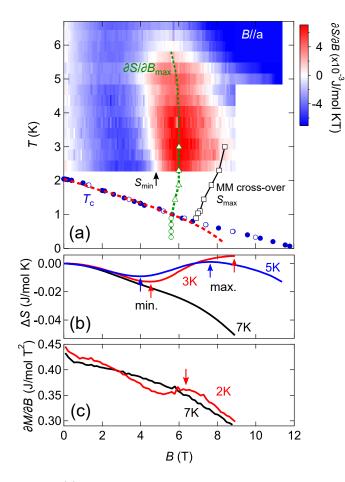


FIG. 2. (a) Color contour plot for the field derivative of magnetic entropy $\partial S/\partial B$ of UTe₂ for $B \parallel a$ -axis. The green dotted line corresponds to the maximum positions of $\partial S/\partial B$. Green open triangles and circles are the position of a minimum in Magnetic Grüneisen parameter, $\Gamma_{\rm B}$ (Fig. 3(d)). The former is a broad minimum, while the latter is a sharp minimum, indicative of a transition of first-order character. The black open squares indicate an entropy maximum at the metamagnetic crossover determined by magnetocaloric effect. The blue solid/open circles are T_c determined by magnetic susceptibility/magnetocaloric effect (Fig. 1 [36]. The red dotted line is a calculation for superconductors in the strong coupling regime. Pauli-limiting effect is omitted. (b) The entropy increment from zero field at different temperatures is plotted against magnetic field. (c) The magnetic susceptibility $\partial M/\partial dB$ at different temperatures is plotted against magnetic field.

temperatures down to 0.33 K using the magnetocaloric effect (MCE), $(\partial T/\partial B)_S$ (Fig. 3(d)). The MCE is often used to detect bulk transitions because it is sensitive to field-induced phenomena [44]. A system with an MM crossover without a sharp transition can be understood with a critical end point (CEP) at a negative temperature (Fig. 3(a)). The entropy shows a broad maximum at the MM crossover field [39] (Fig. 3(b)). The entropy maximum always accompanies a minimum at the low-field side because of paramagnetic background with negative $\partial S/\partial B$. The magnetic Grüneisen parameter, which is a

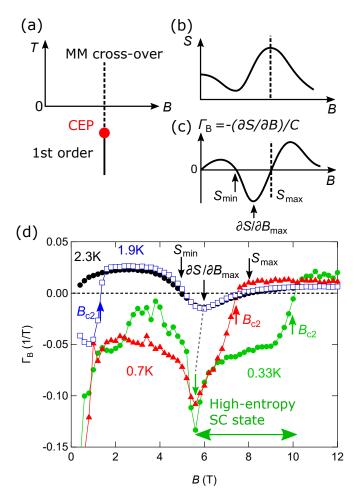


FIG. 3. (a) *T-B* phase diagram with a metamagnetic crossover. When critical end point (CEP) is tuned below zero temperature, the system shows only a crossover. (b) A metamagnetic crossover causes an entropy maximum as a function of magnetic field. (c) Magnetic Grüneisen parameter, $\Gamma_{\rm B} = (\partial T / \partial B) / T = -(\partial S / \partial B) / C$. Metamagnetism causes a sign-change at a metamagnetic crossover [39–41]. (d) Magnetic Grüneisen parameter, $\Gamma_{\rm B}$, for UTe₂ as a function of the magnetic field along *a*-axis. For the data at 2.3 K in the normal-conducting states, three arrows denotes the positions of entropy minimum $S_{\rm min}$, the steepest increase of the entropy $\partial S / \partial B_{\rm max}$ and entropy maximum $S_{\rm max}$. Green double-sided arrow indicates superconducting state at 0.33 K with higher entropy compared to the low-field superconducting state.

T-coefficient of magnetocaloric effect, $\Gamma_{\rm B} = (\partial T/\partial B)/T$, exhibits a sign-change at the entropy maximum because $\Gamma_{\rm B}$ is also expressed as $\Gamma_{\rm B} = -(\partial S/\partial B)/C$ and the specific heat C is always positive (Fig. 3(c)). The sign-change is accompanied by another sign-change (minimum in entropy, $S_{\rm min}$) and a minimum (steepest slope in entropy, $\partial S/\partial B_{\rm max}$). Indeed, at 2.3 K in the normal conducting state $\Gamma_{\rm B}$ shows the expected behavior with a sign-change at 8 T from negative to positive at $S_{\rm max}$ and another sign-change at 5.0 T and a minimum at 6.0 T (Fig. 3(d)). A stepwise reduction to negative values with decreasing field across B_{c2} is observed at a decreased temperature slightly below T_c because the entropy increases with the field up to B_{c2} due to the destruction of the SC order by the magnetic field. The sign-change around 8 T is not any more visible in SC state for T=0.7 and 0.33 K because of the large negative SC contribution. Note that at low temperatures, the sign-change is taken over by the SC phase transition at B_{c2} . While the sign-change caused by the MM crossover disappears, the $\Gamma_{\rm B}$ minimum surprisingly transforms into a sharp negative peak in the SC state. Such a negative peak (positive peak in $\partial S/\partial B$) corresponds to a step-wise increase in entropy. Therefore, the anomaly indicates a first-order character of this transition.

We added the points of the $\Gamma_{\rm B}$ minimum $(\partial S/\partial B_{\rm max})$, green dotted line and green open symbols) and the signchange at the MM crossover with S_{max} (open squares) The MM crossover line touches the SC in Fig. 2. phase boundary at 7 T. The $\partial S/\partial B_{\text{max}}$ in the normalconducting state is connected to the transition line at 5.6 T. Notably, the anomaly sharpens in the SC phase, although an anomaly of Fermi surface (FS) instability, such as the MM crossover, would weaken because of the opening of SC gap on FS. Therefore, the transition inside SC state would not be simply explained by the MM crossover persisting inside SC state, but rather suggestive of an induced transition between two distinct SC states. For $B \parallel a$ -axis, the symmetry is reduced to C_{2h} , which has two odd-parity irreducible representations, $A_u^{2h} = A_u + B_{3u}$ and $B_u^{2h} = B_{1u} + B_{2u}$ [26, 45]. A possible scenario is thus the transition between A_u^{2h} to B_u^{2h} . On the other hand, because of its first-order nature, it can be a transition within the same symmetry, for example, a step-wise increase of B_{3u} weight at the transition between two A_u^{2h} states. In this case, since A_u is fully gapped and B_{3u} is point nodal, a larger entropy is expected above the transition.

We note that the MM crossover is found also in low T_c samples [34]. The low T_c samples do not show an upward curvature in $T_c(B)$ because superconductivity is already suppressed to zero by the magnetic field before the entropy anomaly appears. We further mention that the B_{c2} enhancement above MM crossover is in a sharp contrast to the abrupt disruption of superconductivity above the MM transition for $B \parallel b$ -axis.

In summary, using high-quality single crystals of UTe₂, we investigated the interplay between metamagnetism and superconductivity for the field along the *a*-axis. The improvement of crystal quality largely enhances the upper critical field B_{c2} . In addition, the $T_c(B)$ shows an unusual upward curvature above ~ 6 T, which is reminiscent of the field-induced reinforcement of the SC for $B \parallel b$ -axis. Around the same magnetic field of 6 T, the entropy is enhanced at low temperatures below 6 K because of the weak metamagnetic crossover. Remarkably, the anomaly associated with the crossover persists even in the SC state, where the sharp negative peak in $\Gamma_{\rm B}$ indicates that the crossover becomes a transition of firstorder character below T_c . The transition separates the two SC states and causes a step-wise increase of entropy in the high-field SC state. In UTe₂, the metamagnetism plays a crucial role in stabilizing the superconductivity at high fields for both B||a and b-axes.

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Supplementally material for "Stabilization of superconductivity by metamagnetism in an easy-axis magnetic field on UTe₂"

DETERMINATION OF UPPER CRITICAL FIELD BY AC MAGNETIC SUSCEPTIBILITY

We use mainly AC magnetic susceptibility to determine the upper critical field B_{c2} of UTe₂ to construct the superconducting phase diagram, shown in the main text. Figure S1 displays the AC magnetic susceptibility at different temperatures for B_{\parallel} areas. The irreversibility point is taken as B_{c2} as indicated by arrows.

MAGNETIC ENTROPY OF IDEAL PARAMAGNETS

In ideal paramagnets the only energy scale is Zeeman splitting. Because it is linear in field B, entropy S(T,B) is a function of T/B, namely S(T/B). This is true for any size of magnetic moment J. In Fig. S2 we show a calculation of entropy for an ideal paramagnet (no magnetic interaction) with a size of magnetic moment J=1/2. As can be seen, isentropes are all linear crossing the origin. The full entropy of $R\ln(2)=5.8J/mol\cdot K$ is observed in a region where $k_BT \gg \mu_B B$. Entropy is always decreasing with field and reduced to zero in a region of $k_BT \ll \mu_B B$.

ENTROPY INCREMENT OF A HEAVY FERMION COMPOUND USn₃

In heavy fermion compounds above $T_{\rm K}$, f-electrons act as local moments. In such a temperature range magnetic entropy should be decreasing with magnetic field. On the other hand below $T_{\rm K}$, they form Fermi liquid state, where the entropy depends on the density of states, as $S=\gamma T$, where γ is Sommerfeld coefficient. Therefore, a cross-over from the high-temperature region with decreasing S with the field to low-temperature region with the subtle change of S caused by the field dependence of density of states is expected. The latter is usually small unless $T_{\rm K}$ is so small (γ is so large) that some small magnetic field changes the density of states significantly, because a condition of $\mu_B B \sim k_B T$ is reached. In UTe₂ $\gamma=0.13 \, \text{J/mol·K}$. Here, we show a case of USn₃ with a similar γ value of 0.17 J/mol·K [1].

We plot representative magnetization data of USn₃ in Fig. S3(a) to construct a magnetization mapping shown in Fig. S3(b). Then, $\partial M/\partial T = \partial S/\partial B$ is obtained and finally we have the increment of entropy from zero field ΔS by integrating $\partial M/\partial T$ over magnetic field (Fig. S3(c)). We observe clearly decreasing entropy at high temperatures above ~20 K and more or less constant value at low temperatures. This behavior of entropy is consistent with the reported $T_{\rm K}$ of 13-30 K [2, 3].

ENTROPY INCREMENT OF UTe_2 FOR THE FIELD ALONG a-AXIS

Magnetization is measured at many different temperatures to construct a contour plot (Fig. S4). From the slope along *T*-axis we obtain a contour plot for magnetic-field derivative of entropy, using a thermodynamic relation, $(\partial M/\partial T)_B = (\partial S/\partial B)_T$. Then, entropy increment from zero field can be obtained by integrating $\partial S/\partial B$ over magnetic field.

We plot in Fig. S5 the entropy increment of UTe_2 for the field along a-axis. It shows a paramagnetic behavior with decreasing entropy above ~15 K. Below this temperature it is more or less constant, similar to USn₃. A close inspection for low-temperature region reveals an anomaly, as shown in Fig. S6. A minimum is clearly observed around 5 T and an increase above this field.

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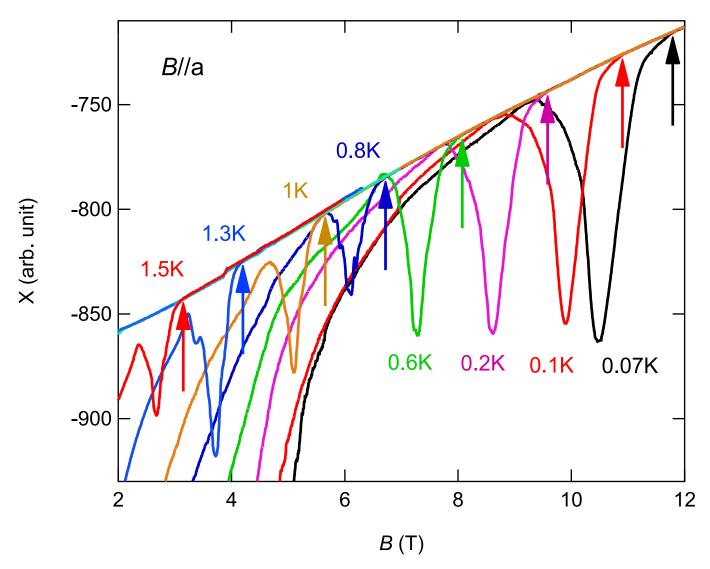


FIG. S1. Determination of the upper critical field B_{c2} of superconductivity in UTe₂ for the field along *a*-axis by ac-magnetic susceptibility χ . Arrows indicate B_{c2} .

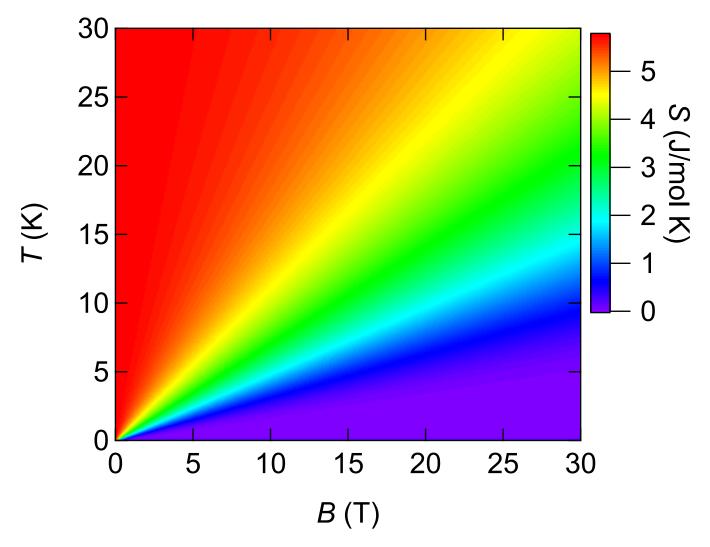


FIG. S2. Entropy of an ideal paramagnet with a size of magnetic moment J=1/2. We set g-factor to 2. Magnetization is $M=N\mu_B \tanh(\mu_B B/k_B T)$.

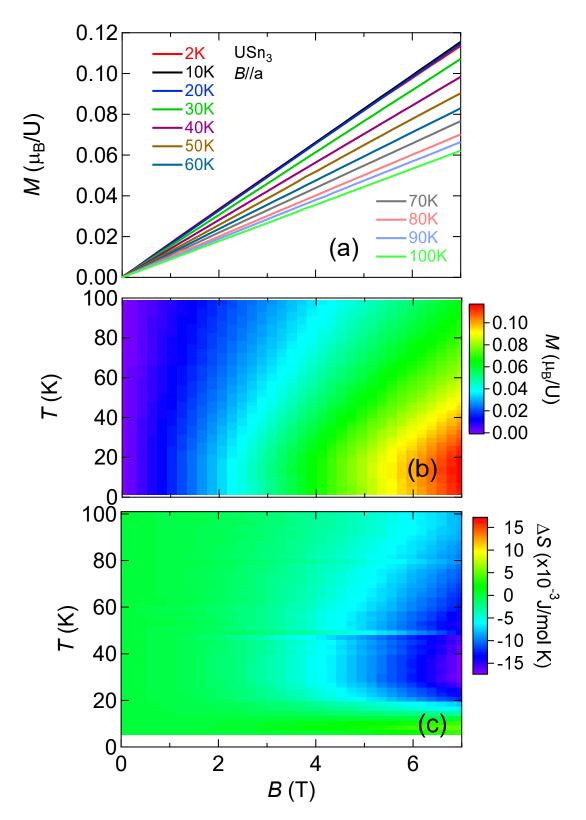


FIG. S3. (a) Representative magnetization data of USn₃ for the field along a-axis to construct entropy contour. M(B) curves are taken with a typical temperature interval of 1 K up to 60 K and 2 K between 60 and 100K. Contour plots of magnetization (b) and entropy increment (c) of USn₃.

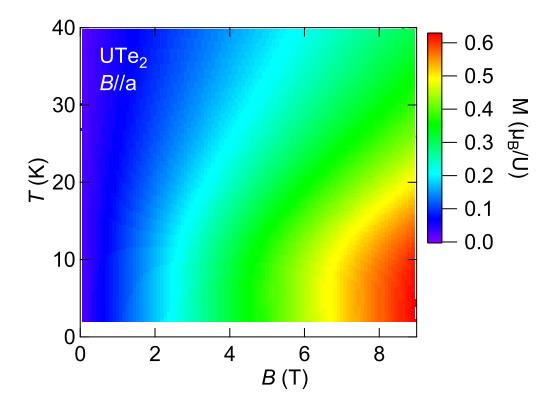


FIG. S4. Contour plot of magnetization of UTe_2 for the field along a-axis.

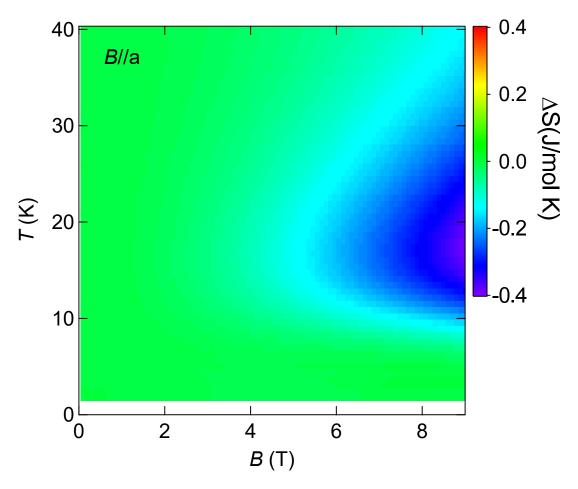


FIG. S5. Entropy increment of UTe_2 for the field along a-axis.

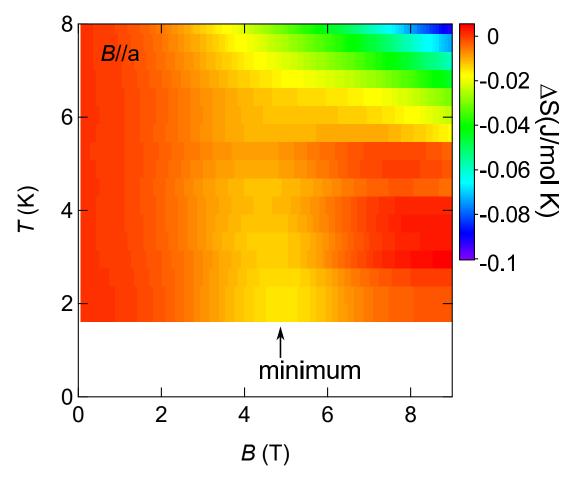


FIG. S6. Blow up of Fig. S5 for the low-temperature region.