Letter

Reinforcement of superconductivity by quantum critical fluctuations of metamagnetism in UTe_2

Y. Tokiwa,[1,*] P. Opletal,[1] H. Sakai,[1] S. Kambe,[1] E. Yamamoto,[1] M. Kimata,[2] S. Awaji,[2] T. Sasaki,[2] D. Aoki,[3] Y. Haga,[1] and Y. Tokunaga[1]

[1]*ASRC, Japan Atomic Energy Agency Tokai, Ibaraki 319-1195, Japan*
[2]*IMR, Tohoku University, Sendai, Miyagi 980-8577, Japan*
[3]*IMR, Tohoku University, Ibaraki 311-1313, Japan*

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The normal-conducting state of the superconductor UTe_2 is studied by entropy analysis for magnetic fields along the b axis, obtained from magnetization using the relation $(\partial M/\partial T)_B = (\partial S/\partial B)_T$. We observe a strong increase in entropy with magnetic field due to metamagnetic (MM) fluctuations (spatially uniform, $Q = 0$). The field dependence is well described by the Hertz-Millis-Moriya theory for quantum criticality of itinerant MM. Notably, the lower bound of the quantum-critical region coincides well with the position of the minimum in the superconducting transition temperature $T_c(B)$. Hence, our results suggest that $Q = 0$ fluctuations reinforce the superconductivity.

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Unconventional superconductivity arises from an anomalous normal-conducting (NC) state [1–3]. The NC states of heavy-fermion, iron-pnictide, and cuprate high-T_c superconductors exhibit non-Fermi liquid (non-FL) behavior with unusual T dependence of resistivity due to quantum fluctuations from magnetic quantum criticality. Extensive studies have established close relationships between superconductivity and magnetic criticality, leading to the widely accepted belief that the superconducting (SC) pairing interaction is provided by such fluctuations. Hence, investigating the NC states of unconventional superconductors is of great importance to gain insights into the SC pairing mechanism.

In this letter, our focus is on the recently discovered superconductor UTe_2, which undergoes a SC transition at $T_c = 1.6$ K [4,5]. The superconductivity has garnered significant attention due to its reinforcement (re-entrance) under high magnetic fields and the presence of multiple SC phases [4,6–10]. The pairing symmetry has been subject to extensive investigations [11–16]. Furthermore, considerable effort has been dedicated to improving the crystal quality, leading to a significant enhancement of T_c to >2 K from the originally reported value [17–20]. Notably, recent progress has allowed the successful observation of quantum oscillations in high-quality single crystals grown using the molten-salt flux (MSF) method [21,22].

The reinforcement of superconductivity is observed when the field is applied along the hard b axis, where a sharp metamagnetic (MM) transition takes place around 35 T [23,24]. There, the SC transition temperature T_c is initially suppressed with magnetic field; however, it remarkably enhances above $B^\star \sim 15$ T. As a result, a minimum in $T_c(B)$ is observed as a function of magnetic field. For certain magnetic field angles between the b and c axes, the magnetic field completely suppresses the superconductivity, but above 40 T, the superconductivity reappears [6,25]. This reinforcement and reappearance bears a resemblance to other uranium ferromagnetic (FM) superconductors, URhGe and UCoGe, of which the superconductivity may be mediated by FM fluctuations [26–30]. Unlike these superconductors, UTe_2 is paramagnetic. The absence of FM and the similarity to FM superconductors open the intriguing possibility of FM quantum fluctuations providing the SC pairing interaction. However, the magnetic susceptibility of recently grown high-quality crystals tends to saturate at low temperatures [20], which is consistent with a temperature-independent Knight shift [31]. Instead, magnetic excitations at incommensurate Q vectors have been observed by neutron scattering experiments [32,33]. Therefore, the nature of magnetic fluctuations responsible for the superconductivity remains unknown [34]. A broad anomaly observed in the NC state around 15 K in several thermodynamic, transport, and nuclear magnetic resonance (NMR) experiments may be related to the key magnetic fluctuations [31,35–37].

In strong magnetic fields along the b axis, fluctuations associated with the MM transition likely play a crucial role. MM is identical to FM in the sense that they both correspond to a spatially uniform ($Q = 0$) magnetic instability, as evidenced by the discontinuity in uniform magnetization $M(Q = 0)$ at the transition field. The only distinction from FM lies in its induction by magnetic field. When the critical endpoint (CEP) of MM is tuned to zero temperature, the quantum fluctuations lead to the formation of an anomalous metallic state [38,39]. Even when a system is not precisely tuned to quantum CEP but is in close proximity with the CEP at a finite temperature, the quantum-critical fluctuations can still induce an anomalous state [38–44]. In UTe_2, an increase in electron mass and longitudinal fluctuations with the field toward the MM is observed and is proposed to drive the reinforcement of superconductivity [9,23,45,46].

Entropy is a direct thermodynamic measure of fluctuations, as it accumulates at a quantum critical point and exhibits a

*tokiwa.yoshifumi@jaea.go.jp

peak as a function of control parameters, such as magnetic field and pressure [47,48]. Because unconventional SC pairing is believed to be mediated by such quantum critical fluctuations, it is interesting to map entropy in some parameter space. In UTe_2, the superconductivity couples to the magnetic field in a very peculiar manner, as evidenced by the nonmonotonic behavior of $T_c(B)$. Therefore, the magnetic field–temperature parameter space is the most interesting to map entropy. To achieve this, we performed high-resolution magnetization measurements for the $B \parallel b$ axis, enabling us to extract the field dependence of entropy in the NC state of UTe_2 using a thermodynamic relation $(\partial M/\partial T)_B = (\partial S/\partial B)_T$. The consistency with specific heat [9] is checked by comparing C/T and $\int (dM/dT)/T\,dB = \int (dS/dB)/T\,dB = \gamma$ at low temperatures in the Fermi liquid (FL) regime, where γ is the Sommerfeld coefficient (see Supplemental Material [49]). In this letter, we reveal an anomalous increase in fluctuations attributed to MM in an entropy map. We investigate the nature of these fluctuations by fitting their field dependence with the Hertz-Millis-Moriya theory, which pertains to the quantum criticality of itinerant MM [38,39]. The theory, widely recognized as a standard, is frequently employed to describe the anomalous behavior associated with MM quantum criticality [40,42,43,52,53].

Magnetization is measured by a vibration-sample magnetometer with an accuracy of $\sim 2 \times 10^{-4}$ emu in magnetic fields up to 24 T and for temperatures between 2.2 and 65 K, in the High-Field Laboratory for Superconducting Materials at the Institute for Materials Research at Tohoku University. We measured magnetization $M(B)$ at various temperatures with typical intervals of 1 K up to 20 K, 2 K up to 40 K and 5 K up to 65 K to obtain a magnetization landscape (see Supplemental Material [49]). From the slope along the T axis, we have a landscape of field derivative of entropy $\partial M/\partial T = \partial S/\partial B$. Then $\partial S/\partial B$ is integrated over B to obtain entropy increment ΔS from zero field, namely, $\Delta S = \int_0^B (\partial M/\partial T) dB$. Note that ΔS does not contain entropy at zero field. A large single crystal with a weight of 94 mg, grown by the chemical vapor transport method, is used. The SC transition temperature, the residual resistivity ratio, and the residual specific heat coefficient divided by the value at the NC state of the same batch are, respectively, $T_c = 1.7$ K, RRR = 25, and $\gamma(0)/\gamma_N = 0.43$. We note that the properties of the NC state remain unaffected by the sample quality. Specifically, the characteristics of magnetic fluctuations and the MM critical field show no variation across crystals of different quality [46,54]. We will delve into the discussion later, explaining how this robustness in the properties of MM leads to the independence of $B^\star$, where $T_c(B)$ exhibits a minimum, from variations in the sample quality.

Figure 1(a) shows the magnetization of UTe_2 for the field along the b axis. Magnetization at 10 and 20 K shows clearly the stronger-than-linear field dependence due to MM fluctuations, which we will discuss later. Note that the magnetization of a paramagnet, such as those described by the Brillouin function, exhibits a downward curvature. As a consequence, the differential magnetic susceptibility ($\chi = dM/dB$) normalized by the zero-field value of a paramagnet is always <1 under magnetic fields $\chi/\chi_{B=0} < 1$. Figure 1(b) displays such a normalized magnetic susceptibility of UTe_2 for the field along the b axis. Reflecting the nonlinear magnetization, χ at 10 and 20 K increases with magnetic field. The nonlinear magnetization (χ >1) is observed up to 30 K, indicating the influence of MM fluctuations in a wide temperature range. Notably, χ at 10 and 20 K changes its slope above ∼15 T. At 2.3 K, χ is nearly constant below ∼15 T, but it deviates from the constant value above ∼15 T.

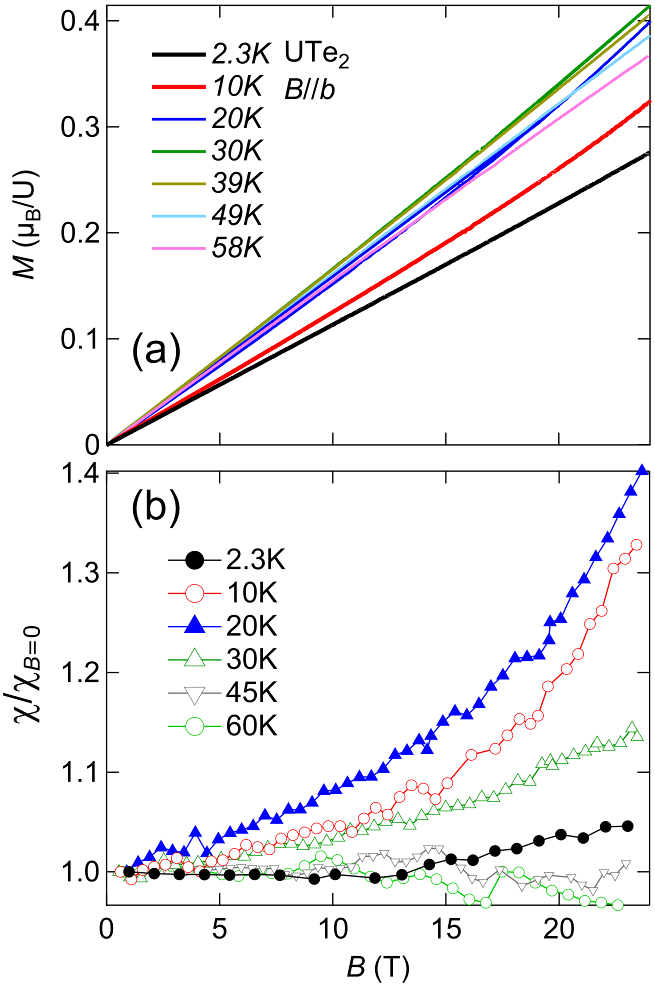

FIG. 1. (a) Magnetization of UTe_2 for the field along the b axis at different temperatures. (b) Magnetic field dependence of differential magnetic susceptibility normalized by the zero field values $\chi/\chi_{B=0}$ of UTe_2 at different temperatures.

From the $M(B)$ measurements, we obtain magnetic entropy increment ΔS, as shown in Fig. 2. For an ideal paramagnet, entropy decreases with magnetic field because magnetic field aligns magnetic moments. In heavy fermion compounds, there is a crossover across T_K [55]. At high temperatures above T_K, paramagnetic behavior is expected, while below T_K, entropy depends on subtle field variations of density of states since, in FL, $S = \gamma T$.

In UTe_2, the paramagnetic behavior with decreasing entropy is found at temperatures above ∼35 K, as shown in Fig. 2. This temperature 35 K corresponds to a maximum of magnetization as a function of temperature, where $\partial M/\partial T = \partial S/\partial B = 0$, in agreement with the reported maximum of magnetic susceptibility around $T_{\chi\max} \sim 35$ K [4,24]. Across

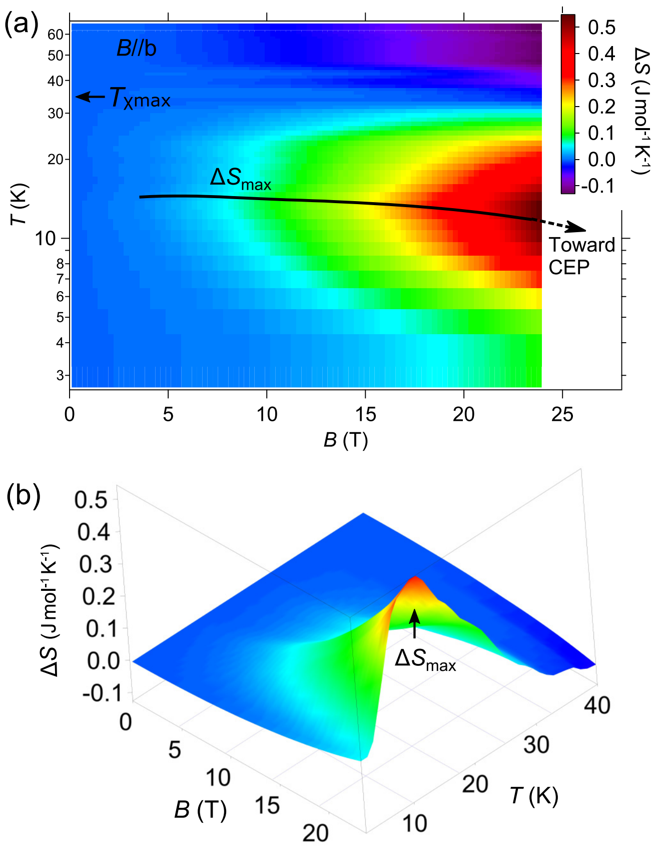

FIG. 2. (a) Color contour plot for magnetic entropy increment ΔS of UTe_2 in $T-B$ phase diagram for the $B \parallel b$ axis. Solid black line corresponds to a maximum of $\Delta S(T)$ as a function of temperature. CEP denotes a critical endpoint for metamagnetic transition. $T_{\chi\max}$ denotes the temperature where magnetic susceptibility at zero field shows a maximum [4,5,24,56,57]. (b) Three-dimensional plot of ΔS as a function of magnetic field and temperature. The arrow denotes the position of ΔS maximum in the temperature dependence at the highest magnetic field of 24 T.

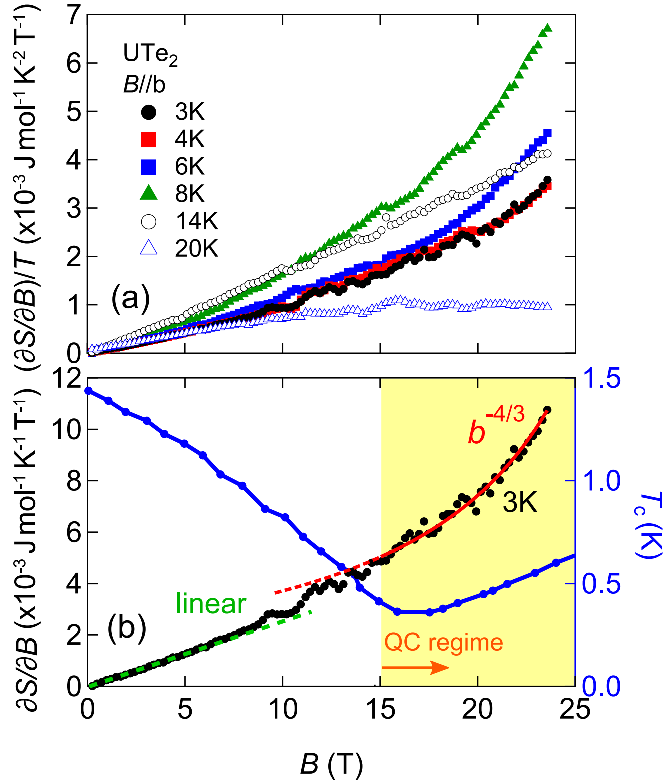

FIG. 3. (a) Magnetic field derivative of entropy divided by temperature $(\partial S/\partial B)/T$ of UTe_2 as a function of magnetic field along the b axis at different temperatures. (b) Comparison between magnetic field derivative of entropy (left axis, black circles) and the superconducting transition temperature T_c (right axis, blue circles) [6]. Green dotted linear line is a guide for the eye. Red solid line represents $b^{-4/3}$ field dependence, where $b = |B - B_c|$ and $B_c = 34.75$ T is the metamagnetic (MM) critical field [9]. QC regime denotes the field range of dominating quantum critical component where entropy follows the theoretical power law for MM quantum criticality of two dimensions (red solid line) [39].

this temperature, $\partial S/\partial B$ changes its sign. Below this temperature, M decreases with decreasing T, therefore, entropy increases with magnetic field $\partial M/\partial T = \partial S/\partial B > 0$. Such an entropy increase is commonly observed in compounds with MM transitions/crossovers due to increasing MM fluctuations toward the critical field [41,42,58]. In Figs. 2(a) and 2(b), we plot a position of $\Delta S_{\max}$, where ΔS is the most as a function of temperature (see also Supplemental Material for the temperature dependence of ΔS at different magnetic fields [49]). It shifts to lower temperatures with magnetic field, approaching the CEP of MM. At lower temperatures below the CEP, the entropy increases becomes smaller. This is attributed to the weakening of magnetic fluctuations caused by the sharpening of MM transition with decreasing temperature. We note that the entropy increase remains finite even at low temperatures, as can be seen in Fig. 3(a), likely playing a crucial role in the reinforcement of superconductivity. Importantly, as demonstrated by Fig. 2, the entropy increase at low temperatures is caused by the MM, which represents a $Q = 0$ instability.

In Fig. 3(a), we plot $(\partial M/\partial T)/T = (\partial S/\partial B)/T$. In the FL state, where $S = C = \gamma T$, this quantity equals the field derivative of the Sommerfeld coefficient $\partial\gamma/\partial B$, which is T independent. The datasets at 3 and 4 K lay on top of each other, indicating formation of an FL state at temperatures below 4 K. They exhibit the upward curvature, indicating an accelerating increase of entropy at high fields. The upward curvature persists up to ~8 K and changes to a downward curvature at high temperatures above the line of $\Delta S_{\max}$. The magnetic field derivative of entropy $\partial S/\partial B$ at 3 K in the FL regime is plotted in Fig. 3(b). At low fields, it is linear in field, as expected from a symmetry consideration $\partial S/\partial B(B) = -\partial S/\partial B(-B)$ with a leading B-linear term. The data gradually deviate from the linear dependence and increase rapidly with a positive curvature at high fields.

According to the Hertz-Millis-Moriya theory [38,39], $\partial S/\partial B$ in the critical regime follows $b^{(d-6)/3}$, where $b = |B - B_c|$ is the distance to the critical field B_c, and d is the dimensionality. Since Fermi surfaces consist of mainly cylindrical ones [21,22,59], $d = 2$ is appropriate, resulting in a power-law dependence $b^{-4/3}$. By fixing $B_c = 34.75$ T of a recently reported detailed study [9], the data are well described by $b^{-4/3}$ >15 T (see also Supplemental Material for fitting with other cases such as three-dimensional (3D) MM and antiferromagnetic (AFM) fluctuations [49]). The best fit among all the considered models is achieved with two-dimensional (2D) MM, suggesting that 2D MM quantum critical fluctuations are dominating at high magnetic fields. The lower-bound field of 15 T coincides well with the field of $T_c(B)$ minimum $B^\star$. Furthermore, above the same field, specific heat [9] also follows the theoretical power law, in agreement with our result (see Supplemental Material [49]). This indicates that the system at low fields is initially noncritical with the linear-in-field dependence and then crosses over to the 2D quantum critical behavior above 15 T. Given that the theory addresses $Q = 0$ fluctuations [39], the coincidence between $B^\star$ and the lower field boundary of the quantum critical regime hints at the involvement of $Q = 0$ MM quantum fluctuations in bolstering the SC transition temperature.

While this correlation is suggestive, it should be interpreted with caution until further evidence solidifies this connection because AFM ($Q \neq 0$) excitations have been identified in neutron scattering experiments, as reported in Refs. [32–34]. A key unresolved question concerns how these AFM fluctuations evolve under magnetic fields. The observation that ΔS without zero-field entropy markedly reflects the anomaly associated with the MM underscores the predominant role of MM at magnetic fields. We note that the change in entropy in this letter, represented by ΔS, accounts for merely 10% of $R\ln(2)$ at its peak. This is substantially smaller than the entropy increase with temperature at zero magnetic field, which reaches 80% of $R\ln(2)$ at 25 K [37]. Consequently, the anomaly due to MM become less discernible in the absolute entropy, obscured by the dominant zero-field entropy (see Supplemental Material [49]). This could be interpreted as, at zero (or low) magnetic fields, AFM contributions being more significant, whereas at high magnetic fields, the influence of MM prevails. Such a trend aligns well with the fundamental behavior of AFM coupling, which tends to diminish in the presence of magnetic fields. Furthermore, a recent NMR study highlights dominant FM fluctuations along the *a* axis [60]. While our findings suggest the dominance of $Q = 0$ fluctuations, further direct investigation into magnetic fluctuations with Q-resolved experiments, particularly through neutron scattering, is essential for further understanding.

The recent improvements of sample quality have enhanced T_c by 25% from the originally reported value [20]. Notably, the upper critical field B_{c2} is enhanced twice for the field along the magnetic easy *a* axis, owing to a weak MM ~8 T [55]. In sharp contrast, the $T_c(B)$ curve for the field along the *b* axis does not change its characteristic shape with a minimum at $B^\star$. Remarkably, $B^\star$ is independent from the sample quality [10]. This is most likely because $B^\star$ is determined by MM fluctuations, which are not influenced by the crystal quality [46]. This assertion is further supported by the fact that the MM critical field is independent from the sample quality [54].

In this letter, we show the accelerated increase of entropy with magnetic field along the *b* axis in UTe_2 due to $Q = 0$ MM fluctuations. The field dependence in the FL regime is well described by the Hertz-Millis-Moriya theory for quantum criticality of itinerant MM. The coincidence between the low-field bound of the quantum-critical fluctuations and $B^\star$ indicates that $Q = 0$ fluctuations boost superconductivity. This is in line with the result for the field along the easy *a* axis, showing that the weak MM strongly enhances the upper critical field [55]. Thus, our results show that $Q = 0$ fluctuations play crucial roles in the spin-triplet superconductivity in UTe_2 and promote further experimental research to more conclusively determine the nature of fluctuations under high magnetic fields.

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