Thermodynamic evidence for high-field bulk superconductivity in UTe$_2$

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(Dated: June 15, 2022)

UTe$_2$ has been of great interest in recent years due to its proposed unconventional magnetic field re-enforced spin-triplet superconducting phases persisting at fields far above the simple Pauli limit for $H \parallel b$. A high-field phase also emerges above 40 T within the field polarized paramagnetic phase when $H$ is applied close to the [011] direction. Here we present thermodynamic evidence of the field stabilized superconducting phases in UTe$_2$ via angular dependent magnetocaloric measurements in pulsed magnetic fields. We observe a pronounced heating effect during the metamagnetic transition when $H$ is applied along the [011] direction, within the high-field phase, indicative of vortex movement in a bulk superconducting state. Our results support the phase diagram established by previous electrical transport measurements. A more subtle feature observed around 15 T supports a phase transition between two separate superconducting states when the field orientation is within a few degrees of the $b$ axis.

I. INTRODUCTION

The recently discovered heavy-fermion superconductor UTe$_2$ is a promising candidate for the realization of chiral spin-triplet superconductivity with equal-spin pairing. Support for this picture comes from its close proximity to magnetic order, its unusually large critical magnetic field (far exceeding the Pauli limit for a weakly coupled BCS superconductor), as well as the observation of only a small change in the Knight shift below its superconducting transition temperature $T_c \approx 1.6$ K [1–7].

UTe$_2$ crystallizes in a body-centered orthorhombic structure ($Immm$) [8]. Unlike closely related orthorhombic (Pnma) URhGe and UCoGe, for which superconductivity emerges within the ferromagnetically ordered state [7], no magnetic order has been observed in UTe$_2$ down to 25 mK [8, 9]. While strong magnetic fluctuations are believed to play a major role in facilitating superconductivity in UTe$_2$ [7], the nature of those is still a matter of contention. Indeed, whilst some experiments give evidence for ferromagnetic fluctuations [1], recent neutron scattering data show excitations at an antiferromagnetic wave-vector, indicating dominant antiferromagnetic fluctuation driven by RKKY interactions between the 4$f$ moments [10]. Studies under hydrostatic pressure also support the presence of antiferromagnetic fluctuations [11, 12].

When a magnetic field is applied along the magnetically hard $b$ axis, a reinforcement of superconductivity is observed above 15 T, which is extended up to $\mu_0 H_m \approx 35$ T. At the latter field, a first-order metamagnetic transition into a field-polarized paramagnetic phase occurs below 8 K, leading to a jump of 0.5 $\mu_B$ in the magnetization and the termination of the superconducting state [2, 3, 13]. A smaller anomaly around 6.5 T was also reported in magnetization data for $H \parallel a$ [13]. As is the case in many heavy-fermion superconductors [14], the similar energy scales of the metamagnetic transition (35 T) and the susceptibility maximum at around 40 K [3, 13] suggest a single mechanism behind both phenomena. It is likely that a Fermi-surface reconstruction as well as a volume/valence change accompanies the metamagnetic transition [15]; thermopower and Hall data show a change of the majority charge and heat carriers from electrons to holes and there is a step-like increase in the resistivity [3]. Based on the Hall data, the estimated carrier density for $H > H_m$ is around a factor of six lower than that for $H < H_m$ [16].

On rotating the magnetic field from $b$ towards $c$, the metamagnetic transition shifts upwards. Interestingly, when the field lies in a narrow angular range around the [011] direction (i.e., $\approx 23.7^\circ$ away from the $b$ axis towards the $c$ axis), transport measurements evidence that superconductivity reemerges within the field polarized paramagnetic phase above $H_m$. It is not clear whether or not this proposed high-field superconducting phase is suppressed by magnetic fields $> 60$ T [2, 7, 17]. Scenarios involving ferromagnetic fluctuations [2] and dimensionality reduction [18, 19] have been invoked to explain the high-field superconducting phase. However, the relevance of the latter mechanism is presently unclear; though the unit cell of UTe$_2$ is elongated along the $c$ direction, and ARPES data suggest the presence of quasi-one-dimensional Fermi-surface sheets due to Te bands [20], the electrical transport is rather isotropic and calculations produce a range of possible Fermi surfaces, some of which are rather three dimensional [7].

To date, one specific heat study indicates a second superconducting phase establishing the reinforcement of the critical field along the $b$ axis [21], evidence for the high-field superconducting phase have been detected us-
II. EXPERIMENTAL DETAILS

Single crystals of UTe$_2$ are grown using chemical vapor transport; the conditions are the same as for sample s4 described in Ref. [28], where further details can be found. To provide initial characterization prior to the pulsed-field experiments, heat-capacity measurements are performed using a commercial calorimeter that utilizes a quasi-adiabatic thermal relaxation technique. In addition, the electrical resistivity $\rho$ is characterized using a standard four-probe configuration with an AC resistance bridge. Resistivity (not shown) and heat-capacity measurements on crystals from this batch show a single sharp transition around 1.9 K [Fig. 1(b)].

Fig. 1(c) shows a schematic drawing of the sample environment for the pulsed-field experiments. The pancake coil for the PDO measurements (10 turns of insulated 50-gauge copper wire) is sandwiched between a G10 holder and the single-crystal UTe$_2$ sample. The sample was coated with a thin film of GE varnish to avoid electrical contact with the layers above. The MCE thermometer is an approximately 100 nm thick semiconducting AuGe film (16 at% Au) deposited directly on the varnish-coated sample to ensure good thermal coupling between sample and film. To improve the contact resistance, Au pads are deposited on the AuGe film. The AuGe film is calibrated against a commercial Cernox sensor; film resistances range from 6 $\Omega$ at room temperature to 250 $\Omega$ at 0.6 K. The sample is glued to the holder with STYCST epoxy to prevent any sample movement due to the large magnetic torque when the field is aligned close to the $b$ axis.

The simultaneous PDO measurements employ equipment similar to that described in Refs. [26, 27, 29, 32]; the technique is well established for mapping the irreversibility and upper critical fields of superconductors in pulsed magnetic fields [31, 32].

The magnetocaloric and PDO experiments were performed in the NHMFL’s mid-pulse magnet, which provides a peak magnetic field of 55 T with a rise time of approximately 30 ms and a total pulse duration of 500 ms. A typical field pulse and its derivative is shown in Fig. 1(d). The sample holder was fixed to the rotating platform of a cryogenic goniometer [33] placed within a simple $^3$He cryostat. The sample was immersed in liquid $^3$He at a bath temperature of 0.6 $\pm$ 0.1 K during the field pulses. Additionally we conducted Piezo torque magnetometry measurements in pulsed magnetic fields up to 75 T by using Membrane-type Surface-stress Sensors at the NHMFL at LANL with a high-frequency ($\approx 300$ kHz) AC.
excitation current of $\approx 500 \, \mu A$. The angular dependent torque measurements were performed at 0.7 K with the sample immersed in liquid $^3$He. In the experiments, we used a balanced Wheatstone bridge between the piezoresistive pathways. Crystals were mounted with the $b$ axis perpendicular to the cantilever plane.

III. EXPERIMENTAL RESULTS

Fig. 1(a) shows the phase diagram of UTe$_2$ as a function of magnetic field and field orientation; the angle $\theta$ describes the field rotating from parallel to the crystallographic $b$ axis ($\theta = 0^\circ$) towards the $c$ axis ($\theta = 90^\circ$). The phase diagram is based on prior magnetization, electric and thermal transport measurements [1–4, 7, 17]; the points surrounding the high-field superconducting phase (SC$_{FP}$) were taken from Ran et al. [2]. Despite sample temperature $T$ excursions of up to $\approx 1.0$ K (described in detail below), far from equilibrium with the $^3$He bath temperature ($\approx 0.6$ K), the field positions of both the high-field metamagnetic and low-field transition out of the SC$_{PM}$ phase obtained from our PDO and MCE measurements are in good agreement with prior data. In the case of the metamagnetic transition this is unsurprising, as $H_m$ is virtually temperature independent for $T < 4$ K [17].

Examples of sample temperature $T$ versus field curves for $H \parallel b$ ($\theta = 0$) and $\theta = 33^\circ$ are shown in Fig. 2(a, b) alongside data from simultaneous PDO measurements (c, d). Referring to the phase diagram [Fig. 1(a)], at sub-Kelvin temperatures and $\theta = 0$, the up-sweep of a 55 T field pulse first traverses the low-field SC$_{PM}$ phase, then the so-called re-entrant superconducting phase (SC$_{RE}$) and the metamagnetic transition at $H_m$ before finally entering the field-polarized (FP) (non-superconducting) phase. By contrast, at $\theta = 33^\circ$, a similar pulse goes through the SC$_{PM}$ phase, a metallic (non-superconducting) phase and the metamagnetic transition (shifted to higher field), where it enters the SC$_{FP}$ phase. As we will now see, these different paths across the phase diagram result in different thermal responses.

Turning first to the MCE data at $\theta = 0$ [Fig. 2(a)], as $H$ initially rises (black curve) there is a steep increase in $T$ from the $^3$He bath temperature ($\approx 0.6$ K) to $\approx 1.1$ K. This heating is attributable to an avalanche-like, dissipative vortex movement in the superconducting SC$_{PM}$ phase, a phenomenon frequently seen in pulsed-field measurements of more conventional superconductors (e.g., Ref. [31]). Thereafter, $T$ relaxes towards the bath temperature until a sharp step upwards denotes the first-order phase transition at $H_m$. Once in the FP state, $T$ again relaxes for the rest of the up-sweep and during the start of the down-sweep (red curve). However, at $H_m$ on the down-sweep there is another sharp increase in $T$, followed by further relaxation down to around 15 T; below $\approx 13$ T there is a gentle rise in $T$, again likely attributable to dissipative vortex motion in the SC$_{FP}$ phase. Note that the down-sweep of $H$ is much slower than the up-sweep, allowing more time for heat generated by vortex motion to dissipate [31].

The simultaneous PDO data at $\theta = 0$ [Fig. 2(c)] reflect these $T$ changes. As the field increases (black curve) there is a sharp fall in $f$ at about 15 T, indicating the SC$_{PM}$ to SC$_{RE}$ transition (the corresponding $T$ versus $H$ curve in (a) flattens at about the same field). The sample exits the SC$_{RE}$ phase at $\mu_0 H_m = 35$ T; once in the non-superconducting FP phase, shifts $\Delta f$ in the PDO frequency are dominated by changes in the sample resistivity $\Delta \rho$, with an approximate proportionality $\Delta f \propto -\Delta \rho$ [2, 20, 27]. At $H_m$, $\rho$ is known to exhibit a sharp increase [2], leading to a downward step in $f$. Above $H_m$, the normal-state resistivity of UTe$_2$ is rather $T$-independent in the range 0.6–2 K [2]; hence, despite the varying $T$ seen in the MCE data, the PDO frequency on the down-sweep of the field (red curve) overlies the up-sweep data. Below $H_m$, slight hysteresis between down-sweep (red) and up-sweep PDO data marks the presence of the SC$_{RE}$ phase before a step upwards (marked by an arrow) shows the transition back to the SC$_{PM}$ phase; as $T$ is lower on the down-sweep [Fig. 2(a)], this latter feature occurs at a slightly higher field than the corresponding feature in the up-sweep.

Below about 15 T, the MCE and PDO data for $\theta = 33^\circ$ [Fig. 2(b,d)], behave in a similar way to their counterparts at $\theta = 0$. However the lack of the SC$_{RE}$ phase at


\[ \theta = 33^\circ \] means that \( T \), rather than flattening, continues to fall until \( H_m \) is reached. Correspondingly, the PDO signal above 15 T at \( \theta = 33^\circ \) decreases roughly linearly, reflecting the increasing normal-state magnetoresistance, rather than flattening out as it did at \( \theta = 0 \) due to the presence of the SC\textsubscript{RE} phase. However, the biggest contrast for \( \theta = 33^\circ \) compared to \( \theta = 0 \) occurs on crossing \( H_m \), where the sample enters the SC\textsubscript{FP} superconducting phase: a significantly weaker, almost reversible, cooling effect is observed. During the down-sweep, the \( T \) change at \( H_m \) is now negative - displaying an overall cooling of the sample when exiting the SC\textsubscript{FP} state [Fig. 2(b)]. (The full angular dependence of the MCE at \( H_m \) is discussed below). Continuing along the down-sweep curves, the 33\(^\circ\) PDO data show an increase in \( f \) due to the normal-to-SC\textsubscript{PM} transition, accompanied by slight heating due to vortex motion revealed by the MCE data.

Having described the signatures of the various phase boundaries in the PDO and MCE data, we now turn to Fig. 3(a), which shows PDO frequencies for 15 angles in the range 0 \( \leq \theta \leq 48^\circ \); as before, black curves signify rising \( H \) and red curves falling \( H \). Note that the field at which the drop in \( f \) associated with the exit from the SC\textsubscript{PM} phase (either into the SC\textsubscript{RE} phase \( \theta \leq 10^\circ \)) or normal state \( \theta > 10^\circ \) occurs at lower fields on the field up-sweep due to the heating caused by dissipative vortex motion (see below); the sample is much closer to the bath temperature on the down-sweep, so that the corresponding step is at higher fields [31].

Corresponding derivatives \((1/\mu_0)(df/dH)\) of the down-sweep data are shown in Fig. 3(b). For the three lowest \( \theta \) values \((0.5^\circ, 10^\circ)\) there is only a weak, broad feature between 15 and 20 T, reflecting that the transition is between two superconducting phases (SC\textsubscript{PM} and SC\textsubscript{RE}). For \( \theta > 10^\circ \), the weak feature is replaced by a well-defined minimum, as it now corresponds to a superconductor (SC\textsubscript{PM})-to-normal transition.

The MCE measurements are summarized in Fig. 4, the increase in \( T \) in the SC\textsubscript{FP} phase around \( \theta = 33^\circ \) clearly stands out, especially in the down-sweep data. This provides thermodynamic evidence that the SC\textsubscript{FP} state observed in UTe\textsubscript{2} is indeed a bulk phase. As mentioned above, due to the large heating effect caused by vortex motion at the beginning of the magnet pulse, no clear phase boundary of the low field superconducting phase can be identified in the up-sweep MCE data. Based on the PDO data (Fig. 3), the SC\textsubscript{PM} phase is suppressed at a field of a few Tesla on the up-sweep. During the down-sweep, the phase boundary into the SC\textsubscript{PM} phase coincides with the onset of gentle sample heating below \( \approx 15 \) T and the corresponding upward step in the PDO data (Fig. 3).

At the close of this section, we again emphasize that though the corresponding features in the PDO and MCE data are weak, there are distinct indications of the boundary between the SC\textsubscript{PM} and SC\textsubscript{RE} phases. This seems to confirm that though both states are superconducting, they are distinct phases with subtly different properties [7].

**IV. DISCUSSION**

Before treating the thermodynamics of the onset of the high-field SC\textsubscript{FP} state in more detail, it is worth considering whether there is an alternative explanation for the previous (non-thermodynamic) data used to identify the apparent superconductivity of this phase.

One possibility might be a low (but nonzero) resistivity metallic phase caused by a field-induced Fermi-surface reconstruction at \( H_m \) that occurs over a restricted range of field orientations. However, existing experimental data provide a number of objections to such an interpretation.

1. As mentioned in the Introduction, Hall-effect and thermopower measurements [3] for \( H \parallel b \) indicate a very significant decrease in the charge-carrier density as one crosses \( H_m \) into the FP (normal) state, leading to a strong increase in the resistivity [2][3].

2. To counter the previous point, one might argue that a significantly different change in electrical properties (i.e., a large increase in carrier density and/or a decrease in resistivity) occurs at \( H_m \), but only over a special, restricted range of \( \theta \). In such a case, one would expect that the metamagnetic transition would also change in character for these an-
FIG. 4. Contour plot of the sample temperature $T$ as a function of the angle $\theta$ for the up-sweep (a) and down-sweep (b) of the magnetic field. The circles indicate the superconducting/ metamagnetic phase transitions discussed in the text, green triangles enveloping the high-field superconducting state were taken from Ran et al. [2]. The initial temperature before the field pulse is approximately $T_0 = (0.6 \pm 0.1)$ K, variations in $T_0$ cause vertical stripes to appear in both contour plots.

1. However, torque magnetometry data (Fig. 6) carried out over a wide range of field orientations show that the position and size of the magnetization jump at $H_m$ vary smoothly and monotonically with $\theta$.

3. An increase in the charge-carrier density at $H_m$ would lead to cooling (see [34, 35] for Ce$_3$Bi$_4$Pt$_3$ and [24] for URu$_2$Si$_2$) in the MCE during the field up-sweep and heating in the down-sweep, which is incompatible with the data in this paper.

4. The PDO data used to detect the SC$_{FP}$ state in Ref. [2] (and those in this paper) behave in a qualitatively similar manner to PDO measurements on more conventional superconductors such as pnictides [31, 32] and cuprates [36], especially in the hysteresis observed between up-sweeps and down-sweeps of the field. By contrast, PDO data measured in systems where there is a large field-induced increase in carrier density but no superconductivity [29, 30] behave in a very different way.

5. The typical energy scales associated with the transition at $H_m$ are $\sim 40$ K (see Introduction above). Any phenomenon associated with increased (normal-state) conductivity due to a Fermi-surface change at $H_m$ would be expected to persist (or slowly die away) over a temperature range similar to this. By contrast, the upper temperature limit of the SC$_{FP}$ phase is about 1.9 K [2], very similar to the critical temperatures of the SC$_{PM}$ and SC$_{RE}$ superconducting phases [7], suggesting a common or closely related origin.

In view of the above points, the following discussion of the thermodynamics occurring at and around $H_m$ assumes that the SC$_{FP}$ phase is superconducting.

As shown in a previous study [22] for $H \parallel b$, the metamagnetic transition at $H_m$ is first order at low temperatures and accompanied by hysteresis losses plus a significant release of latent heat. In the current, field-orientation-dependent study, the temperature change $\Delta T_{FP}$ observed at $H_m$ can be described as follows (see Fig. 5). (i) During the up-sweep, $\Delta T_{FP}$ is positive and decreases with increasing $\theta$ (dashed line in Fig. 5(b)) once the transition is between two normal conducting states (i.e., once we are clear of the region $\theta \leq 10^\circ$ over which the SC$_{RE}$ phase is present). (ii) $\Delta T_{FP}$ increases for $\theta$ between 25$^\circ$ and 35$^\circ$ as the sample transitions into the SC$_{FP}$ state. (iii) $\Delta T_{FP}$ decreases with $\theta$ once again when the SC$_{FP}$ state is suppressed at larger $\theta$. During the down-sweep of the field, $\Delta T_{FP}$ (Fig. 5(b), red points) is always smaller than that during the up-sweep. This is consistent with observations in Ref. [22] for $H \parallel b$. For falling field, $\Delta T_{FP}$ is positive for $\theta < 27^\circ$ and becomes negative for larger angles.

In making a quantitative description of the thermodynamics of the metamagnetic transition, we assume that the overall entropy change is a sum of reversible and irreversible processes,

$$\Delta S = \Delta S_{rev} + \Delta S_{irr} = C_p \Delta T \frac{1}{T} + \frac{\partial Q_{loss}}{T}. \quad (1)$$
FIG. 5. (a) Temperature vs. time during the up-sweep of the magnetic field pulse for $H \parallel b$. The time frame shows the metamagnetic transition and the subsequent relaxation back to the bath temperature, which is approximated by an exponential decay (red line). (b) Temperature change $\Delta T_{FP}(\theta)$ at the metamagnetic transition during the up-sweep (black triangles) and down-sweep (red circles) of the magnetic field. (c, d) Reversible and irreversible component of $\Delta T_{FP}$ as a function of the angle $\theta$ (left axes). The corresponding entropy changes are shown on the right axes of each figure.

where $\Delta S_{rev}$ describes the latent heat released during the transition, which is recovered when the field crosses $H_{m}$ in the opposite sense; and $C_{FP}$ is the heat capacity at constant pressure. The small field width of the metamagnetic transition leads us to assume adiabatic conditions and extract the temperature change $\Delta T$ directly from the magnetocaloric measurements. The time to cross the transition at $H_{m}$ is $\sim 0.6$ ms - significantly longer than the thermal relaxation timescale $\tau$ of the sample which is around 10 ms for our equipment. $\tau$ was estimated from the $T(t)$ behavior above $H_{m}$ [Fig. 5(a)]. We obtain the reversible temperature changes at the metamagnetic transition through $\Delta T_{rev} = (\Delta T_{FP\uparrow} - \Delta T_{FP\downarrow})/2$, where the subscripts “up” and “down” refer to the up- and down-sweeps of the field respectively. On the other hand, irreversible processes such as Joule heating contribute to the temperature change in both field-sweep directions, therefore $\Delta T_{irr} = (\Delta T_{FP\uparrow} + \Delta T_{FP\downarrow})/2$.

Using the fact that $C_{p}/T \approx 250$ mJmol$^{-1}$K$^{-2}$ and assuming that $C_{p}$ is nearly temperature independent below 2 K at 35 T [22], for $\theta < 25^\circ$ we obtain an almost constant value, $\Delta S_{rev} \approx 30$ mJmol$^{-1}$K$^{-1}$. Within the SC$_{FP}$ phase, $\Delta S_{rev}$ increases, peaking at $\Delta S_{rev} \approx 80$ mJmol$^{-1}$K$^{-1}$ close to $\theta = 35^\circ$ [Fig. 5(c)] [37]. Therefore, entering the SC$_{FP}$ phase releases an additional $\approx 50$ mJmol$^{-1}$K$^{-1}$ in latent heat. Assuming (as justified above) that the SC$_{FP}$ represents a field-induced superconducting state, the additional latent heat is likely to result from the formation of a gap at the Fermi energy and an entropy reduction due to pair condensation [7].

The irreversible component $\Delta S_{irr}$ mainly consists of hysteretic losses during the first-order metamagnetic transition and, bearing in mind the similarity of the behaviour of the PDO data in the SC$_{FP}$ state to that in the SC$_{PM}$ phase (see also Ref. [2]), what is likely to be dissipation due to vortex movement. As shown in Fig. 5(d), rotating $H$ to higher $\theta$ leads to an overall decrease in $\Delta S_{irr}$, apart from a local maximum around $\theta = 35^\circ$. As this is roughly in the middle of the $\theta$ range over which the SC$_{FP}$ phase occurs, it possibly coincides with the strongest vortex pinning and thus the largest dissipation caused by vortex motion. Note that while $H_{m}$ increases with increasing $\theta$, the jump in the magnetization at $H_{m}$ at 1.4 K does not change significantly between $H \parallel b$ and $H \parallel [011]$ [23]. Torque measurements shown in Fig. 6 also vary smoothly as a function of angle. Therefore it is unlikely that the additional latent heat released when entering the SC$_{FP}$ phase is of magnetic origin; rather it is probably related to its electronic and/or superconducting properties.

Finally, we remark that the boundaries between the various low-temperature/high-magnetic-field phases of UTe$_{2}$ derived in this work from PDO and MCE data match those in the literature [2, 7] very closely. This is of interest because the zero- or low-field behaviour of UTe$_{2}$ seems very sensitive to the source, growth method, and quality of the crystals used (an excellent summary is given in Ref. [7]). The present study employs crystals from completely different sources to those used to produce the phase diagrams reported in Refs. [2, 7], perhaps suggesting that the high-field properties of UTe$_{2}$ are in
some sense less sensitive to sample dependent disorder than those in zero or small magnetic fields.

V. SUMMARY

Simultaneous magnetocaloric effect and MHz conductivity measurements are carried out on a single crystal of UTe$_2$ as a function of magnetic field orientation, using pulsed magnetic fields of up to 55 T. A pronounced heating effect is observed close to the metamagnetic transition into the high-field SC$_{FP}$ phase. This strongly suggests vortex movement in a bulk superconducting state, giving thermodynamic evidence that the SC$_{FP}$ state represents a field-stabilized bulk superconducting phase of UTe$_2$, an interpretation that is supported by phase diagrams proposed from previous electrical transport measurements. With the field aligned close to the $b$ axis, a more subtle feature is observed around 15 T, suggesting that the superconducting SC$_{PM}$ and SC$_{RE}$ states represent separate, distinct phases.

ACKNOWLEDGEMENTS

We thank Minseong Lee for helpful discussions. A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by the NSF Cooperative Agreement No. DMR-1644779, the U.S. DOE and the State of Florida. This material is based upon work supported by the U.S. Department of Energy, Office of Science, National Quantum Information Science Research Centers. P.F.S.R. acknowledges support from the Los Alamos Laboratory Directed Research and Development program through project 20210064DR. J.S. thanks the DoE BES FWP “Science of 100 T” for support in developing techniques used in these experiments. R.S., Y. L. and M.J. acknowledge support by the NHMFL UCGP program and the G. T. Seaborg Institute Postdoctoral Fellow Program under project number 20210527CR.

APENDIX A: COMPLETE ANGULAR DEPENDENT MAGNETOCALORIC DATA SET

Here we show the entire angular dependent magnetocaloric data set (Fig. 6) measured with the sample immersed in liquid $^3$He. The data was used to generate the contour plots shown in Fig. 7.
FIG. 7. Sample temperature vs. magnetic field for different angles denoted in the graphs, where $0^\circ$ is $H \parallel b$ and $90^\circ$ is $H \parallel c$. Field up- and down-sweep data are depicted as black and red lines respectively.

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