

**Anomalous anisotropy of the lower critical field and Meissner effect in UTe<sub>2</sub>**C. Paulsen,<sup>1,\*</sup> G. Knebel<sup>2</sup>, G. Lapertot,<sup>2</sup> D. Braithwaite,<sup>2</sup> A. Pourret,<sup>2</sup> D. Aoki,<sup>2,3</sup> F. Hardy,<sup>4</sup> J. Flouquet,<sup>2</sup> and J.-P. Brison<sup>2</sup><sup>1</sup>Université Grenoble Alpes, Institut Néel, CNRS BP 166, F-38042 Grenoble France<sup>2</sup>Université Grenoble Alpes, CEA, Grenoble INP, IRIG, PHELIQS, F-38000 Grenoble, France<sup>3</sup>Institute for Materials Research, Tohoku University, Ibaraki 311-1313, Japan<sup>4</sup>Institute for Solid-State Physics, Karlsruhe Institute of Technology, D-76021 Karlsruhe, Germany

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We report on low-temperature susceptibility and magnetization measurements made on single crystals of the recently discovered heavy-fermion superconductor UTe<sub>2</sub> and compare the results with the two ambient pressure ferromagnetic superconductors URhGe and UCoGe. Hysteresis curves in the superconducting phase show a familiar diamond shape superimposed on a large paramagnetic background. The Meissner state was measured by zero-field cooling in small fields of a few Oe as well as ac susceptibility measurements in small fields and resulted in 100% shielding, with a sharp transition. However, the field-cooling Meissner-Ochsenfeld effect (expulsion of flux) was negligible in fields greater than just a few Oe, but becomes nearly 30% of the perfect diamagnetic signal when the field was reduced to 0.01 Oe. The critical current due to flux pinning was studied by ac susceptibility techniques. Over the range in fields and temperature of this study, no signature of a ferromagnetic transition could be discerned. The lower critical field  $H_{c1}$  has been measured along the three crystallographic axes, and surprisingly, the anisotropy of  $H_{c1}$  contradicts that of the upper critical field. We discuss this discrepancy and show that it may provide additional support for a magnetic field-dependent pairing mediated by ferromagnetic fluctuations in UTe<sub>2</sub>.

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Spin-triplet superconductivity (SC) in itinerant ferromagnets close to the ferromagnetic (FM)-paramagnetic (PM) instability was proposed four decades ago [1]. The discovery of the coexistence of ferromagnetism and SC in UGe<sub>2</sub> opened the “rush” to a large variety of experiments [2]. The strong first-order nature of the FM-PM transition under pressure at  $p_c \approx 1.6$  GPa leads to SC occurring only in the FM domain in the pressure range from 1.2 to 1.6 GPa; the maximum of the superconducting temperature  $T_c$  is 0.8 K, but the Curie temperature  $T_{\text{Curie}} \approx 30$  K [3].

The field was enriched by the discoveries of two ambient pressure superconducting ferromagnets, URhGe [4] and UCoGe [5], with  $T_c = 0.25$  and 0.8 K, much lower than the respective  $T_{\text{Curie}} = 9.5$  and 2.7 K. The rapid suppression of  $T_{\text{Curie}}$  in UCoGe with pressure leads to the PM ground state above 1 GPa with the persistence of SC far above the critical pressure [6]. For both systems, the weakness of the FM interaction means that transverse magnetic fields ( $H$ ) applied along the  $b$  axis, perpendicular to the easy magnetization axis  $c$ , of these orthorhombic crystals give rise to a spectacular field enhancement of SC [3,7,8].

The recent observation of SC in orthorhombic UTe<sub>2</sub> [9,10] at  $T_c = 1.6$  K opens the possibility to study at ambient pressure spin-triplet SC in a system with a PM ground state located very close to a PM-FM instability. UTe<sub>2</sub> has the highest susceptibility [11] and strong magnetic fluctuations [12] along the  $a$  axis. However, the transverse field configuration with  $H \parallel b$  attracted the most attention, due to the observation of a

strong field-induced reinforcement of SC on approaching the metamagnetic field  $H_m \approx 35$  T [13–16]. Most of the published magnetization data in FM SC investigate the field dependence of the FM interaction by longitudinal or transverse field variation [17–19].

In URhGe and UCoGe the respective FM sublattice magnetization  $M_0 = 0.4\mu_B$  and  $0.07\mu_B$  per U atom produces an internal field of 800 and 100 G far higher than the estimated value of the lower superconducting critical field  $H_{c1}$  of a few gauss. Thus even at  $H = 0$ , self-induced vortices should occur, as shown for example in the magnetization studies on UCoGe [20].

In this Letter we report low-temperature susceptibility and magnetization measurements on two crystals of UTe<sub>2</sub> ( $T_c = 1.5$  and 1.6 K). The experiments concentrate on (i) the persistence of the PM state well below  $T_c$  K, (ii) the strength of the Meissner effect in field-cooled (FC) experiments, (iii) the proof of a complete superconducting screening in zero-field-cooled (ZFC) magnetization measurements, (iv) the determination of  $H_{c1}$ , and (v) the determination of the London penetration depth and of the superconducting coherence length from  $H_{c1}$  and from the upper critical field  $H_{c2}$ . We compare the results with the FM superconductors URhGe and UCoGe [21].

All the measurements were made using two low-temperature superconducting quantum interference device (SQUID) magnetometers developed at the Institut Néel in Grenoble. A unique feature of the setup is that absolute values of the magnetization can be measured using the extraction method in a field range from 0.01 Oe up to 8 T (for details, see the Supplemental Material [21]).

\* carley.paulsen@neel.cnrs.fr

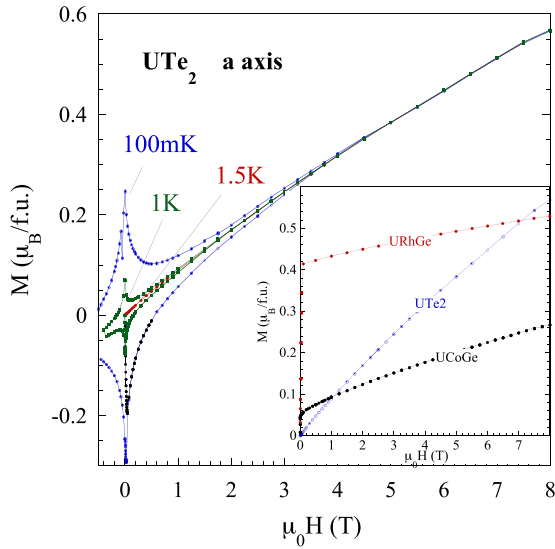


FIG. 1. Magnetization vs field for  $UTe_2$  (sample 1) at 100 mK and 1 K below  $T_c$ , and at 1.5 K just above  $T_c$  in the normal state with the field along the  $a$  axis (easy axis). The inset shows the magnetization of  $UTe_2$  (blue) at 1.5 K,  $URhGe$  (red) at 500 mK, and  $UCoGe$  (black) at 600 mK, i.e., just above their respective  $T_c$  in the normal state, with the field along the easy axis.

Figure 1 shows hysteresis loops for  $UTe_2$  measured at 100 mK and 1 K below  $T_c$ , and at 1.5 K in the normal phase for  $UTe_2$  with the field direction along the easy magnetization  $a$  axis. The slope of the initial magnetization [also shown later more clearly in Fig. 4(a)] corresponds to 100% shielding. The inset of Fig. 1 compares  $UTe_2$  at 1.5 K to the two FM superconductors  $URhGe$  and  $UCoGe$ , with the field applied along their easy  $c$  axis, at 500 and 600 mK above their respective superconducting states, but far below their respective Curie temperatures. The spontaneous moment of both ferromagnetic superconductors appears clearly, however, it is not a saturated moment:  $M(H)$  keeps growing with increasing field. Although there is no spontaneous moment for  $UTe_2$ , the PM magnetization increases quickly and becomes larger than in  $UCoGe$  at about 1 T, and then greater than in  $URhGe$  above 7 T.

The hysteresis loops for  $UTe_2$  have a familiar superconducting diamond shape which is superimposed on a very large PM background response. More hysteresis loops taken close to  $T_c$  are shown in Fig. S5 in the Supplemental Material, as well as a comparison with  $UCoGe$  in Figs. S6 and S7. In contrast to  $UCoGe$  where the superconducting and FM signals are fused together with the FM response dominating [20], our measurements of  $UTe_2$  over the full temperature and field range show that there is no hint of FM behavior down to 80 mK, in agreement with muon spin rotation ( $4\mu SR$ ) experiments [22].

In Fig. 2(a) the dc susceptibility  $M/H$  is plotted against temperature for various applied fields ranging from 0.01 to 200 Oe. Each curve was made by first zero-field cooling (ZFC) the sample. A dc field was then applied and the sample was slowly warmed above  $T_c$ , after which it was recooled in the same field, giving the field-cooled (FC) curve. In small dc fields the value of the ZFC susceptibility corresponds to

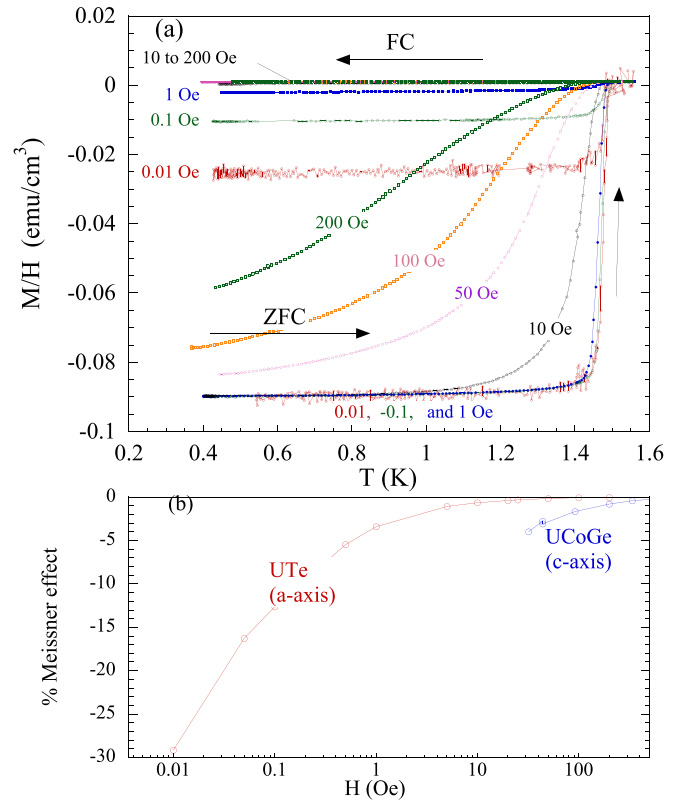


FIG. 2. (a)  $M/H$  vs  $T$  of  $UTe_2$  (sample 1) for various applied fields ranging from 0.01 to 200 Oe from zero-field-cooled (ZFC) and field-cooled (FC) measurements. (b) The percent Meissner-Ochsenfeld effect (expulsion of flux) plotted against the applied field for  $UTe_2$  (red points) and for  $UCoGe$  (blue points). Note that the field range in  $UCoGe$  cannot extend below 100 mT as the sample needs to be monodomain. Over the comparable field range, the expulsion of flux is much greater in  $UCoGe$ .

100% shielding of the field (when demagnetization corrections are made), and the transition is sharp. As the fields are increased, the transition becomes broader and shifts to lower temperatures. The FC susceptibility shows that the Meissner-Ochsenfeld effect (the reversible expulsion of flux as the sample is field-cooled and warmed through  $T_c$ ) for fields greater than a few Oe is negligible. However, for very small fields, the effect becomes more important, reaching about 30% expulsion in a field of 0.01 Oe.

We compare this last result to  $UCoGe$  along the easy axis in Fig. 2(b). There are important differences. First, the internal fields that are present in  $UCoGe$ , of the order 50–100 G, are much greater than  $H_{c1}$ , and as a result  $UCoGe$  is always in the mixed state, and never achieves 100% shielding. In addition, to measure the Meissner-Ochsenfeld effect in  $UCoGe$  means taking into account hysteresis and a coercive field such that the applied field has no meaning while the sample is multidomain [20]. Nevertheless, a typical value of the percent expulsion of the flux from  $UCoGe$  compared to its effective shielding would be about 3% expulsion at 50 Oe, which decreases with increasing field. Although small, this is much greater than the Meissner-Ochsenfeld effect observed in  $UTe_2$  in the same field range as can be seen in Fig. 2(b). The other remarkable feature observed in Fig. 2(b) is the nonsaturating rate of

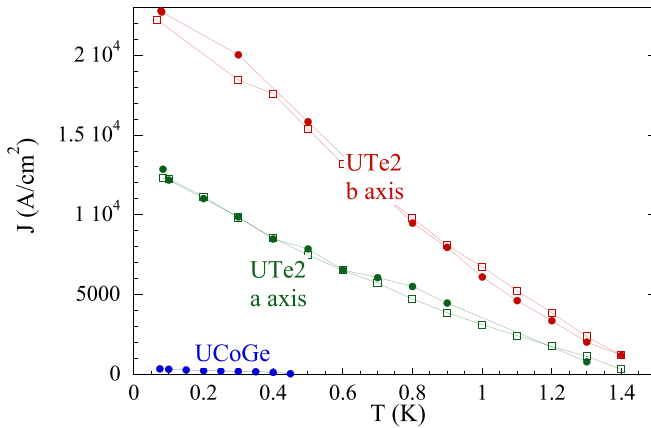


FIG. 3. Current density  $J_c$  derived from the critical state model vs temperature for  $\text{UTe}_2$  (sample 2) measured along the  $a$  and  $b$  axes, and for  $\text{UCoGe}$  along the  $c$  axis. The solid round points are data derived from the imaginary part of ac susceptibility, and the open square points are data derived from the remanent magnetization (discussed in the Supplemental Material [21]).

increase of the Meissner effect down to fields as low as 0.01 Oe in  $\text{UTe}_2$ : If any internal field exists due to a weak FM phase inside the superconducting phase, the resulting dipolar field has to be much smaller than 0.01 G, or in other words, the ordered moment should be much smaller than  $7 \times 10^{-6} \mu_B$ .

A strong hysteresis and a weak Meissner-Ochsenfeld effect suggests strong flux pinning. To confirm this we measured the ac susceptibility  $\chi$  as a function of the ac driving field: An example can be seen in the Supplemental Material Fig. S20 for the  $b$  axis. When flux begins to enter the sample,  $\chi'$  and  $\chi''$  of the ac susceptibility will deviate from their 100% shielding values. The deviations are linear in the applied driving field and the slopes are proportional to  $2/(J_c D)$  for  $\chi'$ , and  $2/(3\pi J_c D)$  for  $\chi''$ , where  $J_c$  is the current density in the critical state model, and  $D$  is the sample width, where we approximate the sample shapes as slabs [23]. The resulting  $J_c(H)$  for  $\text{UTe}_2$  is plotted in Fig. 3, along with  $J_c$  for  $\text{UCoGe}$  measured along the  $c$  axis. Clearly, flux pinning is far greater in  $\text{UTe}_2$ .

The initial magnetization  $M$  vs  $H$  taken at various constant temperatures is shown in Fig. 4(a). For each curve, the sample was first ZFC. The blue dashed line is a linear fit to the 100 mK data over a field range 0–10 Oe. The slope of this fit (when corrected for demagnetization effects) corresponds to a susceptibility of  $-1/4\pi$  ( $-1$  in SI units) or 100% shielding of the magnetic field. For a given temperature, as the field is increased, the curves deviate from this slope, and this is an indication that flux is entering the sample because  $H_{c1}$  has been exceeded. As can be seen in Fig. 4(a), due to the strong pinning, flux enters the sample almost asymptotically and makes the determination of  $H_{c1}$  difficult. In addition, because the samples used in this study are not perfect ellipsoids, the field enters the samples sooner around the edges due to demagnetization effects, further obscuring the real  $H_{c1}$ .

A better way to determine  $H_{c1}$  is to measure the remanent magnetization as shown in Figs. 4(b) and 4(c) and discussed in detail in the Supplemental Material. Figure 4(b) is an example of a series of minor hysteresis loops at 1.1 K for sample 1

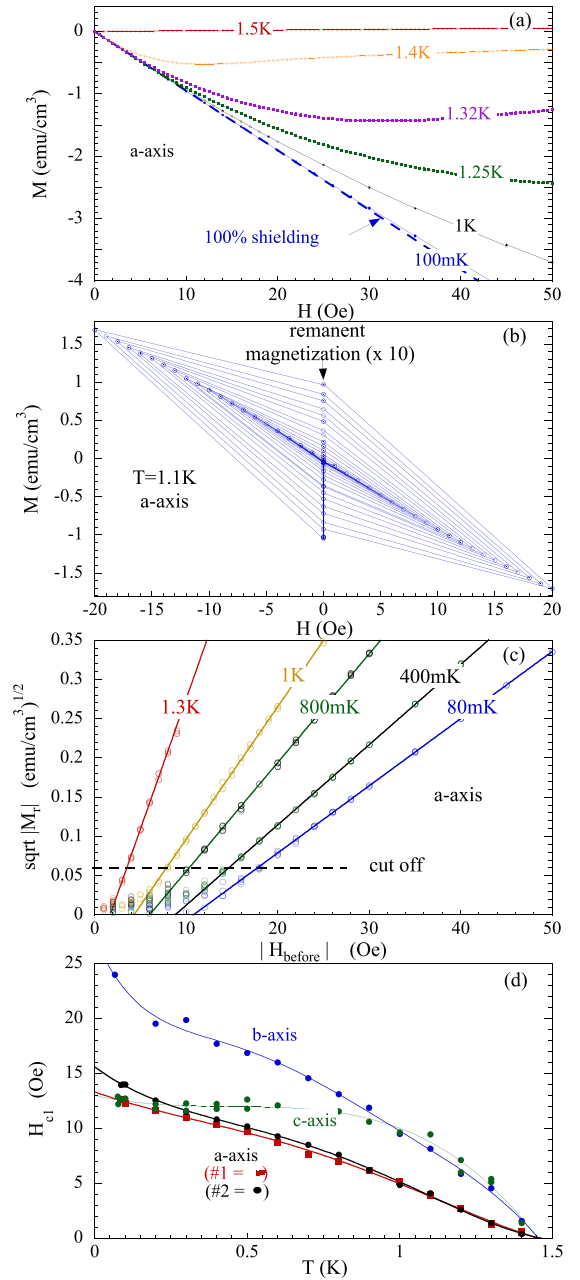


FIG. 4. (a) Initial magnetization  $M$  vs  $H$  taken at various constant temperatures for  $\text{UTe}_2$  (sample 1) with the field along the  $a$  axis. The dashed line is a linear fit to the 100 mK data at low fields, and represents 100% shielding. (b) Series of minor hysteresis loops for sample 1 at 1.1 K where the magnitude of the field was systematically increased in steps of 1 Oe, and then returned to zero to measure the point at which a remanent magnetization begins to appear in the sample (the remanent magnetization has been multiplied by 10). (c) The remanent magnetization is plotted as  $\sqrt{M_r}$  vs  $H_{\text{before}}$  for the  $a$  axis for sample 1. The solid lines are linear fits to the data above the cutoff line. (d)  $H_{c1}(T)$  for  $\text{UTe}_2$  along the  $a$  axis for sample 1 (red) and for sample 2 along the  $a$  (black),  $b$  (blue), and  $c$  (green) axis after correcting for demagnetization effects. The lines are guides to the eye.

where the magnitude of the field was systematically increased in small steps, and then returned to zero to measure the point where flux begins to enter the sample. While  $H < H_{c1}(T)$

the cycles are reversible. However, when  $H_{c1}(T)$  is exceeded, flux begins to enter the sample and the magnetization deviates from the 100% shielding. When the field is then returned to zero, half the flux remains trapped in the sample, and a remanent moment  $M_r$  appears. According to the Bean critical state model  $M_r \propto H_{\text{before}}^2$ , i.e., the last field value before the field was reduced to zero and the remanence was measured [24]. In order to estimate  $H_{c1}$ , we modify the critical state model following [25] by assuming two regimes: when  $H_{\text{before}} < H_{c1}$ ,  $M_r = 0$  and when  $H_{\text{before}} > H_{c1}$ ,  $M_r \propto (H_{\text{before}} - H_{c1})^2$ .

In Fig. 4(c) we plot an example of the remanent magnetizations for sample 1 as  $\sqrt{M_r}$  vs  $H_{\text{before}}$ . The solid lines are linear fits to the high-field data above the cutoff line, thus omitting the low-field values where we expect rounding to occur. The intercept gives  $H_{c1}$ .

In Fig. 4(d)  $H_{c1}$  along the  $a$  axis is plotted versus temperature for sample 1, and for sample 2 for the  $a$ ,  $b$ , and  $c$  axes, where  $H_{c1}$  has been corrected for demagnetization effects: Both samples have a platelet shape, with the field applied parallel to the platelet, except for the  $c$ -axis measurements which was performed perpendicular to the surface. We estimate the value of  $H_{c1}$  at  $T = 0$  to be 14 Oe along the  $a$  and  $c$  axis, and 24 Oe along the  $b$  axis.

We have also studied  $H_{c2}$  along the  $a$  axis using bulk ac susceptibility as shown in Fig. S21 of the Supplemental Material and we found  $H_{c2}(0) = 5.4$  T along the  $a$  axis, in good agreement with published resistivity measurements [9,10].

However, a comparison of  $H_{c1}$  and  $H_{c2}$  data reveals a major inconsistency. Indeed, close to  $T_c$  in the Ginzburg-Landau (GL) regime, the anisotropy of  $H_{c1}$  should be opposite to that of  $H_{c2}$  as  $H_{c1}H_{c2} = H_c^2(\ln \kappa + 0.49)$ , where  $H_c$  is the (isotropic) thermodynamic critical field and  $\kappa = \frac{\lambda}{\sqrt{2\xi}}$  the anisotropic GL parameter. Hence, neglecting the anisotropy of  $\kappa$  due to the logarithm, with  $H_{c2}^b > H_{c2}^a \approx H_{c2}^c$ , one expects  $H_{c1}^b < H_{c1}^a \approx H_{c1}^c$ . In contrast, Fig. 4(c) shows that the relative anisotropies anticipated from  $H_{c2}$  are wrong in all directions. Quantitatively, this is a major effect, as the measured  $H_{c1}$  anisotropy between  $a$  and  $(b, c)$  is a factor 3, whereas the  $H_{c2}$  anisotropy between  $(a, c)$  and  $b$  is between 3 and 5, depending on the measurements notably for  $H \parallel b$  (see, e.g., Ref. [10] [15,16]).

Such a large discrepancy calls for an explanation. Measurement errors of  $H_{c1}$  for  $H \parallel b$  might come from stronger pinning in this direction. The critical current has been found indeed twice as large for  $H \parallel a$  [see Fig. 3(b)], but it is very unlikely that it could explain a factor of 10 error between the two directions. The sample geometries are also similar in both cases, excluding an explanation through a bad estimation of the demagnetization corrections.

So the next step is to question the estimate of  $H_{c1}$  from the relations with the GL parameter: For single-band  $s$ -wave superconductors, these relations hold even in very anisotropic cases (see, e.g., Ref. [26]). However,  $\text{UTe}_2$  is most likely  $p$ -wave, multigap (as most other heavy-fermion superconductors), and topological [27]. The last feature, implying the existence of low-energy surface states might influence pinning, but if it has any influence on the determination of  $H_{c1}$ , it should also be reflected in the critical current measurements. More interestingly, the multigap character [or the nodal gap

structure [28] for one-dimensional (1D) irreducible representations] has been shown, theoretically (e.g., Ref. [29]) and experimentally (e.g., Ref. [30]) to induce strong deviations of the anisotropy of the critical fields from the estimations through the GL parameter: But this holds at low temperature, not close to  $T_c$ .

Last, several works have reported double transitions in their  $\text{UTe}_2$  crystals at zero pressure [31,32]. Note, however, that  $H_{c1}$  and  $H_{c2}$  measurements are only sensitive to the upper transition, until both transitions eventually cross under field, which does not happen below 8 T according to Ref. [32]. Hence, whatever the origin of this double transition (intrinsic or due to inhomogeneities), it cannot help to understand the reported anomalous relative anisotropies of  $H_{c1}$  and  $H_{c2}$ .

A possible explanation for the puzzling anisotropy of  $H_{c1}$ , which cannot be completely ruled out, is a low-field change of slope of  $H_{c2}$ . This is suggested for example by specific heat measurements [33], which find a weak anisotropy between  $a$  and  $b$  axis due to a strong curvature of  $H_{c2}$  along  $a$  (hence a much larger slope at low fields), but still a smaller slope along the  $c$  axis. This may partially help to reduce the discrepancy between  $a$  and  $b$  anisotropies of  $H_{c1}$  and  $H_{c2}$ , but is still not enough to explain the  $H_{c1}$  result, requiring  $H_{c2}^a \gg H_{c2}^b \sim H_{c2}^c$ .

A key to understanding this anomalous anisotropy might be to take into account that the pairing mechanism in  $\text{UTe}_2$  can be tuned by a magnetic field [9,15], a mechanism inducing a change of slopes of  $H_{c2}$  [18,19], which is not taken into account by GL. A field-dependent pairing induces a field dependence of the “bare” critical temperature  $T_c(H)$  that is independent of the mixed state formation, as well as of the Fermi velocities ( $v_F$ ) [19]. Hence, with field-dependent pairing, both critical fields are functions of field (through  $T_c$  and  $v_F$ ) and temperature and it is easy to show that the measured slope at  $T_{sc}$  is  $\frac{dH_{c1}}{dT} = \frac{(\frac{\partial H_{c1}}{\partial T})_H}{1 + \frac{dT_{sc}}{dH} (\frac{\partial H_{c1}}{\partial T})_H}$ . Here,  $(\frac{\partial H_{c1}}{\partial T})_H$  is the “usual” slope defined for field-independent pairing, for which GL relations should hold. This relation between the measured  $\frac{dH_{c1}}{dT}$  and  $\frac{\partial H_{c1}}{\partial T}$  shows that, because  $H_{c1}$  is three to four orders of magnitudes smaller than  $H_{c2}$ , corrections due to  $\frac{dT_{sc}}{dH}$  should be negligible on  $H_{c1}$ , which should reflect the true bare anisotropies. A  $\frac{dT_{sc}}{dH} \approx -0.1$  T/K for  $H \parallel a$  and  $+0.1$  T/K for  $H \parallel b$ , (assuming  $\frac{dT_{sc}}{dH} \approx 0$  for  $H \parallel c$ ) could reconcile the lower and upper critical field measurements (see Supplemental Material for more details [21]).

To summarize, the surprising contradiction between lower and upper critical field anisotropies can be understood as a manifestation of the strong field dependence of the pairing strength in  $\text{UTe}_2$ , which is strongly suppressed along the easy axis and boosted along the hard  $b$  axis. Moreover, this is an indication that pairing is suppressed by a field along the easy axis in this system, an effect comparable to, although weaker than, in  $\text{UCoGe}$  [18,19], which can be seen as additional support for ferromagnetic fluctuation mediated pairing.

Moreover, from our very low-field measurements of the Meissner state, we can put an upper limit to any FM ordered moment above 100 mK of  $7 \times 10^{-6} \mu_B$  in  $\text{UTe}_2$ . Restricted Meissner-Ochsenfeld expulsion is coherent with the observed strong pinning. A possible link between the present strong

pinning and singular topological properties of the superconducting phase deserve to be clarified.

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