Expansion of the high field-boosted superconductivity in UTe$_2$ under pressure

Sheng Ran $^{1,2,3}$, Shanta R. Saha $^{1,2}$, I-Lin Liu $^{1,2}$, David Graf $^4$, Johnpierre Paglione $^{1,2}$ and Nicholas P. Butch $^{1,2,5}$

Magnetic field-induced superconductivity is a fascinating quantum phenomenon, whose origin is yet to be fully understood. The recently discovered spin-triplet superconductor, UTe$_2$, exhibits two such superconducting phases, with the second one reentering in the magnetic field of 45 T and persisting up to 65 T. More surprisingly, in order to induce this superconducting phase, the magnetic field has to be applied in a special angle range, not along any high symmetry crystalline direction. Here we investigated the evolution of this high-field-induced superconducting phase under pressure. Two superconducting phases merge together under pressure, and the zero resistance persists up to 45 T, the field limit of the current study. We also reveal that the high-field-induced superconducting phase is completely decoupled from the first-order field-polarized phase transition, different from the previously known example of field-induced superconductivity in URhGe, indicating superconductivity boosted by a different paring mechanism.

npj Quantum Materials (2021) 6:75; https://doi.org/10.1038/s41535-021-00376-9

INTRODUCTION

The recent evidence for spin-triplet superconductivity in UTe$_2$ has opened an avenue for the study of topological superconductivity$^1$. The superconducting state of UTe$_2$ closely resembles that of ferromagnetic superconductors, but the normal state is paramagnetic and shows no indication of magnetic ordering$^{2-4}$. Spin-triplet pairing is strongly indicated by the extremely large, anisotropic upper critical field $H_{c2}$, nodes on the superconducting gap$^6$, and the temperature-independent NMR Knight shift in the superconducting state$^8$. The superconducting order parameter comprises two components and breaks time-reversal symmetry$^{10,11}$. A nontrivial topology is suggested by the observation of chiral in-gap bound states by scanning tunneling microscopy$^{12}$.

Even more striking, UTe$_2$ hosts two independent field-induced superconducting phases$^{13-16}$, with the higher-field phase reentering at a high magnetic field of 45 T and persisting up to 65 T when the magnetic field is aligned over a limited angular range about the normal direction of the (011) plane. The quasi-two-dimensional Fermi surface revealed by band structure calculations and photoemission measurements$^{17-19}$, as well as the lack of a ferromagnetic ground state, has led to suggestions that the field-induced superconductivity in UTe$_2$ is due to reduced dimensionality instead of magnetic fluctuations$^{20,21}$: a magnetic field applied parallel to quasi 2D conducting layers will stabilize superconductivity when the magnetic energy reaches the hopping amplitude between the conducting layers$^{20,22}$.

In this work, we investigate the evolution of the magnetic field-induced superconducting phases in UTe$_2$ as pressure is applied to samples oriented specifically along the off-axis angle which stabilizes the high-field phase. Over a range of pressures near 1 GPa, the two different superconducting phases merge together, and the electrical resistance remains zero up to at least 45 T, a remarkably large value for a superconductor with a 3 K critical temperature. The high-field-induced superconducting phase is completely decoupled from the first-order transition to a field-polarized state, suggesting that magnetic fluctuations may not be crucial to this reentrant superconductivity. At pressures exceeding the critical pressure at which metamagnetic phase transition extrapolates to zero magnetic field, we observe features with the same temperature dependence as the high-field-induced superconducting phase, further investigation of which might shed light on the mechanism reentrant superconductivity.

RESULTS AND DISCUSSION

Pressure-magnetic field phase diagram

To characterize the evolution of the high-field-induced superconducting phase under pressure, we performed complementary measurements of electrical resistance $R$ and tunnel diode oscillator (TDO) frequency $\Delta f$, which is sensitive to the change of both electrical resistance and magnetic susceptibility. Two samples were studied for which the magnetic field was applied $\sim 25^\circ$ and $30^\circ$, respectively, away from the $b$ axis toward c. At ambient pressure, three distinct phases were observed as shown in Fig. 1: a field-polarized state FP with greatly enhanced magnetization and resistance$^{13,15,16,23}$, the low-field superconducting phase, SC$_{FP}$, coexisting with the paramagnetic state; and the high-field-induced superconducting phase, SC$_{FP}$, existing inside the field-polarized state. The criteria used to infer critical magnetic field for each phase are explained in the Supplementary Information (see Supplementary Figs. 4–7).

Absent in this field configuration is another field-induced superconducting phase that is observed on the low-field side of the metamagnetic field $H_{FP}$ when the magnetic field is applied along the $b$ axis. For magnetic field along the $b$ axis, applied pressure suppresses the metamagnetic field to zero at 1.5 GPa and forces a phase transition inside the paramagnetic state at a crossover pressure $P_{c2}$ from SC$_{PM}$ to SC$_{PM2}$, in zero magnetic field at $\sim 1$ GPa$^{24}$. Recent symmetry analysis indicates that the SC$_{PM1}$
The stability of superconducting and magnetic phases of UTe$_2$. a The zero-field superconducting phase SC$_{PM1}$ exhibits a strongly direction-dependent upper critical field $H_{C2}$. Along the crystallographic b axis, the reentrant superconducting phase SC$_{PM2}$ is stabilized up to a first-order magnetic transition at $H_{FP}$, above which exists a magnetically polarized phase FP. Applied pressure uniformly suppresses all field scales and $H_{C2}$ is no longer visible in the electrical resistance. For a magnetic field oriented between 25° and 30° between the b- and c-axes, the high-field reentrant superconductivity SC$_{FP}$ stabilizes in the FP phase. Applied pressure enhances SC$_{PM2}$, which meets a decreasing $H_{FP}$ at a crossover region $P_c$. Here, SC$_{PM1}$ transitions to SC$_{PM2}$, which survives up to $P_c$, where it is replaced by magnetic order M. Below $P_c$, SC$_{FP}$ is a lower bound on SC$_{FP}$, but above $P_c$, the two are decoupled and SC$_{FP}$ survives beyond $P_c$. An anomaly $H_{FP}$, suggestive of Landau-level superconductivity, emerges above $P_c$. The phase diagram is based on the resistance data shown in Fig. 2.

A subtle difference between the electrical resistance and TDO samples highlights the effects of their slight angular offset (Fig. 2). As the TDO sample sits at a slightly larger angle away from the b-axis, the upper critical field of SC$_{PM}$ has a smaller value and the critical onset field of SC$_{FP}$ has a larger value at ambient pressure. The SC$_{PM}$ and SC$_{FP}$ phases remain connected over a much smaller pressure range. Similarly, the field-polarized state starts above a higher magnetic field because the metamagnetic field is larger, but it is suppressed faster upon increasing pressure and vanishes also at $P_c$.

The discontinuous nature of the metamagnetic transition at $H_{FP}$ is conspicuous in the resistance data (Fig. 3). At low pressures, a sharp upward jump marking the FP phase boundary is replaced at low temperatures by a sharp downward jump marking the SC$_{FP}$ phase boundary. An additional hallmark of first-order transitions, namely field-hysteresis is also readily apparent. In the intermediate pressure range, $H_{FP}$ limits the lower-field superconducting phase SC$_{PM}$. Here, the transitions associated with $H_{FP}$ are again hysteretic. Interestingly, the decoupled SC$_{FP}$ phase appears to also exhibit some small hysteresis, the origin of which may be associated with details of the field reentrance. These features are consistent at all measured pressures.

These details reveal important points about the relationships between the many electronic phases. The low- and high-field superconducting phases always exist on opposite sides of the metamagnetic transition $H_{FP}$, upon which Fermi surface reconstruction has been suggested based on thermoelectric power and Hall effect measurements. In addition, in the low-pressure range, SC$_{FP}$ only appears on the high-field side of the metamagnetic field (Fig. 2a, b), while SC$_{PM}$ only exists at fields lower than the metamagnetic field. The role of the metamagnetic field switches in the intermediate pressure range, where the metamagnetic field truncates the low-field phase SC$_{PM}$. This behavior is apparent in both panels of Fig. 2c, where the high-temperature part of the upper critical field of SC$_{PM}$ curve rises rapidly as temperature decreases, extrapolating to field values far higher than the metamagnetic field, but then, as soon as the upper critical field of SC$_{PM}$ coincides with the metamagnetic field, the behavior suddenly changes and the upper critical field of SC$_{PM}$ becomes almost temperature-independent down to low temperatures. Taken together, these facts imply that the SC$_{PM}$ and SC$_{FP}$ phases separated by the metamagnetic transition might have different superconducting pairings that are unique to PM and FP, respectively. It is particularly striking that both low- and high-field superconducting phases look like they would cover much larger ranges of the field were they not limited by the metamagnetic field.
In the high-pressure range, the critical onset field of SC$_{FP}$ and the metamagnetic phases are well separated, by more than 20 T. This is crucial for the understanding of the pairing mechanism of the SC$_{FP}$ phase. In the case of URhGe, the reentrance of superconductivity is crucial for the understanding of the pairing mechanism of the SC$_{FP}$ phases. These anomalies, denoted as $A_{FP}$, are pronounced in the $\Delta f$ data, and noticeable in $R$ data (Fig. 4). It is not clear whether these anomalies correspond to a thermodynamically distinct phase. In order to trace the evolution of these anomalies, we introduced criteria to define the boundaries as shown in the Supplementary Materials. These anomalies exhibit a clear temperature dependence below 1.2 K. This notable similarity to the temperature dependence of SC$_{FP}$ suggests that $A_{FP}$ is a closely related phenomenon, with a similar energy scale to that of the superconductivity. This distinguishes $A_{FP}$ from the zero-field magnetically ordered phase M, which has a higher-temperature ordering temperature relative to superconductivity, that continues to increase with applied pressure.

An exciting possibility is that $A_{FP}$ is a precursor to superconductivity. Previous theoretical studies have shown that in extreme magnetic field Landau levels will have a dramatic influence on the low-temperature behavior of the upper critical field $H_{c2}$. Indeed, a more recent theoretical study indicates that SC$_{FP}$ might be a form of superconductivity that is enhanced by high-field Landau quantization of the conduction electrons. Therefore, $A_{FP}$ and SC$_{FP}$ may actually be the same superconducting phase occurring at different Landau levels, analogous to Shubnikov-de Haas oscillations. A challenge to this interpretation is that $A_{FP}$ is not accompanied by a zero-resistance state (Fig. 3a), but a plausible explanation for this is that $A_{FP}$ is actually partially superconducting due to the effects of energy-level broadening. An exciting possibility is that $A_{FP}$ serves as the upper bound to SC$_{FP}$, whose dome is cut off continuously.

Above $P_c$, $H_{FP}$ falls approximately between SC$_{FP}$ and SC$_{PM}$, whose ranges of stability would otherwise overlap. Above $P_c$, $H_{FP}$ serves as the upper field limit for SC$_{PM}$.

Above $P_c$, long-range ordered magnetism M sets in while SC$_{FP}$ survives and the anomalous feature $A_{FP}$ emerges as a local maximum in $\Delta f$. Solid dots are from experimental data, dashed lines are guide for the eye. Note that TDO and resistance measurements are performed for slightly different magnetic field direction, which leads to the slightly different phase diagrams for each pressure.

Anomalous features in the high-pressure region

A striking characteristic of the high-pressure FP phase is the emergence of additional features in the field range between the M and SC$_{FP}$ phases. These anomalies, denoted as $A_{FP}$, are pronounced in the $\Delta f$ data, and noticeable in $R$ data (Fig. 4). It is not clear whether these anomalies correspond to a thermodynamically distinct phase. In order to trace the evolution of these anomalies, we introduced criteria to define the boundaries as shown in the Supplementary Materials. These anomalies exhibit a clear temperature dependence below 1.2 K. This notable similarity to the temperature dependence of SC$_{FP}$ suggests that $A_{FP}$ is a closely related phenomenon, with a similar energy scale to that of the superconductivity. This distinguishes $A_{FP}$ from the zero-field magnetically ordered phase M, which has a higher-temperature ordering temperature relative to superconductivity, that continues to increase with applied pressure.

An exciting possibility is that $A_{FP}$ is a precursor to superconductivity. Previous theoretical studies have shown that in extreme magnetic field Landau levels will have a dramatic influence on the low-temperature behavior of the upper critical field $H_{c2}$. Indeed, a more recent theoretical study indicates that SC$_{FP}$ might be a form of superconductivity that is enhanced by high-field Landau quantization of the conduction electrons. Therefore, $A_{FP}$ and SC$_{FP}$ may actually be the same superconducting phase occurring at different Landau levels, analogous to Shubnikov-de Haas oscillations. A challenge to this interpretation is that $A_{FP}$ is not accompanied by a zero-resistance state (Fig. 3a), but a plausible explanation for this is that $A_{FP}$ is actually partially superconducting due to the effects of energy-level broadening. An exciting possibility is that $A_{FP}$ serves as the upper bound to SC$_{FP}$, whose dome is cut off continuously.

### METHODS

#### Crystal synthesis

Single crystals of UTe$_2$ were synthesized by the chemical vapor transport method using iodine as the transport agent. Elements of U and Te with atomic ratio 2:3 were sealed in an evacuated quartz tube, together with 3 mg/cm$^3$ iodine. The ampoule was gradually heated up and held in the temperature gradient of 1060/1000 °C for 7 days, after which it was furnace cooled to room temperature. The crystal structure was determined by X-ray powder diffraction using a Rigaku X-ray diffractometer with Cu–K$\alpha$ radiation. Crystal orientation was determined by Laue X-ray diffraction performed with a Photonic Science X-ray measurement system.
Magnetoresistance and tunnel diode oscillator (TDO) measurements were performed at the National High Magnetic Field Laboratory, Tallahassee, using the 45-T hybrid magnet. A non-magnetic piston-cylinder pressure cell was used for measurements under pressure up to 1.57 GPa, with Daphne oil 7575 as the pressure medium. Pressure was calibrated at low temperatures by measuring the fluorescence wavelength of ruby, which has a known temperature and pressure dependence. The TDO technique uses an LC oscillator circuit biased by a tunnel diode whose resonant frequency is determined by the values of LC components, with the inductance L given by a coil that contains the sample under study; the change of its electrical resistance and magnetic properties results in a change in resonant frequency. Identification of commercial equipment does not imply recommendation or endorsement by NIST. Error bars correspond to uncertainty of one standard deviation.

DATA AVAILABILITY
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: 24 March 2021; Accepted: 9 July 2021; Published online: 06 September 2021

Fig. 3 High-field magnetoresistance, showing phase boundaries at different pressures. Magnetic field is applied at 25° from b axis toward c axis. a Pressure dependence of 0.4 K magnetoresistance, showing the sharpening of phase boundaries at higher pressures and emergence of high-pressure phase $A_{FP}$. b The resistance at pressures 0.69, 0.85, and 1.12 GPa is zero between 11 and 45 T at 0.4 K. Zero resistance persists to zero magnetic field at these pressure values. c For fields lower than $H_{FP}$, the superconducting transitions $H_{C2}$ are broad, but $H_{FP}$ is sharp. As shown in inset, $H_{FP}$ is hysteretic, reflecting the 1st order transition, both at high temperatures in the FP phase and low temperature in the SC$_{FP}$ phase. d Much sharper superconducting phase boundaries occur once $H_{FP}$ limits SC$_{FP}$. As shown in inset, low-field phase boundaries are hysteretic and first order, as is the onset of SC$_{FP}$.

Fig. 4 Additional anomalies emerge in the high-pressure region. $A_{FP}$ phase appears at 1.54 GPa in both TDO and resistance data, which have the same temperature dependence as SC$_{FP}$ phase, indicating $A_{FP}$ phase might be the precursor of superconductivity. Magnetic field is applied at 30° from b axis toward c axis for TDO measurement, and 25° from b axis toward c axis for resistance measurement.

Measurement
Magnetoresistance and tunnel diode oscillator (TDO) measurements were performed at the National High Magnetic Field Laboratory, Tallahassee, using the 45-T hybrid magnet. A non-magnetic piston-cylinder pressure cell was used for measurements under pressure up to 1.57 GPa, with Daphne oil 7575 as the pressure medium. Pressure was calibrated at low temperatures by measuring the fluorescence wavelength of ruby, which has a known temperature and pressure dependence. The TDO technique uses an LC oscillator circuit biased by a tunnel diode whose resonant frequency is determined by the values of LC components, with the inductance L given by a coil that contains the sample under study; the change of its electrical resistance and magnetic properties results in a change in resonant frequency. Identification of commercial equipment does not imply recommendation or endorsement by NIST. Error bars correspond to uncertainty of one standard deviation.

REFERENCES
1. Ran, S. et al. Nearly ferromagnetic spin-triplet superconductivity. Science 365, 684–687 (2019).
2. Sundar, S. et al. Coexistence of ferromagnetic fluctuations and superconductivity in the actinide superconductor UTe$_2$. Phys. Rev. B 100, 140502 (2019).
3. Hutanu, V. et al. Low-temperature crystal structure of the unconventional spin-triplet superconductor UTe$_2$. Acta Crystallogr. Sect. B 76, 137–143 (2020).
4. Duan, C. et al. Incommensurate spin fluctuations in the spin-triplet superconductor candidate UTe$_2$. Phys. Rev. Lett. 125, 237003 (2020).
5. Aoki, D. et al. Unconventional superconductivity in heavy fermion UTe$_2$. J. Phys. Soc. Jpn. 88, 043702 (2019).
6. Metz, T. et al. Point-node gap structure of the spin-triplet superconductor UTe$_2$. Phys. Rev. B 100, 220504 (2019).
7. Bae, S. et al. Anomalous normal fluid response in a chiral superconductor UTe$_2$. Nat. Commun. 12, 2644 (2021).
8. Nakamine, G. et al. Superconducting properties of heavy fermion UTe$_2$ revealed by $^{125}$Te-nuclear magnetic resonance. J. Phys. Soc. Jpn. 88, 113703 (2020).
9. Nakamine, G. et al. Anisotropic response of spin susceptibility in the superconducting state of UTe$_2$ probed with $^{125}$Te-NMR measurement. Phys. Rev. B 103, L100503 (2021).
10. Hayes, I. M. et al. Multicomponent superconducting order parameter in UTe$_2$. Science 373, 797–801 (2021).
11. Shishidou, T., Suh, H. G., Brydon, P. M. R., Weinert, M. & Agterberg, D. F. Topological band and superconductivity in UTe$_2$. Nature 579, 523–527 (2020).
12. Jiao, L. et al. Chiral superconductivity in heavy-fermion metal UTe$_2$. Nature 579, 1250–1254 (2019).
13. Ran, S. et al. Extreme magnetic field-boosted superconductivity. Nat. Phys. 15, 515–520 (2019).
14. Knebel, G. et al. Field-reentrant superconductivity close to a metamagnetic transition in the heavy-fermion superconductor UTe$_2$. J. Phys. Soc. Jpn. 88, 063707 (2019).
15. Knafo, W. et al. Magnetic-field-induced phenomena in the paramagnetic superconductor UTe$_2$. J. Phys. Soc. Jpn. 88, 063705 (2019).
16. Knafo, W. et al. Comparison of two superconducting phases induced by a magnetic field in UTe$_2$. Commun. Phys. 4, 40 (2021).
17. Miao, L. et al. Low energy band structure and symmetries of UTe$_2$ from angle-resolved photoemission spectroscopy. Phys. Rev. Lett. 124, 076401 (2020).
18. Xu, Y., Sheng, Y. & Yang, Y.-F. Quasi-two-dimensional fermi surfaces and unitary spin-triplet pairing in the heavy fermion superconductor UTe$_2$. Phys. Rev. Lett. 123, 217002 (2019).
19. Ishizuka, J., Sumita, S., Daido, A. & Yanase, Y. Insulator-metal transition and topological superconductivity in UTe$_2$ from a first-principles calculation. Phys. Rev. Lett. 123, 217001 (2020).
20. Lebed, A. G. & Yamaji, K. Restoration of superconductivity in high-parallel magnetic fields in layered superconductors. Phys. Rev. Lett. 80, 2697–2700 (1998).
21. Mineev, V. P. Reentrant superconductivity in UTe$_2$. JETP Lett. 111, 715–719 (2020).
22. Lebed, A. G. Restoration of superconductivity in high magnetic fields in UTe$_2$. Mod. Phys. Lett. B 34, 2030007 (2021).
23. Miyake, A. et al. Metamagnetic transition in heavy fermion superconductor UTe$_2$. J. Phys. Soc. Jpn. 88, 063706 (2019).
24. Lin, W.-C. et al. Tuning magnetic confinement of spin-triplet superconductivity. npj Quantum Mater. 5, 68 (2020).
25. Ran, S. et al. Enhancement and reentrance of spin triplet superconductivity in UTe$_2$ under pressure. Phys. Rev. B 101, 140503 (2020).
26. Braithwaite, D. et al. Multiple superconducting phases in a nearly ferromagnetic system. Commun. Phys. 2, 147 (2019).
27. Thomas, S. M. et al. Evidence for a pressure-induced antiferromagnetic quantum critical point in intermediate-valence UTe$_2$. Sci. Adv. 6, eabc8709 (2020).
28. Aoki, D. et al. Multiple superconducting phases and unusual enhancement of the upper critical field in UTe$_2$. J. Phys. Soc. Jpn. 89, 053705 (2020).
29. Imajo, S. et al. Thermodynamic investigation of metamagnetism in pulsed high magnetic fields on heavy fermion superconductor UTe$_2$. J. Phys. Soc. Jpn. 88, 083705 (2019).
30. Niu, Q. et al. Fermi-surface instability in the heavy-fermion superconductor UTe$_2$. Phys. Rev. Lett. 124, 086601 (2020).
31. Niu, Q. et al. Evidence of fermi surface reconstruction at the metamagnetic transition of the strongly correlated superconductor UTe$_2$. Phys. Rev. Res. 2, 033179 (2020).
32. Gruenberg, L. W. & Gunther, L. Effect of orbital quantization on the critical field of type-II superconductors. Phys. Rev. 176, 606–613 (1968).
33. Tešanović, Z., Rasolt, M. & Xing, L. Quantum limit of a flux lattice: superconductivity and magnetic field in a new relationship. Phys. Rev. Lett. 63, 2425–2428 (1989).
34. Song, K. W. & Koshelev, A. E. Strong Landau-quantization effects in high-magnetic-field superconductivity of a two-dimensional multiple-band metal near the Lifshitz transition. Phys. Rev. B 95, 174503 (2017).
35. Park, M. J., Kim, Y. B. & Lee, S. Geometric superconductivity in 3d Hofstadter butterfly. Preprint at https://arxiv.org/abs/2007.16205 (2020).

ACKNOWLEDGEMENTS
We acknowledge D. Agterberg, G. S. Boebinger, A. E. Koshelev, A. Lebed, and V. P. Mineev for inspiring discussions. Research at the University of Maryland was supported by the National Institute of Standards and Technology (NIST), the US National Science Foundation (NSF) Division of Materials Research Award No. DMR-1610349, the US Department of Energy (DOE) Award No. DE-SC-0019154 (experimen
tal investigations), and the Gordon and Betty Moore Foundation’s EPiQS Initiative through Grant No. GBMF4419 (materials synthesis). Work performed at the National High Magnetic Field Laboratory, USA (NHMFL) was supported by NSF through NSF/DMR-1644779 and the State of Florida.

AUTHOR CONTRIBUTIONS
N.P.B. and S.R. conceived and designed the study. S.R. and S.R.S. synthesized the single-crystalline samples. S.R., S.R.S., I.L. and D.G. performed the electrical resistivity and tunnel diode oscillator measurements. S.R., J.P. and N.P.B. analyzed the data and wrote the paper with everyone’s contribution.

COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41535-021-00376-9.
Correspondence and requests for materials should be addressed to S.R. or N.P.B.
Reprints and permission information is available at http://www.nature.com/reprints
Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit https://creativecommons.org/licenses/by/4.0/.

This is a U.S. Government work and not under copyright protection in the US; foreign copyright protection may apply 2021