Quasi-uniaxial pressure induced superconductivity in the stoichiometric compound UTe₂

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The recent discovery of superconductivity in heavy the fermion compound UTe₂, a candidate topological and triplet-paired superconductor, has aroused widespread interest. However, to date, there is no consensus on whether the stoichiometric sample of UTe₂ is superconducting or not due to the lack of reliable evidence to distinguish the difference between the nominal and real compositions of samples. Here, we are the first to clarify that the stoichiometric UTe₂ is nonsuperconducting at ambient pressure and under hydrostatic pressure up to 6 GPa; however, we find that it can be compressed into superconductivity by application of quasi-uniaxial pressure. Measurements of resistivity, magnetoresistance, and susceptibility reveal that the quasi-uniaxial pressure results in a suppression of the Kondo coherent state seen at ambient pressure, which then leads to a superconductivity initially emerged on the *ab*-plane at 1.5 GPa. At 4.8 GPa, the superconductivity is developed in three crystallographic directions. The superconducting state coexists with an exotic magnetic ordered state that develops just below the onset temperature of the superconducting transition. The discovery of the quasi-uniaxial pressure-induced superconductivity with an exotic magnetic state in the stoichiometric UTe₂ not only provides new understandings of this compound, but also highlights the vital role of Te deficiency in developing the superconductivity at ambient pressures.

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I. INTRODUCTION

A key issue in condensed matter physics is to understand the evolution of electronic states caused by an interplay of electron interactions and competing order [1-3]. Unconventional superconductivity, which often develops in the vicinity of antiferromagnetic, ferromagnetic, or charge-ordered states [4-13], is a topic of particular interest. The recently discovered superconductor UTe₂ is one such example, in which superconductivity emerges from a correlated electronic state [14–18] and coexists with magnetism under pressure [19]. This material hosts magnetic fluctuations [20–24] and is believed to be one of the three-dimensional bulk topological superconductors [15,25-29] that exhibits different in-gap states at structurally different kinds of step edges [30]. UTe₂ is also highly sensitive to external control parameters such as pressure and magnetic field [19,31-34]. Moreover, re-entrant superconductivity is observed at high fields [35]. Remarkably, its superconducting ground state displays a large Sommerfeld coefficient (γ) [14,15,36], but without any corresponding thermal conductivity. However, this collection of remarkable properties have, to date, only been observed in the UTe₂ crystals with a nominally nonstoichiometric composition [36]. In this study, we employ the nonsuperconducting compound UTe₂, with a nominally stoichiometric composition, as a target material to investigate the real composition of the obtained sample and the effect of pressure on its transport properties.

Our UTe₂ single crystals were grown using a vapor transport technique with a nominal ratio of 1U:2Te, as reported elsewhere [36]. Single crystal diffraction measurements indicate that our UTe₂ sample crystalizes in an orthorhombic unit cell with the lattice parameters a = 4.1615(0) Å, b =6.1203(0) Å, and c = 13.94300(1) Å in the *Immm* (71) space group, and all ions are fully occupied in the lattice (Fig. 1). To confirm the stoichiometry of our sample and clarify the argument whether the stoichiometry of the UTe₂ sample is associated with its superconductivity [14,28,37–40], we performed a chemical analysis using inductively coupled plasma atomic emission spectroscopy (ICP-AES), a direct method to detect the composition of material. We found that its actual composition is UTe_{1.96}-very close to the nominally stoichiometric UTe2. Our chemical analysis results obtained from the ICP-AES method are in good agreement with the result reported in 1996 by the same method [41]. Our consistent results from the analysis of the ICP-AES and

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FIG. 1. Crystal structure and composition information for the nominally stoichiometric UTe₂ employed in the present study. (a) The single crystal x-ray diffraction patterns for the (hk0), (h0l) and (0kl) zones taken at 300 K. (b) The lattice and structure parameters of our UTe₂ sample.

the single crystal x-ray diffraction measurements manifest that the sample investigated in this study is almost fully stoichiometric UTe_2 .

II. EXPERIMENTAL METHODS

A. Setup for "quasi-uniaxial pressure experiment"

The uniaxial pressure shrinks the lattice along the *c*-axis but elongate the lattice along the *a*- or *b*-axes, while the hydrostatic pressure shrinks the lattice in all three crys-tallographic directions equally. However, an ideal uniaxial pressure condition cannot be realized currently in the experimental measurement of the high-pressure resistance in a diamond anvil cell (DAC). There are two main reasons: one is that the high-pressure measurement in a DAC requires a metallic gasket to separate the two anvils, which can effectively avoid the damage of the anvils at high pressure due to the fact that the gasket can serve as a buffer for the pressure applied; the other is that high-pressure resistance measurement

in a DAC needs an insulating layer to prevent the conductance of the sample from the metallic gasket. In this study, we employed a mixture of cubic boron nitride (c-BN) and epoxy as an insulating layer and placed them into the metallic gasket hole to protect the sample from short circuiting with the metallic gasket. Then, the single crystal sample was put on the top of the insulating layer (Fig. 2). In such a case, the compacted insulating powder can more or less restrain the lateral expansion along the a- or b-axes. Although our pressure environment cannot be the ideal uniaxial pressure that shrinks the lattice along the *c*-axis and elongates the lattice along the *a*- or b-axes, we can say that this is the available environment closest to the real uniaxial pressure in the high-pressure resistance measurements. Therefore, in order to describe the pressure condition adopted in this study appropriately or rigorously, we define the pressure applied on our sample as "quasi-uniaxial" pressure (Fig. 2). To know the sample pressure, we placed a piece of ruby flake on the top of the sample. Since the ruby flake is in the same pressure environment as that of the



FIG. 2. A sketch of our quasi-uniaxial pressure experiment setup displaying the arrangement for the gasket (gray), c-BN insulating layer (yellow), sample (black), and ruby (red) in a diamond anvil cell. The circle on the left displays the enlarged view for the sample and ruby flake.

sample, it can be used to characterize the pressure of the sample. Pressure was determined by the ruby fluorescence method [42].

B. High-pressure resistance measurement along the *c*-axis

To obtain the effective information of the *c*-axis resistance from the pressurized samples, we designed the following experimental setup (Fig. 3). The closer the distance between the two electrodes on the *ab*-plane, the more favorable one can minimize the effect of temperature dependence of resistance



FIG. 3. Experimental setup for the *c*-axis resistance measurements. The blue cuboid stands for the sample. The yellow and green bars represent the electrodes on the top and the bottom of the sample, respectively. *I* and *V* stand for the applied current and measured voltage. The distance between the electrodes is about 5 μ m.

[R(T)] along the *a*- and *b*-axes. As a result, the distance between the electrodes is reduced to ~5 μ m.

C. High-pressure susceptibility measurement

High pressure magnetic susceptibility measurements were carried out in a diamond anvil cell fabricated from a Cu-Be alloy. The sample is surrounded by a secondary coil (pickup coil) and a field-generating primary coil that is wound on the top of the secondary coil. The alternating flux through the pickup coil produces an ac voltage, which is the measured signal. When the sample is cooled below the superconducting transition temperature (T_c) , the field is expelled from the sample due to the superconducting shielding effect, forcing some of the flux lines out of the pickup coil and leading to a reduction in the induced voltage in the pickup coil [3,43,44]. For the primary coil, the alternating magnetic field was stimulated at 13.83 kHz, and the signal was collected at the same frequency. The superconducting transition can be captured by a step-like diamagnetic signal in this ac susceptibility measurement. The modulated magnetic field is provided by the secondary coil that is stimulated at 13.3 Hz, and its amplitude is around 9.9 mT. Here, the modulated susceptibility can be expressed by $\Delta \chi' = \chi'(9.9 \text{ mT}) \cdot \chi'(0 \text{ mT})$. Consequently, the result of $\chi'(B)$ produces a peak-like function in the secondary locked-in amplifier, instead of a step-like function, which is collected by a single phase-locked amplifier. Such a measurement with the magnetic field modulation has been used by the geophysics lab at the Carnegie Institute for the study of pressure effects on superconductivity and pressureinduced superconductivity in materials [44–46].

D. High-pressure specific heat measurement

For the high-pressure specific heat measurements, diamond anvils with 700- μ m flat and nonmagnetic rhenium gaskets were employed. A thin layered mixture of c-BN powder and epoxy was used as the insulating layer. The constantan was glued to one side of the sample with a dimension of about 0.4 × 0.4 × 0.05 mm³ as the heater; a chromel-AuFe (0.07%) thermocouple was fixed on the opposite side. With this technique, the small temperature oscillation (ΔT) generated by a heater is converted to an ac voltage signal. The ac calorimetry method adapted to high pressures is described in Refs. [47,48]. Pressure is determined by the ruby fluorescence method [42].

III. RESULTS AND DISCUSSIONS

A. Resistance versus temperature along the *a*-, *b*-, and *c*-axes separately under quasi-uniaxial pressure and hydrostatic pressure

We applied the quasi-uniaxial pressure to the nonsuperconducting UTe₂ single crystals and performed comprehensive measurements of R(T) along the *a*-, *b*-, and *c*-axes as function of pressure. As shown in Fig. 4(a), 4(g), and 4(m), the transport properties at ambient pressure in three crystallographic directions are highly anisotropic. From the fit of $R \sim \log(T)$, we find that the resistance of our sample is linearly proportional to $\log(T)$ [Fig. 4(a), 4(g), and 4(m)]. This linear behavior indicates that the maxima in the resistivity is



FIG. 4. Characterization of transport properties of single-crystal UTe₂. Temperature dependence of electrical resistance R(T), measured along the *a*-axis (a–f), *b*-axis (g–l), and *c*-axis (m–r), between 1 bar and 6.1 GPa. The insets detail the low-temperature behavior. T^* and T_m denote the onset temperature of the Kondo coherence and magnetic phase transitions. T_c^{onset} and $T_c^{R=0}$ stand for the temperatures of onset and zero-resistance superconducting phase transitions, respectively.



FIG. 5. (a, b) Temperature dependence of R_a and R_b of the UTe₂ sample measured under hydrostatic pressure condition, showing no superconducting transition up to 5.5 GPa.

associated with an intrinsic energy scale (T^*) of Kondo physics [49]. The coherence temperatures (T^*) related to Kondo coherence (KC) are $T^* = 88 \text{ K}$ [Fig. 4(g)], 137 K [Fig. 4(a)], and 217 K [Fig. 4(m)] for currents in the b, a, and c directions respectively. Upon increasing pressure to 0.9 GPa, the measured resistance changes dramatically in all three directions, but a clear coherence temperature $T_b^*(P)$ can only be resolved for the current along the *b*-axis. $T_b^*(P)$ shifts to a lower temperature upon compression [Fig. 3(h)], suggesting that the KC state is suppressed by pressure. An anisotropic response of the coherent temperature of the UTe₂ single crystal to currents in the a, b, and c directions is reminiscent of what is seen in the orthorhombic ferromagnetic superconductors UGe₂ [50], URhGe [51], and UCoGe [52]. It is likely that the layered material with an orthorhombic structure always holds the feature of the magnetic anisotropy. Such an anisotropy may result in the different T^* values exhibited in the R-T curves along the three crystallographic axes, which calls for further investigations in the future.

Around 1.5 GPa, an abrupt drop in resistance develops in R_a and R_b upon cooling [Fig. 4(c) and 4(i)], while R_c continues to grow on cooling [Fig. 4(o)]. At low temperatures, $R_a(T)$ and $R_b(T)$ exhibit jumps in resistivity at ~3.5 K and ~2 K, respectively [see insets of Fig. 4(c) and 4(i)], features of which become more pronounced upon compression. At 2.8 GPa, a zero-resistance state appears in $R_a(T)$ and $R_b(T)$ [Fig. 4(d) and 4(j)]; however, $R_c(T)$ still exhibits semiconducting behavior along the *c*-axis [Fig. 4(p)]. Finally, when the pressure reaches ~5 GPa, zero resistance develops in all three directions [Fig. 4(e), 4(k), and 4(q)]. We repeated the measurements on new samples and found the results are reproducible (see Appendix).

We performed hydrostatic pressure measurements for the UTe₂ single crystal in a similar pressure range. As shown in Fig. 5, no superconducting transition was found under pressures up to 5.5 GPa for the first run of the experiments. To repeat the experimental results, we loaded the second sample into a diamond anvil cell and applied a hydrostatic pressure up to 6 GPa. No superconductivity was observed down to 0.3 K (see Appendix). These results indicate that the stochiometric UTe₂ is not superconducting under hydrostatic pressure conditions less than 6 GPa.

B. Characterization of superconducting properties

The two resistance jumps are closely correlated, leading us to attribute them tentatively to a superconducting (T_c^{onset}) and magnetic (T_m) transition. To confirm this hypothesis, we carried out three further sets of measurement. First, we applied magnetic fields under pressures of 2.4 GPa and 5.1 GPa, and found that the high-temperature resistance jump shifts to lower temperatures [Fig. 6(a)-6(d)], while the second jump (knees), which lies between T_c^{onset} and the temperature of the zero-resistance superconducting phase transition $(T_c^{R=0})$, is not detectable at a field of more than 0.2 T, consistent with the identification that the jump in resistance is associated with a superconducting transition. Second, we simultaneously measured both ac susceptibility and resistance for the sample in the same high-pressure cell at 4.9 GPa [Fig. 6(e)]. A sharp cusp in $\Delta \chi'(T)$ [green points in Fig. 6(e)], indicating the development of diamagnetism, and zero resistance [red points in Fig. 6(e)] were observed, in which the onset of diamagnetism in $\Delta \chi'(T)$ at 2.3 K coincides with the zero-resistance value [53]. Third, we performed high-pressure specific heat (C) measurements on the sample at 6 GPa [Fig. 6(f)]. Two peaks were observed in the temperature dependence of C/T. To clarify the origin of the two peaks, we applied a magnetic field perpendicular to the *ab*-plane and found that the high-temperature peak seen at 3.8 K moves to higher temperature and becomes quite broader at 1 T. This behavior is similar to the feature of a ferromagnetic (FM) phase, i.e. its transition temperature in C(T) is broadened and softened with increasing magnetic fields [54,55]. Since the observed magnetic-like state in our sample has not been found before, further investigation to identify whether it is associated with an antiferromagnetic (AFM) or a FM state, or other exotic magnetic state is greatly needed. In this study, we define it as a magnetic (M) state. By contrast, the lower temperature peak migrates downward with increasing field, further confirming that it is associated with a zero-resistance superconducting transition $(T_c^{R=0})$. Moreover, the temperature of the low-temperature peak detected by the specific heat is almost the same as that detected by resistance measurements, further supporting that it results from a superconducting transition.



FIG. 6. Characterization of superconducting properties in single-crystal UTe₂. (a, b) The temperature dependence of resistance along *b*-axis (R_b) at 2.4 GPa under different magnetic fields, showing a dramatic dependence on the direction of the applied field. (c, d) The resistance along the *a*- and *b*-axes (R_a and R_b) as a function of temperature under different magnetic fields at 5.1 GPa. (e) Planar resistance and ac susceptibility ($\Delta \chi'$) as a function of temperature. Green points are the ac susceptibility, while red points show the resistance. (f) Specific heat coefficient (C/T) with respect to temperature (T) measured at 6 GPa under different fields. The lower temperature peak is associated with a bulk superconducting transition, while the higher temperature peak is magnetic. The inset shows the plot of superconducting transition temperature T_c versus critical field (H_{c2}). The dashed lines represent the Ginzburg-Landau fit to the data of H_{c2} .

C. High pressure x-ray diffraction

To clarify whether the observed superconducting transition of the UTe₂ sample subjected to the quasi-uniaxial pressure is related to a pressure-induced crystal structure phase transition, we conducted the high-pressure synchrotron x-ray diffraction measurements on the powder sample at 20 K at beamline 4W2 in the Beijing Synchrotron Radiation Facility. No new peak was observed and all peaks can be indexed well by the orthorhombic structure with the space group *Immm* up



FIG. 7. X-ray diffraction patterns of the UTe_2 sample collected at 20 K and different pressures.

to 6.1 GPa (Fig. 7). These results indicate that no crystal structure phase transition occurs, providing useful structure information for the pressure condition under which resistance, susceptibility, and heat capacity are measured. Since the quasi-uniaxial pressure environment is a combination of the uniaxial pressure with hydrostatic pressure, isolating the combined pressure is quite helpful in revealing the true pressure effect on the lattice parameters and deserves investigations in the future, although it is difficult in practical experiments.

D. Pressure-temperature phase diagram for the compressed UTe₂ single crystal

We summarize our experimental results in the pressuretemperature phase diagram in Fig. 8. It is seen that the onset temperature (T^*) of the Kondo coherence is suppressed by pressure and is not detectable at ~ 1.5 GPa—the pressure where the superconductivity emerges on the *ab*-plane at 3.6 K, followed by the exotic M phase transition (Figs. 4 and 6). The superconducting state, featured by the upturn of resistance in the c direction, coexists with the M state at less than 2.8 GPa (see Appendix), and the onset transition temperature of these two states (T_c^{onset} and T_M) varies slowly with increasing pressure. At the critical pressure P_{c1} (2.8 GPa), zero resistance of the superconducting state of the ab-plane is observed around 0.9 K [Fig. 4(d) and 4(j)], rising to ~2 K at 4.8 GPa (P_{c2}), where the superconducting state develops in the c direction. The zero resistance T_c of the superconducting state measured in all crystallographic directions for different experimental runs varies between 2 K and 1.8 K [Fig. 4(f)–(r), Appendix).

Unexpectedly, we find that the magnetism develops just below the onset T_c of the superconducting state, suggesting that the superconducting transition gives rise to a M state. This contrasts with the other superconductors, in which the superconductivity emerges from a preexisting but suppressed M state [56–58]. We note the work, reported by Thomas *et al.* [19], that the pressure-induced magnetic ordered state



FIG. 8. Pressure-temperature phase diagram for the compressed UTe₂ single crystal. The evolution of electronic state under uniaxial pressure, demonstrating the emergence of the superconducting and the exotic magnetic (M) state at P_{c1} (1.5 GPa) from a Kondo coherence (KC) state, and a crossover from planar superconductivity to a bulk superconducting state at P_{c2} (4.8 GPa). T^* represents the onset temperature of the KC state and $T_c^{\text{onset}}(R_a)$, $T_c^{\text{onset}}(R_b)$, and $T_c^{\text{onset}}(R_c)$ represent the onset temperature of the superconducting transition detected by resistance measurements for currents in the *a*, *b*, and *c* directions. $T_c^{R=0}(R_a)$, $T_c^{R=0}(R_b)$, and $T_c^{R=0}(R_c)$ stand for the zero-resistance superconducting transition temperature detected by resistance measurements for currents in the a, b, and c directions. $T_M(R)$ and $T_M(C_{ac})$ stand for the M phase transition temperature obtained by resistance and heat capacity measurements. $T_c(ac)$ and $T_c(C_{ac})$ represent the superconducting transition temperature measured through ac susceptibility and specific heat measurements. The superconductivity (SC) region filled with light violet refers to the superconductivity realized on the *ab*-plane only, while the region filled with dark violet refers to the superconductivity realized in all three crystallographic directions.

is different from ours. For our study, the magnetic ordered state in UTe₂ emerges from the superconducting state appearing just below the onset temperature of the superconducting transition (Fig. 8), while a magnetic state presents quite differently under the hydrostatic pressure condition where the superconducting state is suppressed [19]. We presume that the difference may stem from two aspects: the composition of the sample and the pressure environment applied. The composition of their sample is UTe₂ with an unnegligible amount of Te deficiency and the pressure environment is hydrostatic, while the composition of our sample is almost stoichiometric UTe_2 ($UTe_{1.96}$) and the pressure environment is quasi-uniaxial. In the case of quasi-uniaxial pressure, the sample tends to shrink more along the *c*-axis, but much less along the *a*- and *b*-axes, while in the case of hydrostatic pressure, the sample is supposed to be compressed equally in all the three crystallographic directions. Such distinct pressure effects between the two experiments may impact differently on the easy axis of its magnetic ordered structure, which may lead to a diverse magnetic state.



FIG. 9. (a–h) Temperature dependence of R_a and R_b for sample 2 at different pressures.

Our results raise several questions: (1) Is the stoichiometric UTe₂ the parent compound of superconducting UTe_{2- δ}?(2) Is the role of Te deficiency similar to or different from the other unconventional superconductors with anion deficiencies [7,59–61]? (3) Does the pressure-induced superconductivity from the stoichiometric UTe₂ has a nontrivially topological nature? All these questions are attractive issues that deserve further investigation.

IV. CONCLUSION

We report the observation of quasi-uniaxial pressureinduced superconductivity in stoichiometric compound UTe₂ that is not superconducting at ambient pressure. The measurements of resistivity, magnetoresistance, specific heat, and susceptibility reveal that the quasi-uniaxial pressure can suppress the Kondo coherent state and then drive a superconducting transition initially emerging on the *ab*-plane at 1.5 GPa. At 4.8 GPa, the superconductivity is developed in three crystallographic directions. The superconducting state coexists with an exotic magnetic ordered state that presents at just less than the onset temperature of the superconducting transition. These results highlight the vital role of Te deficiency in developing the superconductivity at ambient pressures, and also shed new insight on understandings of the underlying superconducting mechanisms in the correlated electron systems of the UTe2, and even in the other unconventional superconducting compounds.

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APPENDIX: REPRODUCIBLE RESISTANCE MEASUREMENTS ON UTe₂ UNDER DIFFERENT PRESSURE CONDITIONS

1. Reproducible resistance measurements on our UTe₂ single crystal under quasi-uniaxial pressure

We performed the same high-pressure measurements on the UTe₂ single crystals that were cut from different batches. As shown in Fig. 9, R_a and R_b as a function of temperature for sample 2 exhibit a similar behavior to that of the sample 1 (Fig. 4). Since the thickness of the sample 2 is thicker than that



FIG. 10. (a, b) Characterization of superconducting and M phase transition for the single crystal UTe_2 subjected to 6 GPa.



FIG. 11. (a–c) Temperature dependence of resistance in three crystallographic directions, displaying the enlarged view of Fig. 4(a), 4(g), and 4(m).

of the sample 1, we observed a flat residual resistance in $R_b(T)$ starting at ~1.6 K on cooling at 3.7 GPa. It is known that the observed resistance (R) is composed of three parts, sample resistance (R_s), contact resistance between the sample and the electrodes (R_c), and deformation resistance (R_d), which is associated with cracks, i.e. $R = R_s + R_c + R_d$. The last two terms of the observed resistance are related to residual resistance (R_r ; $R_r = R_c + R_d$). If the sample is brittle and thicker, it cracks easier under pressure. In this case, $R_d > 0$ and it gives rise to R_r . *T*-independent behavior with $R_r \sim 0.015\Omega$ at 3.7 GPa starting at ~1.6 K [Fig. 9(c) and 9(g)] and 0.0085 Ω at 4.8 GPa starting at ~2 K [Figs. 9(d) and 9(h)] indicates a superconducting transition because no superconductivity was observed from the same electrodes at temperatures as low as 20 mK in the megabar pressure range [62].

In order to confirm the pressure-induced superconducting and M phase transition for the nominally stoichiometric UTe_2 further, we conducted resistance measurements on the sample that were cut from different batches for the third run and obtained very similar results (Fig. 10).

To display the linear behavior in log(T) clearly, we fit the data equally by a straight line for Fig. 2(a), 2(g), and (2(m) (see Fig. 11).

2. Reproducible resistance measurements on our UTe₂ single crystal under hydrostatic pressure

To repeat the experimental results, we loaded the second sample into a diamond anvil cell and applied hydrostatic pressure up to 6 GPa; no superconductivity was observed down to 0.3 K (Fig. 12). These results indicate that the stochiometric UTe_2 is not superconducting under hydrostatic pressure conditions less than 6 GPa.



FIG. 12. (a, b) R_a and R_b as a function of temperature for the UTe₂ sample measured under a hydrostatic pressure condition up to 6 GPa.

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