Pressure-induced structural transition and new superconducting phase in UTe$_2$

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We report the crystal structure and electronic properties under high pressure up to 10 GPa on a heavy fermion superconductor UTe$_2$ by means of x-ray diffraction and electrical resistivity experiments. The x-ray diffraction measurements under high pressure using a synchrotron light source reveal anisotropic linear compressibility of the unit cell up to 3.5 GPa, while a pressure-induced structural phase transition is observed above $P_{c-T} \sim 4$ GPa at room temperature, where the body-centered orthorhombic crystal structure with the space group $I4/mmm$ changes into a body-centered tetragonal structure with the space group $I41/amd$. The molar volume drops abruptly at $P_{c-T}$, while the distance between the first-nearest neighbor of U atoms, d$_{U-U}$, abruptly increases, implying a switch from the heavy electronic states to the weakly correlated electronic states. Surprisingly, a new superconducting phase at high pressures above 7 GPa was detected at $T_c > 2$ K with a relatively low upper-critical field, $H_{c2}(0)$. The resistivity above 3.5 GPa, thus in the high-pressure tetragonal phase, shows a large drop below 230 K, revealing a drastic change from the heavy electronic states to the weakly correlated state.

1. Introduction

UTe$_2$ is one of the hottest topics in condensed matter physics, because of its unusual superconducting properties, such as topological superconductivity, field-reentrant superconductivity, and multiple superconducting phases in a magnetic field as well as under high pressure. A possible analogy to ferromagnetic superconductors, such as UGe$_2$ and URhGe, had been pointed out at the beginning of the discovery of superconductivity in UTe$_2$.

Indeed, the huge field-reentrant superconductivity detected in the hard-magnetization axis in UCoGe and URhGe resembles that observed in UTe$_2$ for $H \parallel b$-axis. On the other hand, no magnetic ordering was found down to low temperatures, revealing a paramagnetic ground state in UTe$_2$, which is quite in contrast to the above-mentioned ferromagnetic superconductors.

In UTe$_2$, a first order metamagnetic transition is observed at $H_m \sim 35$ T at low temperature. Although UTe$_2$ had been considered at the verge of the ferromagnetic ordering, no microscopic evidence for ferromagnetic interactions was obtained experimentally. On the other hand, inelastic neutron scattering experiments clearly detected the antiferromagnetic fluctuations at the incommensurate wave vector. Recently, STM experiments lead to the observation of the charge density wave (CDW) or the pair density wave (PDW) in the superconducting state. After three years of studies, UTe$_2$ is found to be a remarkable heavy-fermion system, where the electronic, magnetic, and charge instabilities coupled to the occupancy of the $5f$ levels play important roles.

To date, the relationship between the crystal structure and electronic properties is intensively discussed. Hence, we recall the crystal structure of UTe$_2$. UTe$_2$ crystallizes in the body-centered orthorhombic structure with the space group $I4/mmm$ (No. 71, $D_{2h}^9$), where 4 formula units are included in the unit cell ($Z = 4$). The nearest and next nearest neighbor U atoms locate along the c- and a-axes forming a rectangular shape within the ac-plane. It is also pointed out that the nearest U forms a two-leg ladder along the a-axis. The crystal structure is so-called UTe$_2$-type, and few other materials form this structure, demonstrating that it is a quite unique structure.

The superconducting properties of UTe$_2$ depend strongly on its sample quality. It is also claimed that significant deficiency of U rather than Te affects the electronic/superconducting properties of UTe$_2$ from their precise analysis, indicating that there may be an instability in the crystal structure of UTe$_2$.

The pressure response of UTe$_2$ is very sensitive, displaying different superconducting and magnetic states. Applying the pressure, the superconducting transition temperature

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splits above 0.3 GPa, revealing multiple superconducting phases. At around 1 GPa, an unusual enhancement of $H_{c2}$ for $H \parallel a$-axis was detected at low temperature and high field region as a consequence of the multiple superconducting phases.\(^{28,29}\) Even at ambient pressure, multiple superconducting phases were also detected for the $b$-axis in specific heat measurements associated with the field-reentrant superconducting state. Above the critical pressure of $P_c = 1.5$ GPa, superconductivity is suppressed and the magnetically ordered phase, most likely antiferromagnetism, appears under higher pressure.\(^4,29\)

It is surprising that UTe$_2$ reveals such rich and unexpected phase diagrams under pressure and a magnetic field. This is most likely due to a singularity of the crystal structure and its instability. It is important to elucidate the pressure response of the crystallography and the electronic state at high pressures with good hydrostatic conditions using a high-quality single crystal. Here, we performed x-ray diffraction (XRD) experiments and resistivity up to 10 GPa. We found a drastic change in the crystal structure from the orthorhombic structure (space group: $Immm$) to the tetragonal structure ($I4/mmm$) above the critical pressure, $P_{O-T} \sim 4$ GPa at room temperature. Furthermore, the resistivity above 3 GPa shows a new phase transition around 230K, which is almost unchanged with increasing pressure up to 9 GPa. In addition, a reappearance of superconductivity was found at 9 GPa with low $H_{c2}$, indicating weak electronic correlations, in drastic contrast to superconductivity at low pressures. These results have been briefly reported in JPS meeting and LT29 by oral and poster presentation, respectively.\(^{30}\)

2. Experimental Procedure

Single crystals of UTe$_2$ were grown by the chemical vapor transport method using iodine as a transport agent as described in the ref. 2. The quality of single crystals was checked by the sharpness of the x-ray Laue pattern. The residual resistivity ratios at ambient pressure were 20–30, and a sharp specific heat jump due to the superconducting transition was detected at 1.6 K.

The XRD experiments under high pressure were carried out on the BL10XU beamline at the JASRI/SPring-8, Japan using a wavelength of $\lambda = 0.4130$ Å. Since UTe$_2$ is easily oxidized or damaged in particular for tiny pieces of samples, special attention was paid. Three XRD experiments using diamond anvil cells (DAC) were performed, where two of them were done on single crystals with different orientations, and the other was done with pulverized powder from the same batch of the single crystal. For the experiments using single crystals, two single crystals of UTe$_2$, namely a crushed single crystal piece with having (010) plane with a size of 90 $\mu$m × 90 $\mu$m, and a thickness of 30 $\mu$m (labeled with “single (1st)”), and an oriented crystal with having (100) plane with a size of 120 $\mu$m × 80 $\mu$m, and a thickness of 23 $\mu$m by polishing (labeled with “single (2nd)”) were loaded in different DAC setups with SUS gaskets together with helium as pressure transmitting medium in order to get compensated diffraction data, respectively. XRD using a powder sample was also performed using helium as a pressure-transmitting medium. The pressure inside the DAC was measured using the conventional ruby scale both before and after the diffraction measurements. The sample pressure was regulated by a helium gas compression system. An imaging plate was used to obtain two-dimensional diffraction patterns via oscillation photography methods, and the resulting data were integrated along the radial direction to get a 1-dimensional diffraction pattern.

Electrical resistivity under high pressures up to 9 GPa was measured by means of a conventional four-probe method with the current along the $a$-axis, using a Palm Cubic anvil press system\(^3\) installed at the Institute of Solid State Physics, the
University of Tokyo in a $^4$He cryostat down to 2 K. A mixture of Fluorinert FC70 and FC 77 with a 1:1 ratio was used as a pressure-transmitting medium. Good hydrostaticity of the pressure condition in this cubic anvil press was mainly guaranteed by the system itself, where the Teflon capsule inside the cubic pyrophyllite and/or MgO gasket was compressed from the 6 directions simultaneously. At 9 GPa, the lowest temperature was further extended down to 30 mK in a dilution refrigerator, and the magnetic field was applied along the c-axis of the original orthorhombic structure.

3. Experimental Results and Analyses

3.1 x-ray diffraction under high pressure

Figure 1 shows the XRD patterns of a crushed UTe$_2$ single crystal (“single (1st)”) at (a) 0.93, (b) 3.05, and (c) 4.24 GPa at room temperature. To obtain more detailed crystal structure parameters: $a$ = 3.89 Å and $c$ = 9.80 Å. The relative height of the peak intensities was not well reproduced, but they were sufficient to determine lattice parameters. Above 4 GPa, the observed XRD pattern was indexed to a body-centered tetragonal system to give the following crystal structure parameters: $a = 3.89$ Å and $c = 9.80$ Å. The detailed crystal structure analysis of the high-pressure tetragonal phase is described in the following section. The $a$-axis and $c$-axis direction in the low-pressure phase turns to nearly $a$-axis through the structural phase transition, which means...
The calculated volume compressibility $k_i$ is consistent with the above-mentioned hypothesis. The results from single crystals and powder samples are basically the same. Lattice parameters along $a$-, $b$-, and $c$-axes decrease almost linearly with increasing pressure. The compression curves are anisotropic and the obtained linear compressibility ($k_i = -\left(1/V_0\right)(dL_i/dP)_{P=0}$, $i = a, b, c$-axes), where $L_i$ is a lattice parameter along the crystallographic $i$-axis, is $k_a = 7.2 \times 10^{-3}$, $k_b = 4.0 \times 10^{-3}$, and $k_c = 5.8 \times 10^{-3}$ (GPa$^{-1}$) for $a$-, $b$-, and $c$-axes, respectively. The $a$-axis is the most compressive and the $b$-axis is the hardest. The elastic properties of uranium compounds are not well established, so far, due to a lack of sufficient examples. However, it is suggested empirically that the direction of U-U bonds is distinctly the “soft” crystallographic direction in UTX (T: transition metal, X: metalloid) system, since the $5f$-electrons of U participate in the bonding. In UTe$_2$, the direction along the $c$- and $a$-axes corresponds to the 1st and 2nd nearest neighbor U-bonding, respectively, and shows smaller linear compressibility, which is consistent with the above-mentioned hypothesis. The calculated volume compressibility $k_V = k_a + k_b + k_c = 17 \times 10^{-3}$ (GPa$^{-1}$) corresponds to the bulk modulus of $B = 59$ GPa. This value is even smaller than that of the tellurium element itself $B = 65$ GPa. UTe$_2$ is very “soft”. Thus, drastic changes in the physical properties in UTe$_2$ under pressure are caused by such small bulk modulus.

Figure 4 shows the molar volume change as a function of pressure up to 11 GPa at room temperature in single crystals (closed symbol) and powder (open symbol) samples. Pressure dependence of the relative volume change ($\Delta V/V_0 = (V(P) - V_0)/V_0$) with respect to the volume at ambient pressure ($V_0$) reaches already 3.5 % at 2 GPa, which is comparable to the volume reduction in liquid 3He (about 5 %), where the paramagnetic Fermi liquid phase changes to the solid phase on the melting curve at low temperature. Thus, one can easily imagine the rather drastic change that occurs in the electronic properties of UTe$_2$ such as the disappearance of the superconductivity and an occurrence of the magnetic ordering under about 2 GPa.

The volume compression is analyzed by following Murnaghan equation of state,

$$\frac{V}{V_0} = \left[ P \left( \frac{B_0}{B'} \right)^{1/2} + 1 \right]^{-1/2},$$

where $B_0$ and $B'$ are the bulk modulus and its pressure derivative at ambient pressure, respectively, and $V_0$ was calculated from the extrapolated lattice parameters to 0 GPa from the present pressure data. The best-fit parameters are $B_0 = 59$ GPa and $B' = 1.9$. The dotted blue line in Fig. 4 is the calculated curve using these parameters and eq. 1. The molar volume drops at the structural phase transition with a volume change of 10 %. It should be noted that the crystal structure turned back to the original orthorhombic structure below 1 GPa during decreasing pressure, which indicates the latent heat at the structural transition is large.

The results between single crystal (closed symbol) and powder samples (open symbol) are basically similar, but the main difference is the critical pressure of structural transition and coexistence region of low- and high-pressure phases. The single crystal data show the transition to the tetragonal structure almost suddenly at $P_{O-T} = 3.5$ and 4 GPa, while the coexistence of two structures occurs between 5 and 7 GPa in powder samples. This is most likely due to the pressure distribution in the gasket and internal strain and/or defects that could happen in the pulverization of single crystals.

![Image of Figure 4](image-url)

**Fig. 4.** (Color online) Pressure dependence of the molar volume in UTe$_2$ based on the orthorhombic ($I\text{mmm}$) and tetragonal ($I\text{4} \text{mmm}$) structure at 300 K. Closed and open circles are the results from single crystals and powder samples, respectively. An open diamond is taken from Ref. 24. The right axis corresponds to the scaled volume change. The dotted blue line indicates the results of fitting by the Murnaghan equation of state. The solid red and black lines are guides for eyes.

### 3.2 Electrical resistivity under high pressure

Figure 5(a) shows the temperature dependence of the electrical resistivity $\rho(T)$ of UTe$_2$ single crystal with the current along the orthorhombic $a$-axis at different pressures. The $\rho(T)$ curve in UTe$_2$ at ambient pressure is characterized by almost
temperature independent above 100 K and an abrupt decrease of ρ below 50 K, as shown in the dashed line in Fig. 5(a). It should be noted that the bare LDA band structure calculation predicts a Kondo semiconductor in spite of the fact that the relatively large volume of Fermi surfaces (about 20 % in BZ for each hole and electron Fermi surface with the compensation) are experimentally detected. The Coulomb repulsion $U$ and the strong correlation must be taken into account in the calculations in order to explain real Fermi surfaces, suggesting that the electronic states are sensitive to pressure and temperature. At 2 GPa, $ρ(T)$ still resembles the curve at ambient pressure with a gradually enhanced hump around 25 K, indicating that the heavy electronic state is still preserved or even enhanced at low temperatures. At lower temperatures, two anomalies are detected at 8.5 and 2.8 K, which may correspond to the transitions for the pressure-induced WMO and MO phases, respectively. At 3.5 GPa, $ρ(T)$ behavior changes drastically, where the electrical resistivity decreases monotonically with decreasing temperature and shows a clear shoulder around $T^{**} = 230$K and a small upturn at around $T_m = 4$ K. The anomaly at $T_m$ is consistent with the pressure-induced magnetic transition reported previously. With increasing pressure above 5 GPa, the $T_m$ anomaly smeared out, and $T^{**}$ slightly increases monotonically with an initial slope of 1.9 K/GPa as shown in Fig. 5(b). The origin of the anomaly at $T^{**}$ is not clear so far, but one can speculate that it corresponds to a drastic change from single site behavior to a well-coupled array at rather high temperatures, which is basically one order magnitude higher than the crossover temperature $T'$ detected at low pressure. Note that the value of resistivity at room temperature as a function of pressure shows no drastic change at the critical pressure $P_{O-T}$ for structural transition.

In order to investigate the low-temperature properties in more detail, resistivity experiments were extended at very low temperatures down to 30 mK. Figure 6(a) shows the low-temperature part of $ρ(T)$ under high pressure above 3.5 GPa of the high-pressure tetragonal phase. The onset of the pressure-induced superconductivity is detected at 7 GPa below 2 K. At 9 GPa, a clear superconducting transition by zero resistivity was detected below about 1 K. The onset of the superconducting transition temperature $T_{SC}$ is 2.2 K at 9 GPa in a zero magnetic field, where the superconducting transition temperature is defined with 80 % of the residual resistivity. It should be noted that the broadened SC transition can be attributed to the strain and cracks arising due to the structural transition associated with the drastic change of lattice parameters. In fact, the sample unloaded from the Palm Cubic anvil pressure cell after the measurement breaks up into small pieces, which was also seen in the samples taken after the XRD measurements under pressure.

We further investigated the superconducting property at 9 GPa in magnetic field and constructed the phase diagram. Here, the magnetic field was applied along the original c-axis in the low-pressure orthorhombic phase. Figures 6(b) and 6(c) show the temperature dependence of the electrical resistivity at the different magnetic fields and magnetic field dependence at 30 mK, respectively. Pressure-induced superconductivity disappears above 2.5 T, although the small onset of superconductivity persists up to about 3.8 T. We show in Fig. 6(d) the temperature dependence of the upper critical field $H_{c2}$ defined as 80 % of the resistivity of the normal state. The slope of the upper critical field is $-dH_{c2}/dT \sim 1$T/K at $T_{SC} = 2.2$ K, and the value of $H_{c2}(T=0)$ is about 2.5 T, which is smaller than the Pauli limit based on the weak coupling BCS theory. Furthermore, the temperature dependence of resistivity follows the Fermi liquid behavior ($ρ = ρ_0 + A T^2$) with the small $A$ coefficient ($A_{50GPa} \sim 1.4 \times 10^{-3} \mu Ω \cdot cm/K^2$), indicating superconductivity is based on the weakly correlated electronic states without contribution from the 5f-electrons. This can be compared with the $A$ coefficient at ambient pressure, $A_0 \sim 0.9 \mu Ω cm/K^2$. The ratio, $A_{9GPa}/A_0 \sim 600$ is comparable or the same order with $(T^{**}/T^*)^2 \sim 300$. Furthermore, the Sommerfeld coefficient for the specific heat is expected to be less than 10 mJ K$^{-2}$mol$^{-1}$ at high pressure, assuming the Kadowaki-Woods ratio, suggesting a weak electronic correlation. The small initial slope of $H_{c2}$ at 9 GPa is also reasonable, compared to the large values, 34 T/K and 7.5 T/K for $b$ and $c$-axis, respectively detected at ambient pressure, in accordance with the change of the effective masses, since $H_{c2}$ is proportional to the $(m^*T_{xc})^2$ assuming the Fermi surfaces are unchanged.

It is known that conventional superconductivity of the tellurium element is induced above 4 GPa, but the superconducting state of tellurium is easily killed at the magnetic field of 0.03 T. Thus, the observed pressure-induced superconductivity is not due to the remaining tellurium but is intrinsic in the high-pressure tetragonal phase of UTe$_2$.

Figure 7 shows the pressure-temperature phase diagram of UTe$_2$. The structural transition from orthorhombic to tetragonal occurs at $P_{O-T} \sim 5.5$ GPa at room temperature, and $P_{O-T}$ shifts to the higher pressure, $P_{O-T} \sim 5.5$ GPa on cooling. At low pressure above 1.5 GPa, $T_{MO}$ corresponding to a magnetic order increases rapidly with pressure, and it disappears above 3.5 GPa. New anomalies denoted by $T^{**}$ start appearing above 3 GPa, and are almost unchanged up to 9 GPa. Superconductivity reappears around 2 K in the tetragonal phase, where the electronic state is drastically changed without the strong electronic correlations as demonstrated by resistivity curves and the low $H_{c2}$.

4. Discussion

4.1 Pressure-induced structure transition at high pressure
Here, we discuss the pressure-induced structural transition around $P_{O-T} \sim 4$ GPa. We recall that the observed diffraction peaks are quite different from the ones in the orthorhombic phase. The diffraction pattern is found to be reproduced with two domains of the body-centered tetragonal lattice with $a \sim 3.98$ Å and $c \sim 9.80$ Å. The unit cell volume at ambient pressure is $357 \, Å^3$ with $Z = 4$, whereas that above $P_{O-T}$ is about $149 \, Å^3$. This indicates the tetragonal unit cell contains two formula units of UTe$_2$ ($Z \approx 2$). Considering the ex-
tinction rule, the possible candidates of the space group of the high-pressure structure are $I4/mmm$ or its subgroup. Recently, it is theoretically suggested that the ambient-pressure $Immm$ phase of UTe$_2$ transforms into a ten-fold coordinated high-pressure $I4/mmm$ phase at 9 GPa. Thus, finally we could take $I4/mmm$ for the most probable candidate of the high-pressure crystal structure. Given the symmetry of $Z = 2$ and $I4/mmm$, it is natural to assume that U exists on the 4-fold axis and on the mirror (because otherwise, the number of U atoms would be more than 4 in a unit cell). Therefore, assuming that U exists at the site $2a$ ($0$ $0$ $0$), Te with 4 atoms in the unit cell would be limited to site $4c$ ($0$ $\frac{1}{2}$ $0$), $4d$ ($\frac{1}{2}$ $0$ $\frac{1}{2}$) or $4e$ ($0$ $0$ $z$). Among them, the $4e$ site is the one where the U-Te and Te-Te distances are about 3.17 and 3.19 Å. In other cases, the U-Te distance of 1.99 Å with 4e Te, and Te-Te distance of 2.81 Å with 4d Te, are too short considering the ionic radius and strong Coulomb interaction, respectively. The crystal structure, in this case, is shown in Fig. 8(c). The simulated XRD pattern assuming this structure reproduces well the powder XRD pattern of the high-pressure phase. The crystal structure of the low-pressure orthorhombic (LP) and high-pressure tetragonal (HP) phases are depicted in Figs. 8(a) and 8(c), respectively.

For this drastic changes of the crystal structure and lattice parameters, large atomic displacements are inevitably required. Three possible unit cells that could be transformed into the body-centered tetragonal structure are shown in Figs. 8(d)-(f). In all cases, the $ab$-plane for the tetragonal structure is based on the close distances for the 1st and 2nd nearest neighbors, $d_1 \sim 3.78$ Å and $d_2 \sim 4.17$ Å, respectively. In Fig. 8(d), the rung denoted by $d_1$ should be tilted, moreover the atomic position must move to the $a$-axis in order to get the tetragonal structure. The direction for the new $c$-axis in the tetragonal structure approximately corresponds to the reciprocal [011] direction in the orthorhombic structure. Note that the reciprocal [011] direction (24 deg tilted from $b$ to $c$-axis) is different from [011] direction in real space (24 deg tilted from $c$ to $b$-axis). In Fig. 8(e), the rung should be also tilted, but no atomic displacement is required along $a$-axis.

The new $c$-axis approximately corresponds to the [011] direction in real space. In Fig. 8(f), a U atom largely moves along the orthorhombic $c$-axis to get the body-centered tetragonal structure. This is rather simple displacement of U atoms, indicating that the orthorhombic $b$-axis switches to the tetragonal $c$-axis.

![Fig. 5](image_url) (Color online) (a) Resistivity at different pressures up to 9 GPa for $J \parallel a$-axis in UTe$_2$. The data at ambient pressure shown by the dotted line are taken from Ref. 2. (b) Pressure dependence of $T^\ast$.

![Fig. 6](image_url) (Color online) (a) Low-temperature resistivities at different pressures above 3.5 GPa in UTe$_2$. (b) Low-temperature resistivities at different fields at 9 GPa. (c) Field dependence of the resistivity at the lowest temperature, 30 mK at 9 GPa. (d) Superconducting phase diagram at 9 GPa. Here $T_N$ or $H_S$ are defined by the temperature or field, at which the resistivity is reduced to the value of 80% from the normal state resistivity.
Although we cannot determine which atomic displacement is correct, we speculate that the [011] or the reciprocal [011] directions is important and it is related to the structural instabilities. Compared with the crystal structures in LP and HP phases focusing on the U position, the rectangular arrangement of U atoms in the ac-plane in the LP phase seems to correspond to the basal plane in the HP tetragonal phase. In this structure transformation, the crystal along the a-axis keeps its direction, and the ac-plane including the U rectangular tilts about 25 degrees from the b to c-axis. With this transformation, the [011] direction in the reciprocal space in the LP orthorhombic phase, most likely, turns to the [001] axis in the HP tetragonal axis. It means that the reciprocal [011] direction in the LP phase has been already potentially important in the stability of the crystal structure. It should be noted that the above-mentioned transformation is consistent with the change of the Laue spots as shown in Fig. 1(b) into 1(c).

Since our experiment was carried out on single crystals, not all the diffraction peaks from the tetragonal structure can be obtained. It brings difficult to determine the atomic position of Te precisely. It is known that several prototype compounds crystallize in the tetragonal I4/mmm type, with AB\textsubscript{2} composition. For example, MoS\textsubscript{2} and WGe\textsubscript{2} are typical compounds, where the c/a ratio is quite similar to the pressure-induced HP phase in UTe\textsubscript{2} and z parameters are 0.335 in both cases. From the theoretical calculation, Hu et al. predict $z = 0.337$.\textsuperscript{36} Here, we assume the z parameter to be 0.335 in the HP phase in UTe\textsubscript{2} as for the prototype compounds. The pressure dependence of the 1st and 2nd nearest neighbor U distances are depicted Fig. 9. The diverse inter-atomic distances become more simply in the high-symmetry structure of the I4/mmm tetragonal phase. Note that UTeAs, UTeP, and UTeGe are known to form the tetragonal structure with the space group I4/mmm. The c/a ratio is, however, much larger, and 4 molecules exist in the unit cell ($Z = 4$).

It is worth mentioning that the c-axis in the high-pressure phase corresponds to the reciprocal [011] direction in the original low-pressure structure, which is tilted by 24 deg from the b to c-axis. Huge field-reentrant superconductivity is observed around this field direction above the metamagnetic field $H_m \sim 40–50$T. According to the band calculation of GGG+U ($U = 2$ eV) based on the $5f$-itinerant model, the maximal and minimal cross-sectional areas of the electron Fermi surface coincide for the field direction to the reciprocal [011] direction, which is the so-called Yamaji angle. The increase of the density of states is generally expected under a magnetic field at Yamaji angle, and indeed in UTe\textsubscript{2}, the specific heat increases with the field, showing a positive jump at $H_m$.\textsuperscript{19} Furthermore the drastic change of of Hall resistivity is also found for the field direction along b-axis and the reciprocal [011] direction.\textsuperscript{18,19}

This suggests that Fermi surface instabilities play a role in field-reentrant superconductivity around [011] direction above $H_m$. The switching principal axis to the original reciprocal [011] direction can be another mark of these Fermi surface instabilities. One can thus expect further singularities along the [011] or the reciprocal [011] direction by uniaxial stress, thermal expansion and ultrasound experiments.

It is interesting that the flat crystal surface perpendicular to the reciprocal [011] as well as [001] direction appear often in as-grown single crystals by the chemical transport method and by the flux method. The natural crystal surface is generally related to the growth speed, which could be affected by the crystal structure. This may indicate a precursor for the singularity of the reciprocal [011] direction.

### 4.2 Pressure-induced electronic phase transitions

There is two pressure-induced phase transition that appears in UTe\textsubscript{2} for its electronic states in the high-pressure tetragonal phase. One is characterized by the kink of the electrical resistivity at $T^{**} = 230$ K and the other one is a superconducting phase transition below 2 K. The former transition was also observed in the previous study.\textsuperscript{20} In their report, the $T^{**}$ anomaly is observed for the geometry $H \parallel b$ which makes internal strain along the b-axis ($u \parallel b$), with their semi-hydrostatic Bridgman anvil cell, but no anomaly was observed for the geometry $H \parallel c$ ($u \parallel c$), suggesting that the $T^{**}$ anomaly is sensitive to the uniaxial stress. The hydrostaticity of the pressure in our Palm Cubic anvil-type pressure cell is guaranteed by the design itself independent of the pressure-transmitting medium. Therefore, The $T^{**}$ anomaly can be favorable to the compression along the b-axis in the LP phase. When the unit cell is compressed along the b-axis, the atomic displacement occurs to avoid strong Coulomb repulsion, particularly between Te(2) sites mentioned above. It is known that several uranium pnictides and chalcogenides exhibit high magnetic ordering temperatures. Thus it is not surprising that
the magnetic order could occur at high-temperature in UTe$_2$.
The nearest neighbor uranium distance in the high-pressure phase of UTe$_2$ reaches 3.9 Å which is sufficiently larger than the so-called Hill limit. Therefore it could be possible that the $T^{**}$ anomaly is attributed to a switch from 5$f$ single site behavior to coupled 5$f$ array. The nature of the transition is the primary task for future study. The resistivity anomaly is resembling a conventional 2nd-order phase transition in a ferromagnet (or antiferromagnet without a superzone boundary effect - possible for the suggested structure assuming an AF coupling within one unit cell with magnetic propagation vector $k = (0 0 0)$). However, without further study, we cannot exclude other options such as a valence change or a kind of Lifshitz transition at the pressure-driven transition.

It is clear that the observed superconductivity at high pressure cannot be attributed to the heavy electronic state, as shown by low $H_{c2}(0)$ and small $A$ coefficient. The drastic change of electronic states associated with the structural change may invoke a new superconducting state without 5$f$-electron correlation. A key point is that the distance between the next-nearest neighbors of U atoms suddenly increases at the critical pressure from orthorhombic to tetragonal structure as shown in Fig. 9, despite the fact that volume decreases with pressure; this leads to the non-trivial results of 5$f$ occupancy, which jumps to 4 % higher than that at ambient pressure.

Similar conclusions were recently given on the pressure studies, where the transition from the orthorhombic to tetragonal transition is reported at around 5 GPa under the quasi-hydrostatic condition, associated with an increase of the Bulk modulus by 45 % which reaches a value above the critical pressure of the structural transition. This is close to a full valence state and the spectacular boost of $d_{U-1}$ by 8 %. The value of $d_{U-1}$ is even larger than that at ambient pressure. Furthermore, a new key result is that a trivalent state of U atom recovers at high pressure in the tetragonal structure.

Qualitatively these observations are in agreement with the theoretical calculations, in which a structural transition from orthorhombic to tetragonal is predicted at 9 GPa; the main pressure effect is to boost the chemical bonding of the pair of Te electrons. A large Bulk modulus is also predicted just above the critical pressure. Quantitative differences critical parameters between experiments and theory is most likely the difficulty of the treatment for the strong correlation effect in UTe$_2$.

5. Summary

We have performed the XRD experiments using single crystals and powder samples to clarify the pressure effect of the crystal structure as well as the electrical resistivity measurements under high pressure up to 9 GPa. X-ray diffraction study under high pressure reveals anisotropic linear compressibility of the unit cell, up to 3 GPa, and a pressure-induced structural phase transition above 4 GPa at room temperature. The body-centered orthorhombic crystal structure with the space group $Immm$ transforms into a body-centered tetragonal structure with the space group $I4/mmm$.

In addition, we have observed a drastic change in the electronic states accompanied by the structural phase transition and the reappearance of superconductivity above 7 GPa, which does not correspond to heavy fermion superconductivity. This could be linked to a quite rare case, where the $U^{3+}$ configuration is recovered at high pressure.

![Fig. 8](image-url)

**Fig. 8.** (Color online) (a) Orthorhombic structure with the space group $Immm$ at low pressure. (b) A view from $b$-axis. The distances of the 1st and 2nd nearest neighbors of U atoms are $d_1 = 3.78$ Å and $d_2 = 4.17$ Å, respectively. (c) Tetragonal structure with the space group $I4/mmm$ at high pressure. (d)(e)(f) Three possible unit cells for the transformation to the body-centered tetragonal structure are displayed by bonds between U atoms.

Acknowledgement

The authors would like to thank W. Knafo, and L. Havela for the fruitful discussion, and S. Nagasaki for the technical support. This work was performed with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposal Nos. 2020A0741, 2020A0740, and 2021A1527) and under the Inter-University Cooperative Research Program of the Institute for Materials Research, Research Center for Nuclear Materials Science of Tohoku University (Proposal Nos. 202012-IRKAC-0017, 202012-IRKAC-0056, 202112-IRKAC-0029, and 202112-IRKAC-0041). This work was financially supported by KAKENHI (JP19K21840, JP19H00648, JP20K03827, JP20K20889, JP20H00130, JP20KK0061, JP22H04933, JP22KK03516).

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Fig. 9. (Color online) Pressure dependence of the distance of the 1st nearest neighbor of U atoms, $d_1$ (closed symbols). The 2nd nearest neighbor, $d_2$ at low pressure is also shown (open symbols). The results of single crystals, 1st run, 2nd run and powder sample are shown by circles, square, and triangle, respectively.
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