Metamagnetic Transition in Heavy Fermion Superconductor UTe$_2$

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We have studied the magnetization of the recently discovered heavy fermion superconductor UTe$_2$ up to 56 T in pulsed-magnetic fields. A first-order metamagnetic transition has been clearly observed at $H_{m} = 34.9$ T when the magnetic field $H$ is applied along the orthorhombic hard-magnetization $b$-axis. The transition has a critical end point at $\sim 11$ K and 34.8 T, where the first order transition terminates and changes into a crossover regime. Using the thermodynamic Maxwell relation, we have evaluated the field dependence of the Sommerfeld coefficient of the specific heat directly related to the superconducting pairing. From the analysis, we found a significant enhancement of the effective mass centered at $H_{m}$, which is reminiscent of the field-reentrant superconductivity of the ferromagnet URhGe in transverse fields. We discuss the origin of their field-robust superconductivity.

Heavy fermion superconductivity (SC) is one of the most interesting topics in strongly correlated electron systems (SCES). In particular, the discovery of coexistence of ferromagnetism (FM) and spin triplet SC in UGe$_2$ opens a new chapter.\(^1\) The appearance of new materials such as URhGe\(^2\) and UCoGe\(^3\) with low Curie temperature ($T_{\text{Curie}}$) shows the key role of Ising interaction and was hints on elegant way to obtain field-reentrant SC or its reinforcement by lowering $T_{\text{Curie}}$ in transverse field scan with respect to the initial perpendicular FM sublattice magnetization.\(^4,7\)

Recently, SC was discovered in the paramagnetic (PM) UTe$_2$, with a relatively high superconducting transition temperature, $T_{\text{sc}} = 1.6$ K.\(^8,9\) SC is believed to be of unconventional spin-triplet type, since the $H_{c2}$ highly exceeds the Pauli limit. In contrast to the previous spin-triplet FM superconductors, the ground state of UTe$_2$ is PM at least down to $T_{\text{sc}}$, most probably locating near a FM instability.\(^8,9\) UTe$_2$ crystallizes in a body-centered orthorhombic structure (space group: $Immm$) [see the inset of Fig 1(a)]. The nearest-neighbor (NN) U atoms in UTe$_2$ align along the $c$-axis with the shortest U-U distance $d_{\text{U-U}} = 3.78$ Å.\(^10\) A similarity with the previous FMSC is the field robust SC phase for $H \parallel b$-axis, which is perpendicular to the easy magnetization $a$-axis at low fields. The SC phase still exists even at 20 T,\(^5\) although the temperature dependence of $H_{c2}$ appears sample dependent.\(^9\) It is expected that magnetic fluctuations will drastically develop at high fields, being coupled with the Fermi surface instabilities when the magnetic polarization reaches a sufficiently critical value.

A key observation in UTe$_2$ for $H \parallel b$-axis is a maximum of the magnetic susceptibility $\chi$ at $T_{\text{max}} \sim 35$ K.\(^8,10\) In metallic SCES, many compounds show metamagnetic behavior at $H_{m}$, which have a similar energy scale to their $T_{\text{max}}$\(^11,12\). It is also interesting to mention the expected magnetic anisotropy from the point of view of the crystal structure. In many U compounds, the magnetic moments are aligned along a direction perpendicular to the axis connecting the NN U atoms in the magnetic ordered state.\(^13\) The NN U atoms of URhGe and UCoGe, for example, make a zigzag-chain along the $a$-axis, resulting in a FM ground state with the easiest magnetization $c$-axis.\(^7\) By applying transverse magnetic fields ($H \parallel b$-axis), reentrant SC in URhGe\(^2\) and field-reinforced SC in UCoGe\(^5\) occur. The enhancement of the FM fluctuations in the $bc$-plane is considered to have an important role.\(^7\) For UTe$_2$, the development of magnetic fluctuations in the $ac$-plane is expected at high fields.

Here, we performed magnetization ($M$) measurements in UTe$_2$ for $H \parallel a$ and $b$-axes in pulsed-magnetic fields up to 56 T and found a first order metamagnetic transition (MMT) accompanied by a huge jump of $M$ ($\Delta M \sim 0.6 \mu_B/\text{f.u.}$) for $H \parallel b$-axis at $H_{m} \sim 34.9$ T. We also observed a small anomaly in the $M(H)$ curve for $H \parallel a$-axis near 6.5 T. The MMT seems to occur when $M$ for $H \parallel b$ reaches that for $\mu_0 H \sim 6.5$ T along the $a$-axis. We discuss the $H$-dependence of the Sommerfeld coefficient ($\gamma$) derived from the temperature dependence of $M$ using a thermodynamic Maxwell relation, and the $H$-robustness of $T_{\text{sc}}$ triggered by the enhancement of the effective mass.

Single crystals of UTe$_2$ were grown using the chemical vapor transport method.\(^8,9\) $M$ in pulsed-magnetic fields was measured by the conventional induction method, employing coaxial pick-up coils. Pulsed-magnetic fields up to 56 T were applied using nondestructive pulse-magnets having typical durations of $\sim 36$ ms installed at the International MegaGauss Science Laboratory of the Institute for Solid State Physics of the University of Tokyo. The measurements were done for the field applied along the $a$ and $b$-axes and at low temperatures down to 1.4 K. Below 7 T, the temperature dependence of $M$ was measured by a commercial SQUID magnetometer at temperatures down to 1.8 K for $H \parallel a, b, c$-axes.

Figure 1(a) shows temperature ($T$) dependence of the magnetic susceptibility $M/H$ with $H$ applied along the orthorhombic principal axes. Consistently with previous reports,\(^8,10\) there is no indication of any phase transitions down to 1.8 K, suggesting a PM ground state before the establishment of SC. $M(T)$ for $H \parallel b$-axis shows a broad maximum at $T_{\text{max}} \sim 36$ K, which hardly depends on $H$ at least up to 7 T [Fig. 1(c)]. On the other hand, as shown in Fig. 1(b), $M(T)$ for $H \parallel a$...
increases upon cooling without saturation at low fields, but \( M(T) \) tends to saturate around 5 K above \( \sim 5 \) T. These saturations may correspond to the anomaly around 7 T in \( M(H) \) curve shown later.

Next, we show the results of \( M(H) \) in a pulsed field. The results at low field and obtained \( H_{c2}(T) \) curves are shown in the Supplemental Material.\(^{14} \) Figure 2 presents \( M(H) \) curves of \( \text{UTe}_2 \) for \( H \parallel a \)- and \( b \)-axes at 1.4 K. For the easiest magnetization \( a \)-axis at low fields, \( M \) monotonically increases and tends to saturate with increasing fields. Note that a small anomaly around 7 T is observed, as seen in the differential susceptibility \( dM/dH \). Remarkably, a huge jump in \( M \) (\( \Delta M \sim 0.6 \mu_\text{B}/\text{f.u.} \)) appears for \( H \parallel b \)-axis at \( H_m = 34.9 \) T, accompanied by a clear but rather small hysteresis loop. It is noted that \( \Delta M \) slightly decreases after several thermal and field cycles without any changes of \( H_m \) and the slope of \( M(H) \). It may arise from the strong field angle dependence or the damage due to the magnetostriction. The data shown in Fig. 3 between 4.2 K and 17 K, which are used for the derivation of Sommerfeld coefficient in the specific heat \( \gamma \), are obtained in the same run. The hysteresis is clearly seen in \( dM/dH \) in Fig. 3(c). These results indicate that the MMT in \( \text{UTe}_2 \) is of first order. A nonzero intercept of a linear extrapolation of \( M(T, H \rightarrow 0) \) from \( H > H_m \) suggests that a finite ordered moment exists in the field-induced phase; as if there is a switch from the PM to FM phase at \( H_m \). Furthermore, the remaining large slope of \( M dM/dH \sim 0.01 \mu_\text{B}/\text{T} \) above \( H_m \) is a mark of surviving heavy quasiparticles in the polarized FM phase. Clearly, the \( b \)-axis becomes the easy-magnetization axis above \( H_m \). As for \( \text{URhGe} \), the magneto-crystalline effect changes drastically with the field. Interestingly, the value of \( M \) occurring the metamagnetic transition for \( H \parallel b \) is close to that showing an anomaly for \( H \parallel a \)-axis, namely \( M \sim 0.5 \mu_\text{B}/\text{f.u.} \). It is quite common in heavy fermion system that for a critical value of \( M \), Fermi surface instabilities associated with enhancement of the magnetic fluctuations occur.\(^{7,15} \)

In order to know the \( T \) evolution of the MMT, we measured the \( M(H) \) curves at different temperatures, as shown in Fig. 3(a). \( \Delta M \) becomes smaller with increasing \( T \). Interestingly, the value of \( H_m \) is almost \( T \)-independent, in stark contrast to the typical itinerant metamagnets locating near the FM critical point.\(^{16} \) Figure 3(b) presents the \( M(T) \) curves at constant various fields replotted from the results in Fig. 3(a). The low-field \( M(T) \) with a broad maximum at \( T_{\text{max}} \) changes into a rapid increase of \( M(T) \) on cooling above \( H_m \). There exists a critical end point (CEP) around 11 K and 34.7 T with a sign change of the \( M(T) \) slope. The signature of the CEP is further confirmed by the differential susceptibility \( dM/dH \) in Figs. 3(c) and (d), which will be discussed later. The field-switch from PM to FM for \( H \parallel b \)-axis resembles to that observed in FM compounds such as \( \text{UGe}_2 \) with the detection of FM wings in \( (T, P, H) \) phase diagram.\(^{16–18} \) The significant differences here are: i) the PM-FM line, \( H_{m}(T) \) is weakly temperature-dependent in \( \text{UTe}_2 \), ii) the phenomena occur for a transverse field with respect to the initial easy-magnetization axis in \( \text{UTe}_2 \), contrary to the case in \( \text{UGe}_2 \) where the FM wing structure is observed for the longitudinal field scan.\(^{17,18} \)

From the \( M \) measurements shown in Figs. 2(b) and 3, the magnetic phase diagram in \( \text{UTe}_2 \) for \( H \parallel b \)-axis is summarized in Fig. 4(a). At 1.4 K, the \( H \)-induced MMT takes place at \( H_m = 34.9 \) T for \( H \)-up sweep measurements. With increasing temperature, \( \Delta H \) decreases and is suppressed to zero at the CEP (11 K and 34.7 T). Above the CEP, the crossover-like broad maximum in \( dM/dH \) appears at \( H_m \), and \( H_m \) decreases with increasing temperature. Since the anomaly in \( dM/dH \) becomes indiscernible above 30 K. Note that the extrapolation to the higher temperature seems to be connected to \( T_{\text{max}} \) at low fields. These results agree with the scaling of \( H_m \) and \( T_{\text{max}} \).\(^{11} \) Thus the MMT and \( T_{\text{max}} \) are dominated by a same single energy scale. We also note that the phase diagram is consistent with the results obtained from magnetoresponse.\(^{19} \)

In order to clarify the evolution of the electronic states
through the MMT, we have analyzed the $M(T)$ data [Fig. 1 (b) for $H \parallel a$ and Fig. 3 (b) for $H \parallel b$] using a thermodynamic Maxwell relation following the previously successful reports, such as in heavy fermion metamagnet CeRu$_2$Si$_2$\cite{20,21} and the reentrant FMSC URhGe.\cite{15} The relation between $M$ and the entropy $S$ is known from the Maxwell relation: $(\partial S/\partial H)_T = (\partial M/\partial T)_H$. At low temperature, $S = \gamma T$ was observed in UTe$_2$.\cite{8,9} From these relations, we can directly access the $H$ dependence of $\gamma$ as, $(\partial \gamma/\partial H)_T = (\partial^2 M/\partial T^2)_H$. Using the $M(T)$ data at various temperatures and assuming that $M(T)$ varies as $T^2$ at low $T$ on the basis of the Fermi liquid state, we have evaluated $\gamma$ as a function of $H$. Figure 4 (b) shows the $M(H)$ curves at 4.2 K and 0 K. The latter was derived by extrapolation to 0 K assuming $M(T, H) = M(0, H) + \beta T^2$, where $\beta$ is a coefficient of $T^2$-dependence of $M$. As expected, $M$ at below (above) $H_{\text{CEF}}$ is smaller (larger) at lower $T$, indicating the reliability of assumption of the $T^2$-dependence. As

![Figure 3](image1.png)

![Figure 4](image2.png)
shown in Fig. 4 (c), \(dy/dH\) for \(H \parallel a\) and \(b\) axes is evaluated using \(dy/dH = \Delta\). In both \(H\)-directions, a peak structure is observed at 6 T and 34.8 T for \(H \parallel a\) and \(b\). The former is in agreement with \(dM/dH\) anomaly [see Fig. 2(a)]. Remarkably, for the \(b\)-axis a very sharp singularity with a drastic sign change is observed at \(H_m\). The change in sign of \(dy/dH\) takes place within an analyzed \(H\)-step of 0.1 T, reflecting a sharp \(M\) jump at \(H_m\). The \(\gamma(H)\) derived by integrating \(dy/dH\) is shown in Fig. 4 (d). The reported value of \(\gamma = 118 \text{ mJ mol}^{-1}\text{K}^{-2}\) is used for \(\gamma(0)\).\(^9\) Of course as the MMT at \(H_m\) is of first order, discontinuity in \(\gamma(H)\) can exist at \(H_m\). The choice of a converging value at \(H_m\) coming from low field \((H < H_m)\) or high field \((H > H_m)\) scan is in agreement with the singularity in the field dependence of the coefficient of \(T^2\)-term of the resistivity \(\rho(H)\),\(^9\) which often scales \(\gamma^2\) in heavy fermion compounds. \(\gamma(H)\) for \(H \parallel a\) weakly depends on \(H\) and shows a small minimum at 6 T, which are in agreement with the \(A(H)\).\(^9\) As seen in other heavy fermion metamagnets, a clear peak in \(\gamma(H)\) centered at \(H_m\) is observed for \(H \parallel b\)-axis. This fact strongly suggests the development of fluctuations on approaching to \(H_m\). The singularity of \(\gamma(H)\) at the first order transition \(H_m\) demonstrates that the dynamic correlation survives through \(H_m\) in agreement with recent work on so called quantum annealed criticality\(^{22}\).

Finally, we discuss the possibility of the \(H\)-reinforced SC in \(\text{UTe}_2\) on going to \(H_m\) for \(H \parallel b\) based on a simple mass enhancement mechanism, which is proposed for the case of the reentrant SC in \(\text{URhGe}\).\(^7,15,23,24\) In similarly to \(\text{UTe}_2\), shown in Fig. 4(d), a mass enhancement was observed in \(\text{URhGe}\) through the \(A(H)\)\(^{23}\) and \(\gamma(H)\)\(^{15}\) coefficients for \(H\) applied hard magnetization \(b\)-axis: a peak appears at \(H_B \sim 12\) T, where FM magnetic moment reorients from the \(c\)-to \(b\)-axis, and reentrant SC becomes more stable than the low-field SC. The total effective mass \(m^*(H)\) can be expressed as,

\[
m^*(H) = m^+(H) + m_B,
\]

where \(m^+(H)\) and \(m_B\) are the correlation and band masses, respectively. As seen in Fig. 4 (d), \(m^* \sim \gamma(H)\) is strongly enhanced at \(H_m\). If the band structure is assumed to be independent of \(H\), namely \(m_B \sim \gamma_0\) and \(\gamma_0 = 61 \text{ mJ mol}^{-1}\text{K}^{-2}\),\(^9\) the development of the fluctuation corresponds to the enlargement of the \(m^*\). Thus, the pairing coupling constant \(\lambda \equiv \frac{\Delta}{m^*}\) is indicated to become larger with \(H\) along the \(b\)-axis.\(^8,9\) Assuming \(m^+(H) \sim \gamma(H) - \gamma_0\), \(\lambda(H)\) is derived as shown in Fig. 5. \(\lambda(H)\) increases with \(H\), in agreement with the previous results.\(^8,9\) Moreover, it does not tend to saturate before \(H_m\). We have estimated \(T_{\text{sc}}\) employing a simplified McMillan-type formula, \(T_{\text{sc}} = T_0 \exp \left( - \frac{\lambda}{m^*} \right)\), where \(T_0\) is a constant and is determined as the experimental \(T_{\text{sc}}\) at 0 T.\(^7,23,24\) As shown in Fig. 5, \(T_{\text{sc}}\) becomes nearly doubled at \(H_m\). Although this model is too rough to describe realistic systems, we believe that it will stimulate further experimental and theoretical studies.

Fermi surface instabilities seem to occur in \(\text{UTE}_2\) for \(H \parallel a\) and \(b\) crossing a critical value of the magnetization. A new ingredient, compared to the previous FMSC cases of UCuGe and \(\text{URhGe}\), is that SC appears in the PM ground state. Thus, arguments used on the field increase of the SC coupling constant \(\lambda\) in transverse field-scan due to the suppression of \(T_{\text{Curie}}\) with field appears to be not relevant here, because no FM has been detected in \(\text{UTE}_2\) so far. A simple image one can consider is that, on cooling the Fermi surface is fully established (below \(T^{\text{max}}\)) and strong Ising fluctuations occur along the \(a\)-axis in the low fields. A field applied along \(b\)-axis lead to drastic change of the orientation of fluctuations as observed in \(\text{URhGe}\).

Metamagnetism in strongly correlated FM systems can have various origins.\(^{25}\) For example, as emphasized earlier, the reentrance of FM just above a critical pressure \(P_c\), in longitudinal field scan is shown in \(\text{UCuGe}\).\(^{17,18}\) The collapse of \(T_{\text{Curie}}\) in transverse field scan accompanied with a switch of magnetocrystalline energy is shown in \(\text{URhGe}\).\(^{26}\) A field-controlled valence transition\(^{26}\) and a Fermi surface reconstruction produced by relative Zeeman decoupling of the subbands\(^{27}\) can be invoked.

A fascinating road is to go deeper in band structure calculation. The first LDA calculation gives the idea that \(\text{UTE}_2\) will be a Kondo semiconductor; a shift of the \(5f\) level can restore the experimental fact that \(\text{UTE}_2\) is rather a good metal at zero field.\(^9\) The magnetic field is an elegant parameter to act on Fermi surface through the change of the \(5f\) configuration and also the valence. Our result must push to progresses in field and pressure effects on \(\text{UTE}_2\), aiming to find the striking difference in the magnetic fluctuations at a similar magnetic polarization along the \(a\) and \(b\)-axes when \(M\) reaches 0.5 T/f.u.

In summary, we have studied magnetization in pulsed-fields up to 56 T for the easy- (hard-) magnetization \(a (b)\) axis of heavy fermion superconductor \(\text{UTE}_2\). A sharp magnetization jump due to the first-order metamagnetic transition is observed for \(H \parallel b\)-axis at \(H_m = 34.9\) T. In addition, a small anomaly in the \(M(H)\) curve for \(H \parallel a\)-axis is observed at \(H \sim 6.7\) T. From careful temperature and field scans of the magnetization, we find that the the first order character terminates at the critical end point (\(\sim 11\) K, 34.8 T). A singularity of the metamagnetic transition is of first order, which is originally induced from the PM ground state. We also propose a possible \(H\)-reinforcement of superconductivity, as indicated by the field dependence of the effective mass derived from a thermodynamic Maxwell relation. \(\text{UTE}_2\) provides a playground to study the interplay of SC, metamagnetism and Fermi surface instabilities. We finally note that the metamagnetic transition in \(\text{UTE}_2\) was also observed by Knafo et
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