

Change of superconducting character in UTe_2 induced by magnetic field

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UTe_2 is a recently discovered spin-triplet superconductor. One of the characteristic features of UTe_2 is a magnetic field (H)-boosted superconductivity > 16 T when H is applied exactly parallel to the b axis. To date, this superconducting (SC) state has not been thoroughly investigated, and the SC properties as well as the spin state of this high- H SC (HHSC) phase are not well understood. In this letter, we performed AC magnetic susceptibility and nuclear magnetic resonance measurements and found that, up to 24.8 T, the HHSC state has bulk nature and is quite sensitive to the H angle and that its SC character is different from that in the low- H SC (LHSC) state. The dominant spin component of the spin-triplet pair is along the a axis in the LHSC state but is changed in the HHSC state along the b axis. Our results indicate that H -induced multiple SC states originate from the remaining spin degrees of freedom.

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Superconductivity occurs when a coherent quantum fluid is formed from electron pairs. For most superconductors, although the total spin (S) of the pairs is in the singlet state ($S = 0$), it is also possible in the triplet state ($S = 1$). Such superconductors, called *spin-triplet superconductors*, are coherent quantum fluids with spin and orbital degrees of freedom. Spin-triplet superconductors would involve rich physics but are very rare. Therefore, the nature of the spin-triplet pairing state was initially studied by analyzing the superfluidity of ^3He [1,2]. The recent discovery of ferromagnetic (FM) superconductors [3–5], in which the ferromagnetism and superconductivity arise from same electrons, has made it possible to study the spin-triplet pairing state in the superconductors. Additionally, a spin-triplet superconducting (SC) candidate UTe_2 has been recently discovered [6]; the SC transition temperature T_c is 1.6–2.0 K [7,8]. Although UTe_2 undergoes no FM transition, it was considered an end member of FM superconductors owing to its physical similarity to FM superconductors [6,7]. However, recent experimental results unveiled the presence of the incommensurate antiferromagnetic fluctuations as well as the FM fluctuations [9,10].

The results of the nuclear magnetic resonance (NMR) Knight-shift (K) measurements to superconductors have provided important information about the spin state in the SC state [11,12]. However, in FM superconductors, this information is obscured because of the internal field produced

by FM-ordered moments. Thus, UTe_2 provides a special opportunity for studying spin-triplet physics because the lack of FM moments means that precise K measurements can be obtained.

UTe_2 crystallizes in the $Immm$ space group (#71, D_{2h}^{25}). The possible SC symmetry and irreducible representation of spin-triplet superconductivity corresponding to the orthorhombic crystal structure of UTe_2 in the zero field and $H \parallel b$ are listed in Tables I and II, respectively [7,13].

As a result of performing the NMR Knight-shift measurements under low external fields [14–17], we found that UTe_2 is a spin-triplet superconductor with spin degrees of freedom. The important aspect to be clarified is the behavior of the remaining spin degrees under various experimental conditions, such as the application of a magnetic field and/or pressure.

The upper critical field of superconductivity (H_{c2}) is strongly directionally dependent [18,19]. When H was perfectly aligned along the b axis, H -boosted superconductivity was observed up to ~ 35 T [6,18–21]. The use of microscopic measurements to investigate this high- H SC (HHSC) state is critical for understanding the nature of spin-triplet superconductivity as well as the SC mechanism of UTe_2 .

A ^{125}Te -enriched single crystal $5 \times 3 \times 1$ mm³ in size and with $T_c \sim 1.67$ K was prepared by applying a chemical vapor transport method [7]. The characterization of the present sample is described in the Supplemental Material (SM) [22]. Figure 1(a) shows the ^{125}Te -NMR spectra for $H \parallel b$, which are plotted against $K = (f - f_0)/f_0$. Here, f is the NMR frequency, and f_0 is the reference frequency determined as $f_0 = (^{125}\gamma_n/2\pi)\mu_0H$ with a ^{125}Te -nuclear gyromagnetic ratio $^{125}\gamma_n/2\pi = 13.454$ MHz/T. As shown in Fig. 1(b), there

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TABLE I. Classification of the odd-parity SC order parameters for point group with D_{2h} in a zero field. The irreducible representation (IR) and its basis functions are listed. The dominant spin component in the SC state is also shown.

D_{2h} (zero field)	Basis functions	SC spin comp.
A_u	$k_a\hat{a}, k_b\hat{b}, k_c\hat{c}$	
B_{1u}	$k_b\hat{a}, k_a\hat{b}$	c
B_{2u}	$k_a\hat{c}, k_c\hat{a}$	b
B_{3u}	$k_c\hat{b}, k_b\hat{c}$	a

are two crystallographically inequivalent Te sites, $4j$ and $4h$, with the Te sites $mm2$ and $m2m$ in UTe_2 ; these Te sites are denoted as Te1 and Te2 sites, respectively. Correspondingly, we observed two ^{125}Te NMR peaks, as has been reported previously [24]. An NMR peak with a smaller [larger] K in $H \parallel b$ was assigned as a Te(1) [Te(2)] peak, in accordance with a previous study [15,24].

For the accurate alignment of the sample, we utilized the Te(1)-NMR shift as an angle marker and an NMR probe with a two-axis rotator. The two angles θ and ϕ have been defined as shown in Fig. 1(c); the sample orientation was adjusted by tuning θ and ϕ such that the Te(1) shift became the minimum value, as shown in Figs. 1(d) and 1(e). The accuracy of the alignment was estimated to be $\pm 0.2^\circ$ for θ and $\pm 0.5^\circ$ for ϕ , where θ (ϕ) is the angle between the b and a (c) axes. The details of how to align the samples are described in the SM [22].

To confirm the SC phase diagram, we measured the tuning frequency (ν_{tune}) and radiofrequency reflection coefficient for the NMR tank LC circuit using a vector network analyzer, where L and C are the inductance and capacitance, respectively. Here, $\nu_{\text{tune}} \sim 1/\sqrt{LC}$ is a good measure for tracking the superconductivity because the inductance of the NMR coil with the sample, i.e., $L = L_0(1 + q\chi_{AC})$, where q is the filling factor, changes at the SC onset. Thus, the change in AC susceptibility (χ_{AC}) due to SC diamagnetism can be detected *in situ* by measuring the change in ν_{tune} across T_c or H_{c2} .

Figure 2(a) shows the variation in $-\Delta\nu/\nu_{\text{tune}}$, as measured by sweeping H at 1.5, 1.0, and 0.6 K. At 1.5 and 1.0 K, the SC transitions were indicated by sudden decreases in the $-\Delta\nu/\nu_{\text{tune}}$, as shown by the arrows. Although UTe_2 is in the SC state at 0.6 K, $-\Delta\nu/\nu_{\text{tune}}$ was found to exhibit a characteristic H dependence. Increasing $H > 14$ T corresponded to sharp decreases in $|\Delta\nu/\nu_{\text{tune}}|$; however, further increase beyond 16.5 T coincided with increasing $|\Delta\nu/\nu_{\text{tune}}|$, indicating a kink at $H_{\text{kink}} \sim 16.5$ T. Figure 2(b) shows the T

TABLE II. Classification of odd-parity SC phases occurring in UTe_2 under a b -axis magnetic field. The typical order parameters belonging to each IR are listed in Table I.

IR of C_{2h} (under field)	$A_u^{H\parallel b}$	$B_u^{H\parallel b}$
H direction	$A_u^{H\parallel b}$	$B_u^{H\parallel b}$
$H \parallel b$	$A_u + iB_{2u}$	$B_{3u} + iB_{1u}$

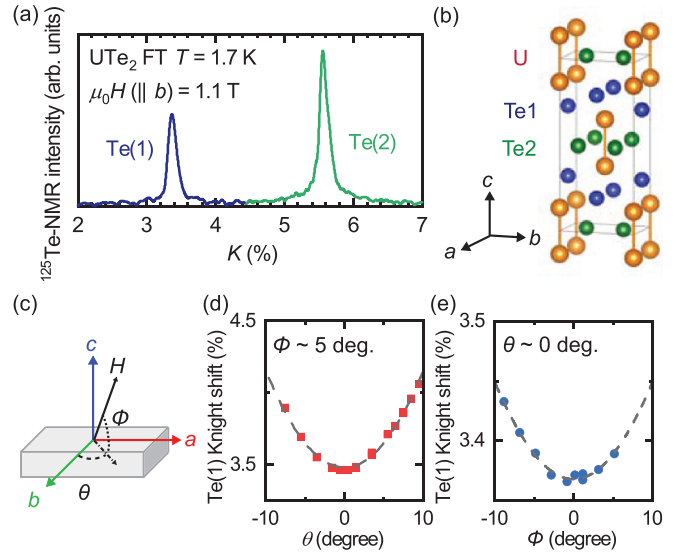


FIG. 1. (a) ^{125}Te -nuclear magnetic resonance (NMR) spectra measured in $H \parallel b$. An NMR peak with the smaller [larger] K is called Te(1) [Te(2)] in this letter. (b) The crystal structure of UTe_2 [23]. There are two Te sites in UTe_2 . (c) Definition of the angles θ and ϕ against the crystalline axes in UTe_2 . (d) [(e)] The θ [ϕ] dependence of the K in the Te(1) peak. The b axis [$(\theta, \phi) = (0, 0)$] is determined from the minimum of K at the Te(1) site.

dependence of $-\Delta\nu/\nu_{\text{tune}}$ for $\mu_0 H = 7.5, 15.5, 16.5$, and 24 T on cooling.

The minimum value of $|\Delta\nu/\nu_{\text{tune}}|$, in relation to SC diamagnetism, was observed at 16.5 T, consequently demonstrating the same tendency as T_c . This indicates that the HHSC state has bulk properties of UTe_2 . The H and T dependencies of $-\Delta\nu/\nu_{\text{tune}}$ suggest that the SC character changed at $\mu_0 H_{\text{kink}} \sim 16.5$ T. In fact, the HHSC state was found to be very sensitive to the angle θ . Superconductivity was observed within $\pm 3^\circ$ at 24 T, as shown in Fig. 2(c), and the unexpected minute θ rotation ($\theta \sim 4^\circ$) that occurred during the experiments completely suppressed the HHSC, although the SC diamagnetism in the low- H SC (LHSC) state was nearly unchanged, as shown by the dotted curve in Fig. 2(a). This result is in good agreement with the results presented in previous reports [18,19]. Based on the T - and H -scan measurements of $-\Delta\nu/\nu_{\text{tune}}$, we developed the SC phase diagram shown in Fig. 2(d). The H_{kink} anomaly, like the overall phase diagram, is consistent with the phase transitions determined by the recent specific heat measurements [25]. Because the responses to H and θ are different between the LHSC and HHSC states, it is reasonable to consider that the kink at H_{kink} marks a phase transition between the two SC states. Such a transition between two bulk SC states was also confirmed in the work [25] by linear magnetostriction and thermal dilatation, evidencing anomalies due to vortex pinning in both phases. As shown in Fig. 2(a), the results of the H -sweep measurement at 0.6 K revealed clear hysteresis behavior at $\mu_0 H^* \sim 4$ T; it was found to be related to an anomaly of the vortex state because the anomaly was not observed in the H dependence results for the electronic term in specific heat measurements [26]. The details of this anomaly have been studied and will be reported in a separate paper.

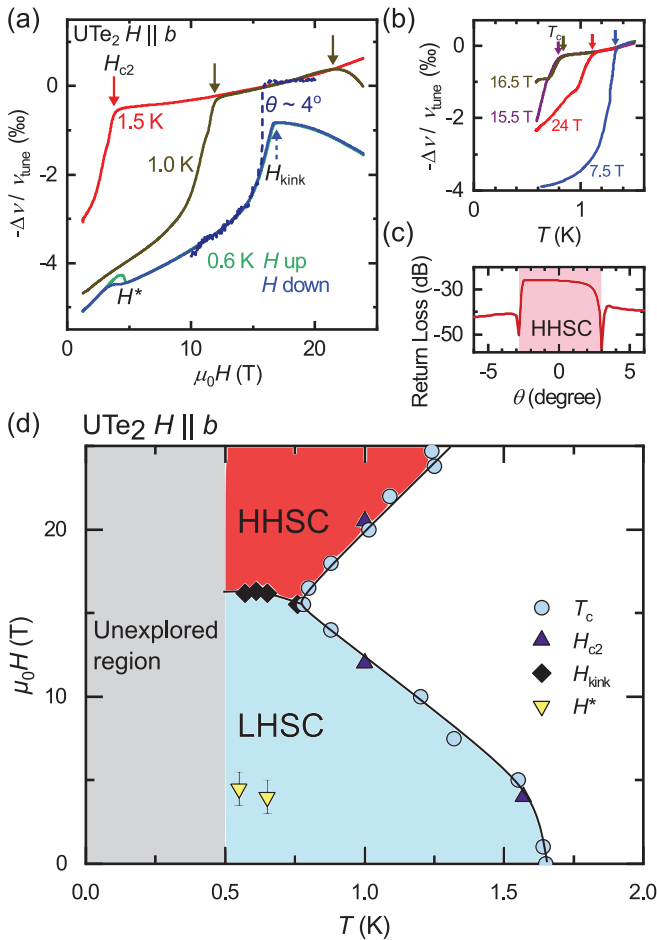


FIG. 2. (a) H dependence of $-\Delta\nu/\nu_{\text{tune}}$ in relation to the AC magnetic susceptibility χ_{AC} , up to 24.8 T, as measured at 0.6, 1.0, and 1.5 K. At 0.6 K, $-\Delta\nu/\nu_{\text{tune}}$ exhibited a kink at H_{kink} , denoted the dotted arrow; the H dependence of $-\Delta\nu/\nu_{\text{tune}}$, which was determined by performing H -up and H -down sweeps, is shown. The dotted curve shows the H dependence of $-\Delta\nu/\nu_{\text{tune}}$ when the minute θ rotation unexpectedly occurred in the sample. The solid arrows denote H_{c2} . (b) Temperature dependence of $-\Delta\nu/\nu_{\text{tune}}$ as measured at 7.5, 15.5, 16.5, and 24 T. T_c in the field is denoted by the arrow with the same color. (c) Angle dependence of the return loss of the nuclear magnetic resonance (NMR) tank circuit at 24 T. When the sample was in the superconducting (SC) state, the quality factor of the circuit Q was lower owing to the change in the impedance of the circuit. High- H superconductivity was observable within $\pm 3^\circ$. (d) H - T phase diagram determined by performing T - and H -scan measurements of $-\Delta\nu/\nu_{\text{tune}}$.

To investigate the SC properties, particularly the spin susceptibility in the HHSC state, we performed ^{125}Te NMR measurements at the Te(2) peak with a larger K . Figures 3(a) and 3(b) show the Te(2)-NMR spectra measured at various temperatures < 2.5 K at 1 and 24 T, respectively. At 1 T, the single-peak spectrum gradually shifted to the low- K side in the normal state and sharply shifted immediately below T_c ; this was accompanied by spectrum broadening. The ^{125}Te NMR spectrum measured under conditions of 24 T and 2.5 K revealed a double-peak structure that is attributable to its high resolution; the right peak was found to have a 0.04% larger

K than the main peak. The H dependence of the Te(2)-NMR spectrum is shown in the SM [22]. Several possibilities were considered for the origin of the larger K peak; they include the occurrence of a mosaic structure and/or minute U-atom deficiency in the single-crystal sample. In the former case, the misalignment of the mosaic was estimated to be 2.0° (8.1°) on the a (c) axis; additionally, the ^{125}Te -NMR measurements for the higher T_c samples are critical for the latter possibility because T_c seems to be very sensitive to a U-atom deficiency [7,8,27]. Further experiments are required to clarify the origin of the larger K peak. As T was decreased, the two peaks gradually shifted to the lower K side in the same manner. Because the resolution of the higher K peak is not sufficient for analysis, we focus on the main peak shown by the arrows.

Figure 3(c) shows the T dependence of K of the main peak, as determined from the NMR spectra measured at 1, 10, 15, 20, and 24 T. A decrease in K below T_c was clearly observed for 1 and 10 T; the magnitude and H dependence of the K decrease below T_c are in agreement with previous results [15,16]. In contrast, at 20 and 24 T, K gradually decreased without any appreciable anomaly at $T_c(H)$. To quantify the Knight-shift decrease (ΔK) ascribed to the superconductivity, the normal-state T dependence was subtracted from the observed K_b and ΔK was plotted, as shown in the inset of Fig. 3(c). Here, ΔK was near zero above 15 T, although this field is still in the LHSC. This behavior is consistent with previous NMR measurements [16]. A similar $\Delta K \sim 0$ trend was previously observed in the a -axis Knight-shift measurement results for the LHSC state, where the dominant SC spin component oriented along the a axis [17]. Thus, these results indicate that b -axis spin-polarized superconductivity is induced by a b -axis magnetic field.

We will now discuss possible SC states in the HHSC region. Considering the observed spin-susceptibility and field-boostered behavior, the ground state of the HHSC is $A_u^{H\parallel b}$, as presented in Table II; this is because the SC spin component is parallel to $H \parallel b$ in the HHSC region. This is consistent with the theoretical suggestion [13,28]. Although ΔK changes smoothly, the kink anomaly in the field dependence $-\Delta\nu/\nu_{\text{tune}}$ implies a phase transition between the HHSC and LHSC states; thus, the LHSC state is determined to be $B_u^{H\parallel b}$. These results strongly support the B_{3u} scenario at a low-field limit [14–17]. Under $H \parallel b$, B_{3u} at zero field becomes $B_u^{H\parallel b}$ with crossover (without any transition). Note that, for thermodynamic limitation, the tricritical point with three second-order phase transitions is not allowed. Thus, the phase transition line inside the SC region should be first order, or there is a hidden phase transition line with second-order phase transition [13]. The results of the crude up-down H -sweep measurement of $-\Delta\nu/\nu_{\text{tune}}$ at 0.6 K revealed the occurrence of one kink without any hysteresis near H_{kink} [Fig. 2(b)]; this seems to exclude the first-order phase transition scenario. Rather, we suggest the presence of another phase transition line characterized with the SC properties of the HHSC state such as $\Delta K = 0$. Further precise NMR measurements are required to understand the relationship between the LHSC and HHSC phases.

In addition, it is noteworthy that the enhancement of the superconductivity against H was found to be stronger than that previously reported [6,18,21]. Because the value of T_c at $H =$

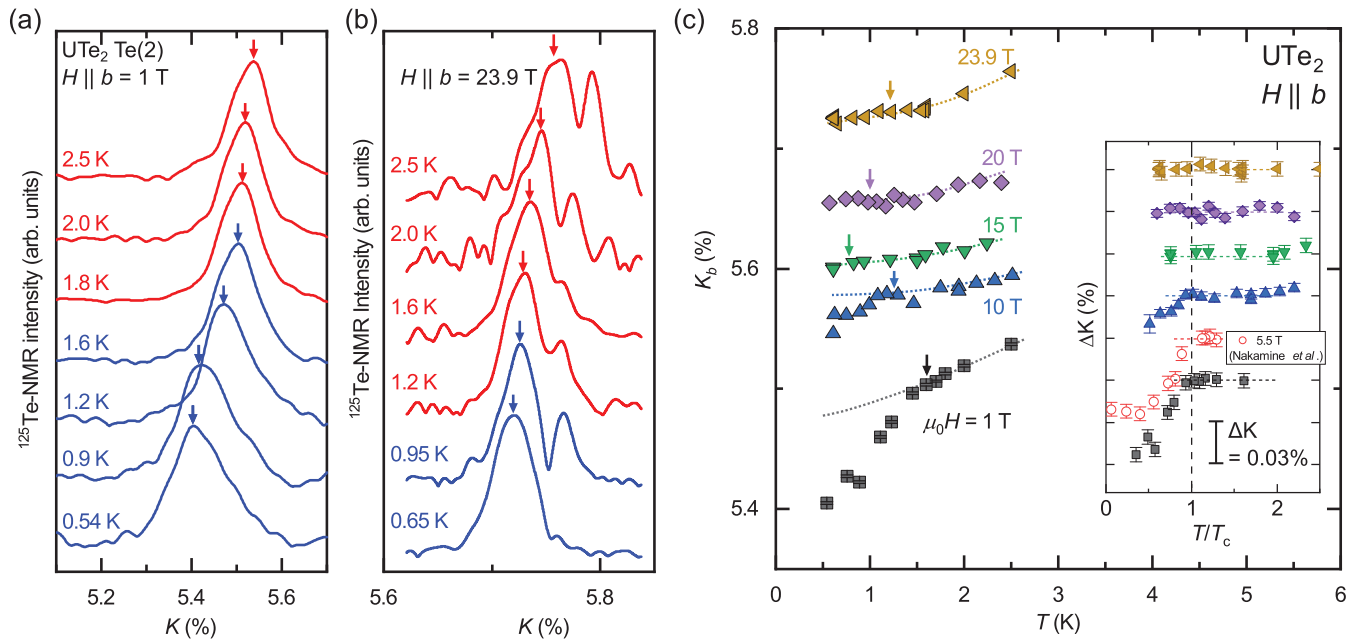


FIG. 3. Te(2)-nuclear magnetic resonance (NMR) peaks measured at various temperatures < 2.5 K at (a) ~ 1 T and (b) ~ 24 T. (c) Temperature dependence of K , as determined by analyzing the Te(2)-NMR peak shown by the arrow. The dotted line is the normal-state behavior extrapolated using the second-order polynomial function as explained in the Supplemental Material [22]. (inset) Temperature dependence of the change in the Knight shift from the normal state. The horizontal dashed line represents the baseline, and the vertical dashed line represents the transition temperature.

0 for the current sample (1.67 K) was slightly higher than that of previous samples (~ 1.5 K), the upturn behavior is seemingly dependent on the sample quality, suggesting the intrinsic properties of UTe_2 . A similar level of superconductivity robustness by $H \parallel b$ was observed in the FM superconductors URhGe [29] and UCoGe [30], in which critical FM fluctuations were determined to play an important role [31–34]. Because superconductivity occurs in the paramagnetic state of UTe_2 , such critical FM fluctuations were not anticipated. Alternatively, we speculate that the critical fluctuations related to the incommensurate antiferromagnetic fluctuations [9,10], which may be induced by $H \parallel b > 16.5$ T, play an important role in the mechanism governing the HHSC state. It is interesting that the SC pairing interaction can be tuned by adjusting H applied along the b axis; this seems to be a common feature of U-based FM and nearly FM superconductors with Ising anisotropy under normal-state magnetic conditions, although the SC pairing interaction is not clarified in UTe_2 .

In conclusion, we have determined from the results of *in situ* χ_{AC} and NMR measurements at magnetic field strengths up to 24.8 T that the HHSC state has bulk properties of UTe_2 and that the spin component of the triplet pair orients along the b axis in the HHSC state, which is different from that in the LHSC state. The results presented here provide decisive evidence that the spin degrees in a spin-triplet pair

can be controlled by an external magnetic field H . This is a unique phenomenon that is not expected in spin-singlet superconductors but is inherent to spin-triplet superconductors. Exploring unique phenomena related to the spin degrees of freedom in spin-triplet superconductors is important because this information can facilitate their application. This study is currently in progress.

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