

Enhancement of antiferromagnetic spin fluctuations in UTe_2 under pressure revealed by ^{125}Te NMRDevi Vijayan Ambika,^{1,2} Qing-Ping Ding¹, Corey E. Frank^{3,4}, Sheng Ran,⁵ Nicholas P. Butch^{3,4} and Yuji Furukawa^{1,2,*}¹*Ames National Laboratory, U.S. DOE, Ames, Iowa 50011, USA*²*Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA*³*Department of Physics, Maryland Quantum Materials Center, University of Maryland, College Park, Maryland 20742, USA*⁴*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*⁵*Department of Physics, Washington University, St. Louis, Missouri 63130, USA*

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Characterizing magnetic fluctuations is one of the keys to understanding the origin of superconductivity in the spin-triplet superconductor UTe_2 , which exhibits two superconducting (SC) phases (SC1 and SC2) under pressure: SC1 where the superconducting transition temperature of T_c decreases with pressure, whereas T_c of SC2 rises with pressure. Previously, D. Ambika *et al.* [Phys. Rev. B **105**, L220403 (2022)] reported the possible coexistence of ferromagnetic (FM) and antiferromagnetic (AFM) spin fluctuations in UTe_2 under pressure from their nuclear magnetic resonance (NMR) measurements. To delve into the relationship between the magnetic fluctuations and the two SC phases, we have carried out detailed ^{125}Te NMR measurements on a single crystal of UTe_2 with $T_c = 1.6$ K at various pressures ranging from 0 to 2.05 GPa. By comparing the temperature T dependence of nuclear spin-lattice relaxation rates divided by temperature $1/T_1T$ with that of the Knight shift K for magnetic fields along the a , b , and c directions, we evidence the enhancement of AFM spin fluctuations with increasing pressure. Based on the results, we suggest that FM spin fluctuations are more favorable for SC1 and AFM spin fluctuations are crucial for SC2. Our findings will inspire further study on this material to understand the peculiar SC phases in detail.

DOI: [10.1103/4gcm-mbl7](https://doi.org/10.1103/4gcm-mbl7)**I. INTRODUCTION**

The discovery of superconductivity in UTe_2 with a superconducting transition temperature T_c of ≈ 1.6 K [1,2] and now raised up to ≈ 2.1 K [3,4] has triggered intense research activity to characterize the physical properties of the compound, as it has been suggested to be a possible candidate for spin-triplet superconductors [1,2,5,6]. The spin-triplet superconducting (SC) state is suggested from a highly anisotropic upper critical field H_{c2} that exceeds the Pauli limit [1,2,5,6], and also from a tiny reduction of the Knight shift (K) below T_c in all three crystallographic directions [7–13]. In addition, recent phase-sensitive measurements utilizing the Josephson effect have also supported the spin-triplet superconducting state [14].

One of the key parameters to characterize materials, especially for the mechanism of superconductivity, is magnetic fluctuation. Ferromagnetic (FM) fluctuations have been initially considered to play an important role in the triplet pairing, as suggested by nuclear magnetic resonance (NMR) measurements [15,16] where Ising-like FM fluctuations along the a axis were reported. At the same time, antiferromagnetic (AFM) spin fluctuations with the incommensurate wave-vector of $q = (0, 0.57, 0)$ was also revealed by neutron scattering (NS) measurements [17–20], suggesting the importance of the AFM spin fluctuations as well as the FM fluctuations, although more recent NS measurements suggest

interband spin excitons arising from f -electron hybridization as a possible origin of the magnetic excitations [21,22]. Recent NMR measurements also pointed out the existence of both FM and AFM fluctuations at ambient pressure [23,24].

UTe_2 also exhibits many intriguing properties under pressure. With the application of pressure, two SC phases appear around 0.25 GPa [25,26]. As depicted in Fig. 1, the SC phase with lower T_c (SC1) is suppressed continuously with increasing pressure. In contrast, the T_c of the SC2 phase is enhanced, takes a maximum $T_c \approx 3$ K at around 1.2 GPa, and then the SC2 phase is suppressed rapidly at higher pressures around $p_c \approx 1.5$ GPa. Above the critical pressure p_c , an AFM magnetic ordered phase appears [26,27,30] where two phases were suggested: weak magnetic order (WMO) state below T_{WMO} and long-range magnetic order state below T_{MO} as illustrated in Fig. 1.

Previously, we reported from NMR measurements on a single crystal of UTe_2 at pressures from 0 to 1.57 GPa under a magnetic field H parallel to the b axis, a possible coexistence of FM and AFM spin fluctuations under pressure, with the AFM spin fluctuations becoming enhanced with increasing pressure [28]. Recent theoretical studies also point out that the interplay between these spin fluctuations is pivotal in understanding the emergence of multiple SC phases [31,32], therefore, it is crucial to investigate the evolution of spin fluctuations in detail under varying pressure conditions.

In this paper, we carried out ^{125}Te NMR measurements on a ^{125}Te -enriched single crystal of UTe_2 under magnetic fields H parallel to the three crystalline axes a , b , and c at various

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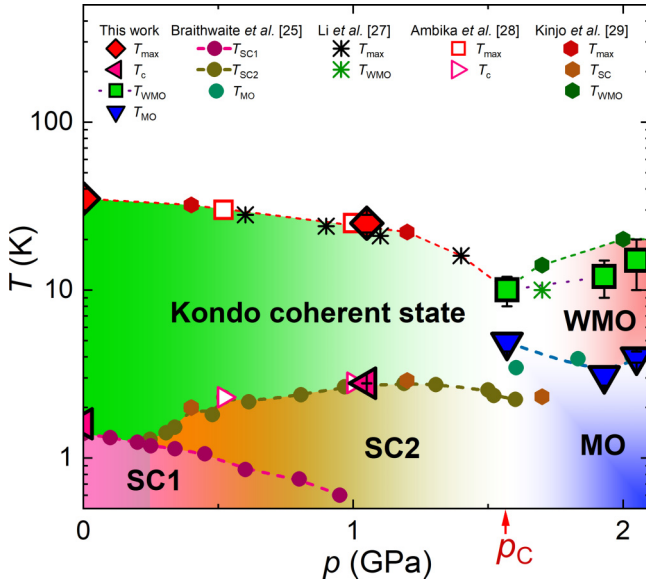


FIG. 1. Pressure-temperature (p - T) phase diagram of UTe_2 . T_{max} is the temperature at which a broad maximum is observed in the temperature dependence of the magnetic susceptibility or Knight shift K under $H \parallel b$ reported from Refs. [27–29], T_c is the superconducting critical temperature from Refs. [25,28,29], T_{WMO} is the weakly magnetic ordered temperature from Refs. [27,29], T_{MO} is the magnetic ordering temperature from Refs. [27,29]. The slightly larger filled symbols are the corresponding temperatures from the present NMR measurements (see text). Note that, although the SC1 phase inside the SC2 phase, shown in the figure, has been reported to be different from SC1 from the NMR measurements (named SC3) [13], we write it with one SC phase (SC1) for simplicity.

pressures ranging from 0 to 2.05 GPa to investigate the details of how the magnetic fluctuations change with pressure. From the results of nuclear spin-lattice relaxation rate divided by temperature $1/T_1T$ measurements under the $H \parallel a$, b , and c directions in comparison with the Knight shift K results, we found the enhancement of AFM spin fluctuations under pressure, where the hyperfine field fluctuations associated with the AFM fluctuations are found to mainly enhance along the a and c axes at the Te sites. Based on the analysis of the data, we suggest that FM spin fluctuations are more favorable for SC1 and AFM spin fluctuations are crucial for SC2.

II. EXPERIMENTAL DETAILS

The ^{125}Te -enriched single crystal of UTe_2 with $T_c = 1.6$ K (at $H = 0$ T) at ambient pressure was synthesized by the chemical vapor transport method using iodine as the transport agent [1]. NMR measurements of ^{125}Te (nuclear spin $I = \frac{1}{2}$, gyromagnetic ratio $\gamma_N/2\pi = 13.454$ MHz/T) nuclei were conducted using a laboratory-built phase-coherent spin-echo pulse spectrometer for the pressures 0, 1.05, 1.93, and 2.05 GPa with a NiCrAl/CuBe piston-cylinder cell using Daphne 7373 as the pressure-transmitting medium, which guarantees high hydrostaticity up to 2.2 GPa at room temperature [33]. The calibration of pressure was accomplished by ^{63}Cu nuclear quadrupole resonance in Cu_2O [34,35] at 77 K. The superconducting transition temperature $T_c = 2.8$ K

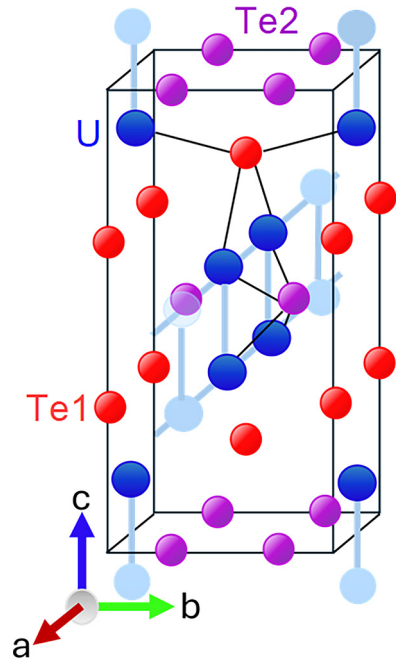


FIG. 2. Crystal structure of UTe_2 . The U atoms (blue circles) form a two-leg ladder structure along the a axis. Two inequivalent tellurium (Te) sites are indicated as Te1 (red circles) and Te2 (violet circles). The light blue circles representing the U atoms are added to show the two-leg ladder structure.

for 1.05 GPa at zero magnetic field ($H = 0$) is determined in the ac susceptibility measurements using an NMR tank circuit. The ^{125}Te -NMR spectra were obtained by sweeping H at fixed NMR frequencies f under H parallel to the a , b , and c axes where the misalignment of the crystal is expected to be less than a few degrees. The $1/T_1$ was measured with a saturation recovery method. $1/T_1$ at each T was determined by fitting the nuclear magnetization M versus time t using the exponential function $1 - M(t)/M(\infty) = e^{-(t/T_1)}$ for ^{125}Te NMR, where $M(t)$ and $M(\infty)$ are the nuclear magnetization at t after the saturation and the equilibrium nuclear magnetization at $t \rightarrow \infty$, respectively. The NMR data for $H \parallel b$ at $p = 0$ and 1.57 GPa are taken from those reported in our previous paper [28]. The uncertainty in the NMR shift and T_1 was estimated by analyzing the maximum and minimum bounds of scattering of the data for each measurement. Furthermore, uncertainties for derived quantities were determined using error propagation methods.

UTe_2 crystallizes in a body-centered orthorhombic structure with the $Immm$ space group [36], where the U atoms form a two-leg ladder structure with legs along the a axis and rung along the c axis, as shown in Fig. 2 [37]. There are two crystallographic inequivalent Te sites occupying $4j$ and $4h$ sites with local symmetries $mm2$ and $m2m$, respectively. Following the previous paper [15], these sites are denoted by Te1 and Te2. The Te1 site is located within the distorted tetrahedron formed by two first and two second-nearest-neighboring U atoms, which belong to three different ladders. On the other hand, Te2 is surrounded by the four nearest-neighbor U atoms forming a square-like structure within a ladder. The NMR spectra exhibit two distinct NMR lines, expected from the

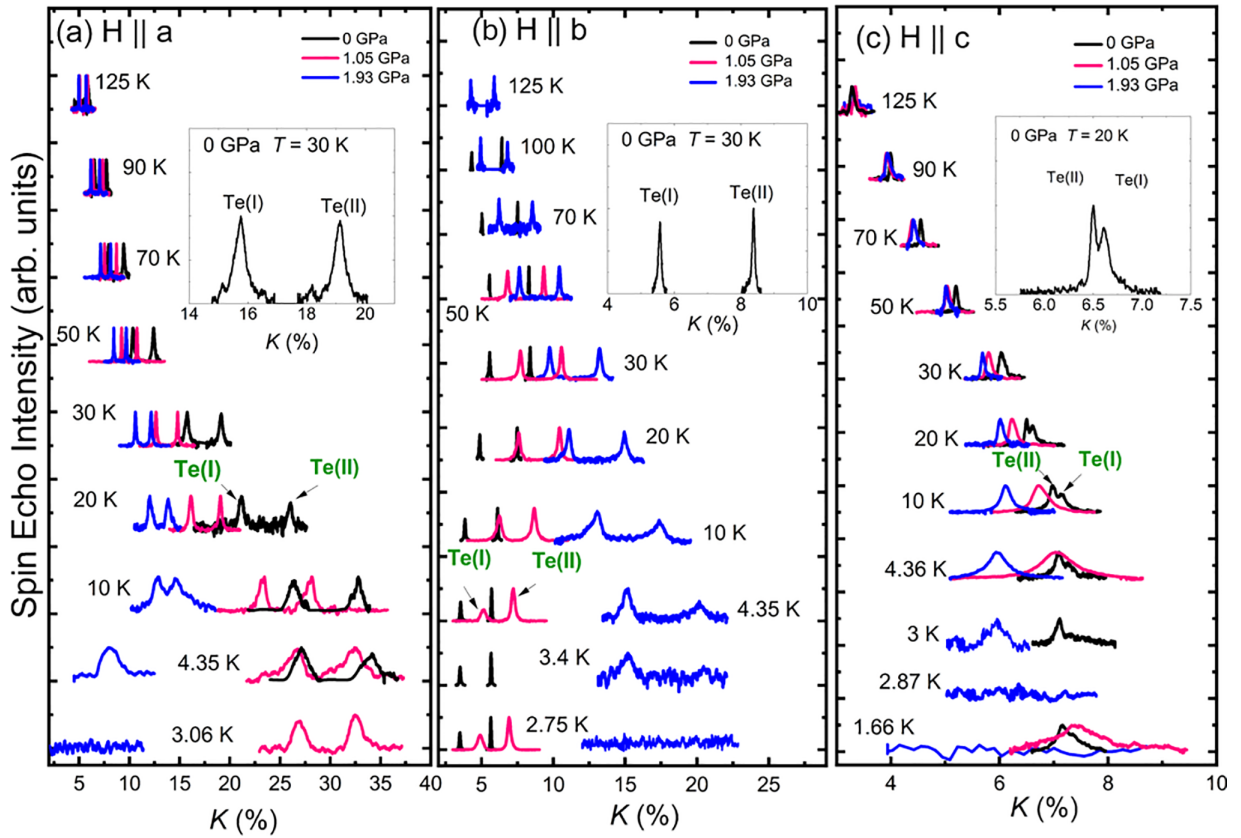


FIG. 3. Temperature dependence of the field-swept ^{125}Te NMR spectra measured on the UTe_2 single crystal for (a) $H \parallel a$ under pressure (0 GPa at $f = 50.35$ MHz, 1.05 GPa at $f = 12.2$ MHz, and 1.93 GPa at $f = 12.2$ MHz); (b) $H \parallel b$ (0 GPa at $f = 41$ MHz, 1.05 GPa at $f = 12.2$ MHz, and 1.93 GPa at $f = 55.6$ MHz); and (c) $H \parallel c$ (0, 1.05, and 1.93 GPa at $f = 50.35$ MHz, respectively). The horizontal axis is the Knight shift K defined by $K = (H_0 - H)/H$, (where $H_0 = 2\pi f/\gamma_N$, f is the NMR resonance frequency, and H is the external magnetic field).

two Te sites, Te1 and Te2 [15]. The specific assignment of these lines to each Te site has remained to be determined [23]. Thus, we designate the peaks as Te(I) and Te(II), following the nomenclature used in the previously published paper [15,23]. However, in this paper, we argue that Te(I) and Te(II) are most likely assigned to the Te1 and Te2, respectively, in the crystal structure as reported in the Supplemental Material (SM) [38]. This is the same with the initial site assignment of Te NMR lines in Ref. [15], and Fujibayashi *et al.* also pointed out such a possibility in Ref. [23].

III. RESULTS AND DISCUSSION

A. ^{125}Te NMR spectrum

Figures 3(a), 3(b), and 3(c) show the temperature dependence of the H -swept ^{125}Te -NMR spectra of a single crystal UTe_2 for three different magnetic field directions parallel to the a ($H \parallel a$), the b ($H \parallel b$) and the c ($H \parallel c$) axes, respectively, under different pressures of ambient, 1.05, and 1.93 GPa. The horizontal axes are Knight shift K defined by $K = (H_0 - H)/H$, where H_0 is the Larmor field given by $2\pi f/\gamma_N$ with f being the NMR resonance frequency.

At ambient pressure, two distinct ^{125}Te -NMR lines for $H \parallel a$ and $H \parallel b$ and two partially overlapped peaks for $H \parallel c$ are observed, consistent with the previous reports [15,23]. Following the previous NMR papers [15,23], the two lines with

smaller and greater K for $H \parallel a$ and $H \parallel b$ are denoted to Te(I) and Te(II), respectively, and the line with the larger K value under $H \parallel c$ is assigned to Te(I), as shown in Figs. 3(a), 3(b), and 3(c). At 1.05 GPa below the critical pressure $p_c \approx 1.5$ GPa, similar spectra, but slightly different K values have been observed.

In contrast, at $p = 1.93$ GPa above p_c , although similar spectra with slightly different K were observed at higher temperatures above ≈ 40 K, quite different temperature dependencies of NMR spectra and K were observed below that temperature. In particular, no signals were observed below ≈ 3 K for all H directions. This is due to the pressure-induced long-range magnetic ordering above p_c [27,29,30]. Furthermore, we also notice that, for $H \parallel b$, the signal intensity of Te(II) starts decreasing below ≈ 10 K, and only the signal of Te(I) can be well observed at 3.4 K as shown in Fig. 3(b). This could be due to a short-range (or weakly) magnetic ordered state. The intensity difference between the two Te sites we observe for 0 and 1.05 GPa for $H \parallel b$ is confirmed to be due to the difference in the nuclear spin-spin relaxation rate. We discuss the temperature dependence of the NMR spectra for $H \parallel b$ above p_c in more detail below.

Figure 4 depicts the temperature dependence of ^{125}Te Knight shift K for both Te sites for all the measured pressures and H directions. The K values of Te(II) are greater than those of Te(I) for the a and b axis directions. This is due

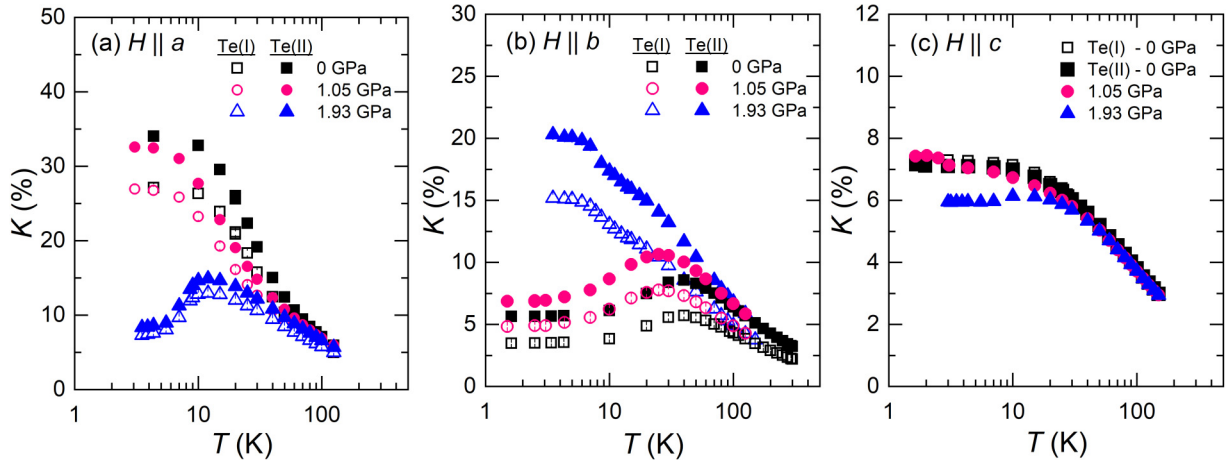


FIG. 4. T dependencies of K s for both Te(I) and Te(II) under different pressures (ambient pressure in black, 1.05 GPa in pink, and 1.93 GPa in blue) under H parallel to all three crystalline axes: (a) $H \parallel a$, (b) $H \parallel b$, (c) $H \parallel c$.

to the difference in hyperfine coupling constants, which are estimated to be $A_{aa}[\text{Te(I)}] = 3.8 \text{ T}/\mu_B$ and $A_{aa}[\text{Te(II)}] = 4.7 \text{ T}/\mu_B$ for the a axis, and $A_{bb}[\text{Te(I)}] = 3.5 \text{ T}/\mu_B$ and $A_{bb}[\text{Te(II)}] = 5.2 \text{ T}/\mu_B$ for the b axis, respectively, at ambient pressure, from the so-called K - χ plot analysis (see the SM [38]). In contrast, along the c axis, the K values of Te(I) and Te(II) are nearly the same, and A_{cc} for Te(I) and Te(II) is estimated to be $4.1 \text{ T}/\mu_B$ at ambient pressure [38]. These coupling constants were also found to be nearly independent of p [29].

The temperature dependence of K for $H \parallel a$ at ambient pressure is similar to the results reported previously [23,42] where, with decreasing T , the K values increase and then level off at low temperatures below 10 K. It was reported that the different T dependence between K and χ_a at low temperatures below $\approx 10 \text{ K}$ is due to the U atom defects in low- T_c crystals [42] where χ increases with decreasing temperature due to the defects while K , reflecting the intrinsic behavior of χ , shows a nearly constant behavior at low temperatures. With increasing p , the values of K are slightly suppressed at 1.05 GPa. However, at 1.93 GPa above p_c , a markedly different temperature dependence of K was observed, exhibiting a maximum around 10 K, as shown in Fig. 4(a). This significant change in the T dependence of K under high p is consistent with the magnetic susceptibility measurements [27]. It is noted that the temperature of the broad maximum is close to $T_{\text{WMO}} \approx 12 \text{ K}$, the onset of a weakly magnetic ordered (WMO) state reported in earlier studies [27,29].

Similarly, a drastic change in the T dependence of K for $H \parallel b$ under pressures can be seen in Fig. 4(b). While K under $H \parallel b$ shows broad maxima at $T_{\text{max}} \approx 35\text{--}40 \text{ K}$ at ambient pressure and $T_{\text{max}} \approx 25 \text{ K}$ at 1.05 GPa, consistent with the magnetic susceptibility data [1,2,27], K keeps increasing and saturates below $\approx 8 \text{ K}$ at 1.93 GPa. These results suggest that the magnetic easy axis changes from the a axis to the b axis upon crossing p_c , and is in good agreement with the χ data under pressure [27].

In the case of $H \parallel c$, the application of pressure leads to only a moderate change in the temperature dependence of K , where a slight suppression of K at 1.93 GPa is observed below $\approx 30 \text{ K}$, showing a weak but visible broad maximum around 10–12 K, which is close to $T_{\text{WMO}} \approx 12 \text{ K}$. The above-

discussed values of K for $H \parallel b$ and $H \parallel c$ under pressure are consistent with previously reported results [29].

B. ^{125}Te NMR spectra for $H \parallel b$ above p_c

As described above, the NMR signal intensities of Te(I) and Te(II) exhibit different temperature dependencies under a pressure of 1.93 GPa at low temperatures, but this is not observed below p_c . To further examine this signal-intensity change, we measured the NMR spectra under $H \parallel b$ at a different pressure of 2.05 GPa above p_c . The spectra are presented in Fig. 5(c), together with the previous data at 1.57 GPa [28] and the spectra at 1.93 GPa shown in Figs. 5(a) and 5(b), respectively. As described, in the case of 1.93 GPa, the NMR line of Te(II) begins to diminish below $\approx 10 \text{ K}$, and only the Te(I) line can be well observed at 3.5 K, and then both signals disappear between 2.75 and 3.5 K, as shown in Fig. 5(b) [also Fig. 3(b)]. A similar behavior is observed at 2.05 GPa where the Te(II) signal intensity starts decreasing below $\approx 10 \text{ K}$ and both signals disappear between 3.7 and 4.36 K. In contrast, at 1.57 GPa, although the signal intensities of Te(II) slightly decrease with respect to that of Te(I) below 10 K, both signals remain present at 5.5 K, and both signals disappear at nearly the same temperature of 4.36 K. As mentioned above, the disappearance of both signals is due to the pressure-induced long-range magnetic order above p_c . Based on these results, we estimate the long-range magnetic ordering temperature T_{MO} to be $\approx 4.9(6) \text{ K}$ at 1.57 GPa, $\approx 3.14(4) \text{ K}$ at 1.93 GPa, and $\approx 4.0(3) \text{ K}$ at 2.05 GPa.

The disappearance of the Te(II) line above T_{MO} could be due to the short-range magnetic ordered state. To better understand the difference in the change of the signal intensities of the two signals, we plotted the ratio of the signal intensity of Te(II) to that of Te(I) as a function of temperature, where the signal intensities were corrected by the nuclear spin-spin relaxation time T_2 . As shown in Fig. 5(d), the ratio clearly begins to decrease below $\approx 10(3) \text{ K}$ for 1.57 and 1.93 GPa and $\approx 15(5) \text{ K}$ for 2.05 GPa, unlike 0 GPa and 1.05 GPa, where the ratio stays close to one. Those temperatures are close to the value of $T_{\text{WMO}} \approx 9.5 \text{ K}$ at 1.7 GPa reported from the magnetic susceptibility measurements [27].

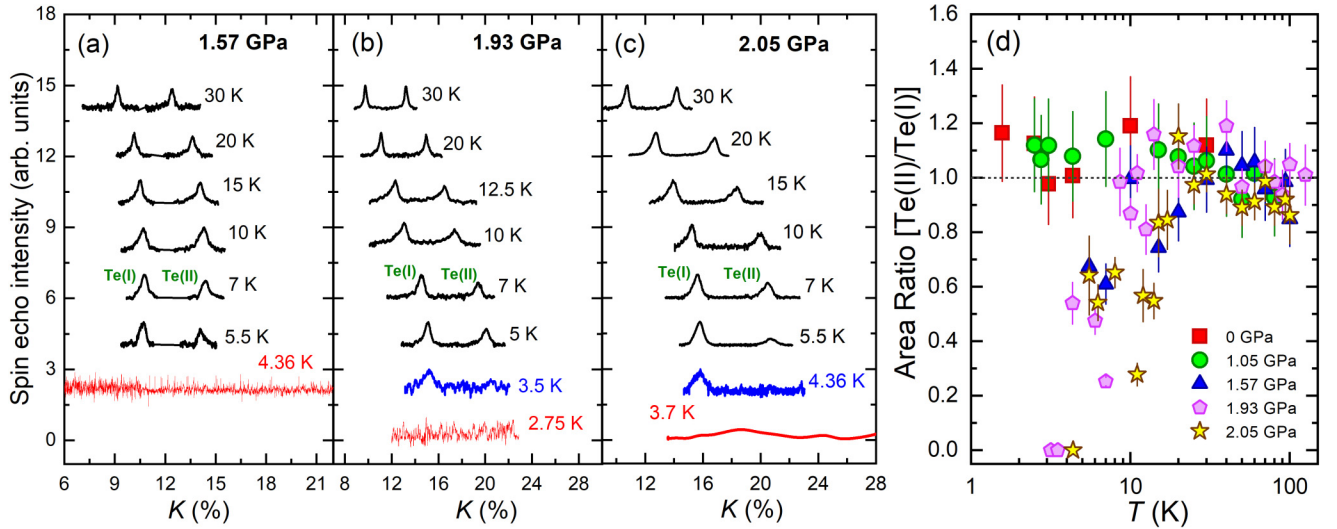


FIG. 5. Temperature dependence of the H -swept ^{125}Te -NMR spectra of a single crystal UTe_2 for (a) $p = 1.57$ GPa, (b) 1.93 GPa, and (c) 2.05 GPa for $H \parallel b$ where the horizontal axis is the Knight shift K . For $p = 1.57$ and 2.05 GPa, the spectra were measured at $f = 50.35$ MHz ($H_0 = 3.74$ T). $f = 55.60$ MHz ($H_0 = 4.132$ T) was used at $p = 1.93$ GPa. (d) Temperature dependence of the signal intensity ratio of Te(II) to Te(I) for $p = 0, 1.05, 1.57, 1.93,$ and 2.05 GPa. Here, the data for 1.57 GPa are from Ref. [28].

C. ^{125}Te spin-lattice relaxation rate $1/T_1$

To shed light on the evolution of the magnetic fluctuations with pressure, we measured the ^{125}Te spin-lattice relaxation rate ($1/T_1$) at the peak positions of the Te(I) and Te(II) lines at $p = 0, 1.05,$ and 1.93 GPa under three different magnetic field directions ($H \parallel a, H \parallel b,$ and $H \parallel c$), whose results are summarized in Figs. 6(a)–6(i). A general trend is that $1/T_1 T$ for both Te sites under all three magnetic field directions appears to increase with increasing pressure. In particular, as reported before [28], $1/T_1 T$ for Te(I) enhances more than that of Te(II) under $H \parallel b$ at $p = 1.05$ GPa, which was attributed to the enhancement of AFM spin fluctuations under pressure. With the present new $1/T_1 T$ data, although we were not able to resolve the T_1 values for the two Te lines along the c axis because of the overlapping of the two signals, it is clearly evident that, for not only $H \parallel b$ but also for $H \parallel a$, $1/T_1 T$ for Te(I) enhances more than that of Te(II) under pressures of 1.05 and 1.93 GPa. The values of the $1/T_1 T$ are consistent with the previously reported values at ambient pressure for both Te sites for all three magnetic field directions [23] and under pressure for the Te(I) site with the magnetic field parallel to the b and c axes [29].

We analyze the current T_1 data following the previous procedure [15]. By writing the directional magnetic fluctuation components, $R_{i,\alpha}$ where $i = \text{I, II}$ and $\alpha, \beta, \gamma = \{a, b, c\}$ for each Te(i) as

$$R_{i,\alpha} = \frac{\gamma_N^2 k_B}{2} \sum_{q,\xi} |A_{\text{hf},i}^{\alpha\xi}(q)|^2 \frac{\chi''_{\xi}(q, \omega_N)}{\omega_N}, \quad (1)$$

$(1/T_1 T)_{i,\alpha}$ can be described by $(1/T_1 T)_{i,\alpha} = R_{i,\beta} + R_{i,\gamma}$ where (α, β, γ) are mutually orthogonal directions. Here $\chi''(q, \omega_N)$ is the imaginary part of the dynamic magnetic susceptibility at the NMR frequency (ω_N) along the ξ ($=\{a, b, c\}$) directions and $A_{\text{hf},i}^{\alpha\xi}(q)$ is the q -dependent hyperfine coupling

tensor for ^{125}Te nuclei. Utilizing the $1/T_1$ data measured under the three different H directions, we derived the temperature dependence of $R_{i,\alpha}$ for each direction at three different pressures, as shown in Figs. 7(a)–7(c). It is noted that a recent NMR study comparing a “early stage” $T_c = 1.6$ K crystal and “ultraclean” $T_c = 2.1$ K crystal showed a difference in T dependence of $1/T_1 T$ at low temperatures (mainly below ≈ 10 K) due to the U deficiency in the low- T_c crystal [24]. Therefore, since the high-temperature (above ≈ 10 K) behavior seems to be consistent across the samples [24], we discuss the magnetic fluctuations based on our data only above 10 K, which are shown with the solid symbols in Figs. 7(a)–7(c), together with the data below 10 K shown with the translucent symbols for reference. In the figures, we also plotted the temperature dependence of the corresponding K of both the Te sites in each panel (the right axes of the figures). Since K reflects the temperature dependence of the real component $\chi'(\vec{q}, \omega_0)$ with $q = 0$ and $\omega_0 = 0$, the comparison between $R_{i,\alpha}$ and K allows us to examine the T dependence of $\sum_{q,\xi} |A_{\text{hf},i}^{\alpha\xi}(q)|^2 \frac{\chi''_{\xi}(q, \omega_N)}{\omega_N}$ with respect to the uniform static susceptibility $\chi'(0, 0)$.

At ambient pressure ($p = 0$ GPa), the temperature dependence of $R_{i,\alpha}$ roughly scales with the corresponding K , suggesting that the $q = 0$ ferromagnetic fluctuations are dominant. It is noted that $R_{i,c}$ for Te(I) is greater than those for Te(II) along the c axis at low temperatures, as has been pointed out in Ref. [23]. Furthermore, the temperature dependence of $R_{i,c}$ for Te(I) does not scale with that of K , whereas for Te(II), both $R_{i,c}$ and K are scaled (see the SM [38]). These results indicate that Te(I) picks up the additional magnetic fluctuations other than $q = 0$, providing evidence for the hyperfine field fluctuations with AFM nature along the c axis at Te(I). It is also noted that, even though K for Te(I) is smaller than that of Te(II) at low temperatures, $R_{i,a}$ of Te(I) is slightly greater (or almost similar) than that of Te(II). This indicates a site depen-

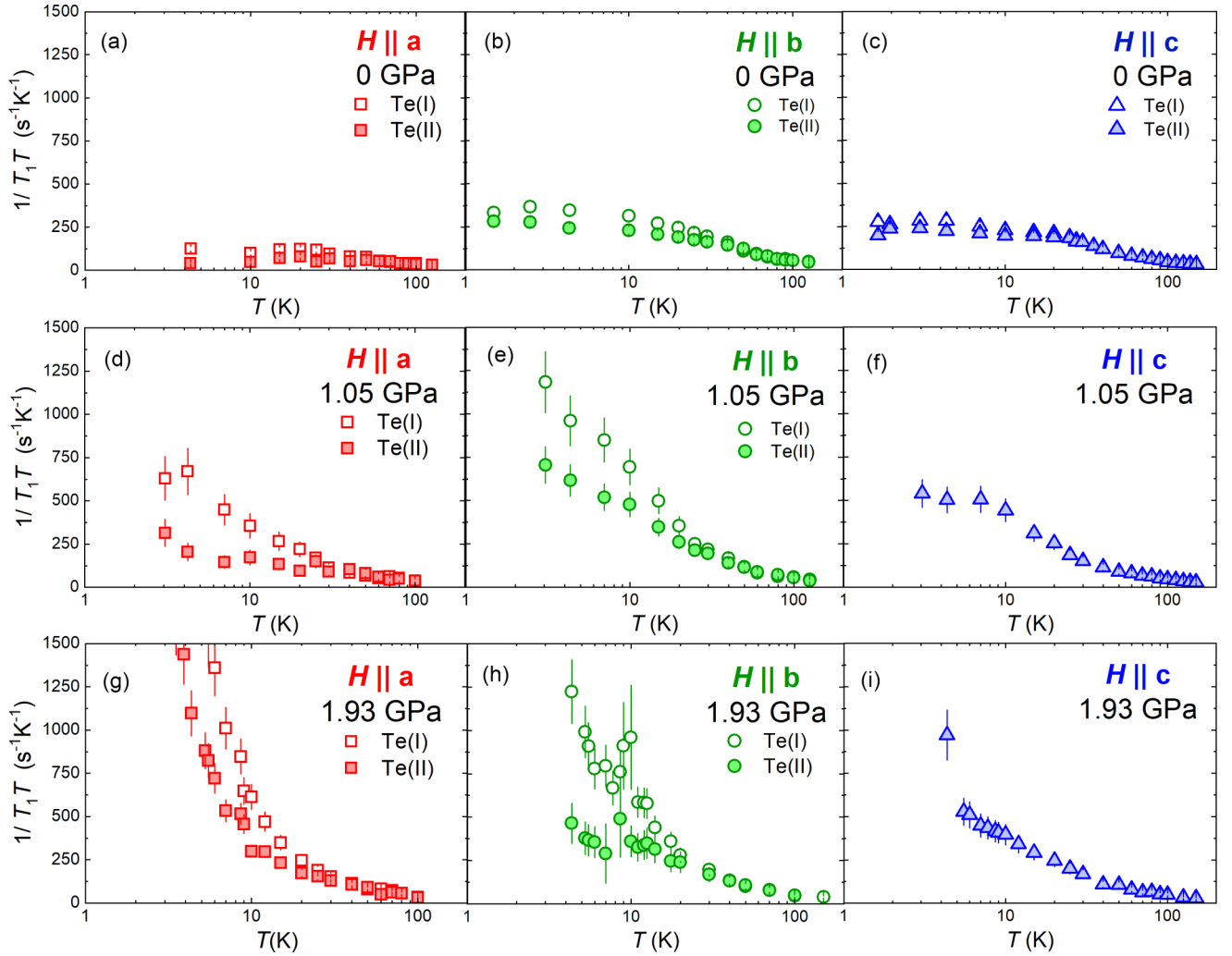


FIG. 6. Temperature dependence of $1/T_1T$ for Te(I) (open symbols) and Te(II) (solid symbols) at (a)–(c) $p = 0$ GPa, (d)–(f) 1.05 GPa, and (g)–(i) 1.93 GPa under $H \parallel a$ (in red), $H \parallel b$ (in green), and $H \parallel c$ (in blue). The data at ambient pressure under $H \parallel b$ are from Ref. [28].

dence in magnetic fluctuations, consistent with the previous NMR report [24], suggesting that Te(I) detects the hyperfine field fluctuations associated with the AFM nature along the a axis as well, even though the ferromagnetic fluctuations are dominant. The existence of the AFM fluctuations along the a axis could be more clearly seen in the temperature dependence of $R_{i,a}$ divided by the corresponding K , $R_{i,a}/K$, as shown in the SM [38]. It is noted that the magnetic fluctuations picked up at the Te sites originate from U spin fluctuations via hyperfine interactions. Since the Te1 and Te2 sites are located at different sites with distinct local environments, the differences in magnetic fluctuations detected at the Te(I) and Te(II) sites can be attributed to the distinct hyperfine coupling tensors (see the SM [38]) where Te(I) and Te(II) are most likely assigned to the crystallographic Te1 and Te2 sites, respectively.

At $p = 1.05$ GPa, the significant enhancement of $R_{i,a}$ and $R_{i,c}$ at 1.05 GPa can be seen in Fig. 7(b), which can be attributed to an additional contribution of the AFM spin fluctuations under pressure. Especially, a clear difference in $R_{i,c}$ between Te(I) and Te(II) is observed, evidencing that again Te(I) detects the AFM spin fluctuations more than Te(II). This

is consistent with the previous NMR measurements under pressures up to 1.57 GPa [28]. It turns out that the different temperature dependence of $1/T_1T$ of Te(I) and Te(II) for $H \parallel b$ under pressure reported before in Ref. [28] was due to the different temperature dependence of $R_{i,c}$ for Te(I) and Te(II) because no difference in $R_{i,a}$ in the two Te sites was observed.

When p is greater than the critical pressure p_c , although $R_{i,c}$ for Te(I) and Te(II) is similar to that at 1.05 GPa showing the AFM spin fluctuations picked at the Te(I) site, it is clear that $R_{i,b}$ for both Te sites is enhanced compared with the lower-pressure case below p_c . Note that, although the K for the b axis direction increases at this p , the enhancement of $R_{i,b}$ for both Te sites is greater than the increase in K . Therefore, AFM spin fluctuations are actually enhanced along the b axis. This is quite different from the lower-pressure case where the weak b axis magnetic fluctuations were observed. Although we do not have a clear idea to explain the large enhancement of AFM spin fluctuations along the b axis at present, it could be related to the change in the magnetic easy axis from the a to b axes above p_c [27] or could be due to the different

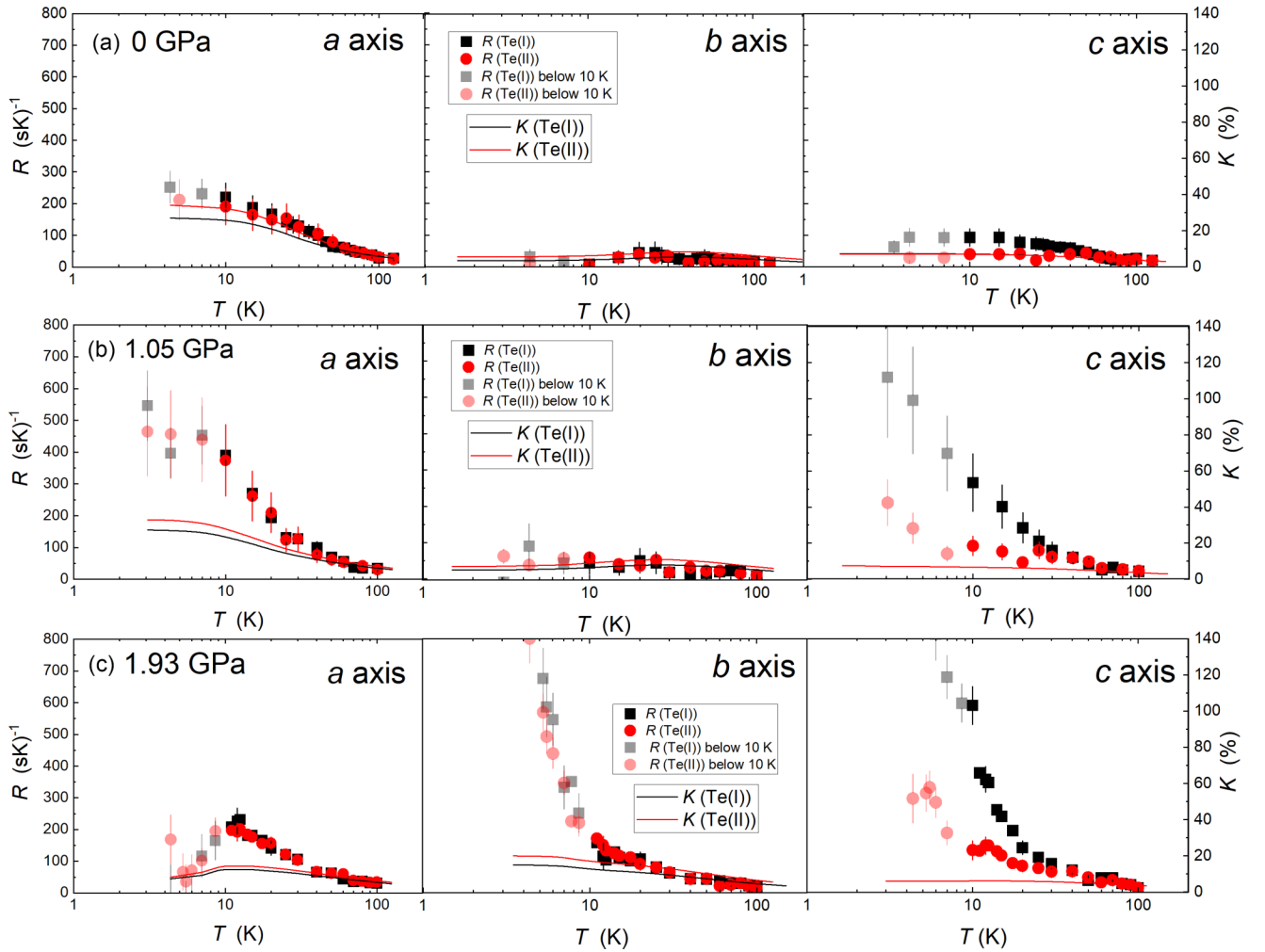


FIG. 7. Temperature dependence of R_a (left), R_b (middle), and R_c (right) for (a) $p = 0$ GPa, (b) 1.05 GPa, and (c) 1.93 GPa for Te(I) (black squares) and Te(II) (red circles), together with the temperature dependence of the corresponding K of Te(I) (black curves) and Te(II) (red curves). The R_i data below 10 K are also shown with translucent symbols for reference.

wave vector in the antiferromagnetically ordered state [30]. Further studies are needed to clarify the magnetic fluctuations above p_c . It is also noted that the $R_{i,a}$ for both Te sites are slightly suppressed. This suppression cannot be explained by the large reductions in K , again evincing the hyperfine field fluctuations associated with AFM nature along the a axis.

Finally, it is important to mention that, as pressure increases, the T_c of SC2 increases while the T_c of SC1 decreases. Given the observed enhancement of AFM spin fluctuations with pressure, the AFM spin fluctuations would play an important role in the appearance of SC2. In this context, it is interesting to point out that recent theories proposed a possibility of the spin-triplet state induced by AFM spin fluctuations [31,32,43]. Conversely, the AFM fluctuation would likely be unfavorable for SC1 state as the T_c of SC1 decreases upon pressure, which would suggest the importance of FM fluctuations for SC1.

To qualitatively illustrate the correlation between AFM spin fluctuations and superconductivity in UTe_2 , we present the evolution of AFM spin fluctuations under pressure in a contour plot (Fig. 8). Here the magnitude of AFM spin

fluctuations is tentatively represented by $(1/T_1 T)_{\text{AFM}} = 1/T_1 T[\text{Te(I)}] - 1/T_1 T[\text{Te(II)}]$ using the $1/T_1$ data measured under $H \parallel b$, which mainly corresponds to the difference in R_c for the two Te sites. Although it is better to use the quantities estimated from the comparison between the R_α and K_α data to represent the AFM spin fluctuations, we used only the $1/T_1 T$ data under $H \parallel b$ to make the contour plot due to the limited number of datasets for R_α under pressure. Thus, the plot reflects the partial contributions of the AFM fluctuations. Nevertheless, the contour plot captures the main feature of the enhancement of AFM fluctuations with increasing pressure, concomitant with the rise of T_c for SC2. It is noted that the small reduction in T_c close to p_c could be due to the long-range magnetic ordering appearing above p_c . A similar suppression of T_c near the AFM phase has been observed in iron-based superconductors, such as $\text{FeSe}_{1-x}\text{S}_x$ under pressure [44]. As mentioned above, T_c for SC1 is suppressed as pressure is augmented. Given the FM spin fluctuations dominate at ambient pressure, these results would suggest that FM spin fluctuations are more favorable for SC1 and AFM spin fluctuations are crucial for SC2, as pointed out above. A recent NMR study by Kinjo *et al.* reported that the $\mathbf{d}$ vector has no b component

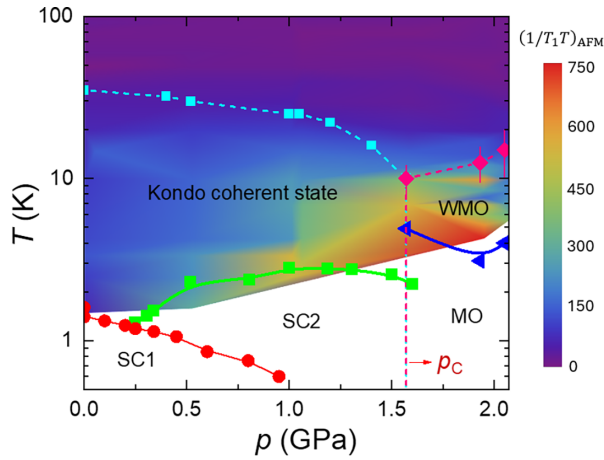


FIG. 8. p - T contour plot showing the magnitude of AFM spin fluctuations tentatively represented by $(1/T_1T)_{\text{AFM}} = 1/T_1T[\text{Te(I)}] - 1/T_1T[\text{Te(II)}]$ as defined in the text. The $1/T_1T$ data with $H \parallel b$ for 0.0–2.05 GPa are used to estimate $(1/T_1T)_{\text{AFM}}$. The T_1 data for $p = 0, 0.52, 1$, and 1.57 GPa are from Ref. [28], and the T_1 data for $p = 2.05$ GPa is given in the Supplemental Material [38]. The various transition temperatures, including T_c , T_{max} , T_{WMO} , and T_{MO} from Fig. 1 are also plotted.

in SC2 while it exhibits a finite component along the b axis in SC1 (and also SC3 defined in Ref. [13]). It is interesting if these differences are related to the distinct magnetic fluctuations suggested for the different SC phases. Further studies are therefore highly requested to elucidate the origin of superconductivity as well as the SC properties in the SC1 and SC2 phases.

IV. SUMMARY

We performed ^{125}Te NMR measurements on a ^{125}Te -enriched single crystal of UTe_2 ($T_c = 1.6$ K) to investigate the evolution of magnetic fluctuations in UTe_2 under pressure. Following the previous NMR studies under pressure, which suggest that the AFM spin fluctuations are enhanced with the application of pressure [28], we measured $1/T_1T$ under the magnetic field parallel to the three crystalline axes at various pressures ranging from 0 to 2.05 GPa. As a result, we evidenced the enhancement of AFM spin fluctuations under pressure. In particular, from the directional resolved dynamical spin susceptibilities derived from the $1/T_1T$ data of the

three magnetic-field directions, we found the hyperfine fluctuations at the Te sites associated the AFM spin fluctuations are mainly enhanced along the a and c axes, which is the origin of the enhancement of $1/T_1T$ for $H \parallel b$ under pressure. It is also shown that the Te(I) site picks up the enhancements of hyperfine field fluctuations along the c axis compared with the Te(II) site. From this difference, combined with the calculations of the hyperfine tensors for the two Te sites in the crystal (see the SM [38]), we suggest that Te(I) and Te(II), denoted in the NMR lines, are assigned to Te1 and Te2, respectively, denoted in the crystal.

We also revealed the magnetically ordered state above the critical pressure p_c from the NMR measurements. From the disappearance of the Te NMR lines, the long-range magnetic order temperatures were estimated to be $\approx 4.9(6)$ K at 1.57 GPa, $\approx 3.1(4)$ K at 1.93 GPa, and $\approx 4.0(3)$ K at 2.05 GPa. The short-range (or weakly) magnetic order was also detected by the different temperature dependence of two Te NMR lines below 10–15 K, which we attributed to the development of ferromagnetic correlations within the U ladders.

Our results suggest that AFM spin fluctuations play an important role in the appearance of SC2, as T_c of SC2 increases with pressure. On the other hand, as the SC1 phase is suppressed with pressure, the AFM spin fluctuations are unfavorable for the phase. Therefore, we suggest that FM spin fluctuations are more favorable for SC1, and AFM spin fluctuations are crucial for SC2. Further studies are strongly requested to elucidate the origin of superconductivity of the SC1 and SC2 phases, as well as to characterize the two SC states.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [45], embargo periods may apply.

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