

Longitudinal Spin Fluctuations Driving Field-Reinforced Superconductivity in UTe_2

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Our measurements of ^{125}Te NMR relaxations reveal an enhancement of electronic spin fluctuations above $\mu_0 H^* \sim 15$ T, leading to their divergence in the vicinity of the metamagnetic transition at $\mu_0 H_m \approx 35$ T, below which field-reinforced superconductivity appears when a magnetic field (H) is applied along the crystallographic b axis. The NMR data evidence that these fluctuations are dominantly longitudinal, providing a key to understanding the peculiar superconducting phase diagram in $H\parallel b$, where such fluctuations enhance the pairing interactions.

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The uranium-based superconductor UTe_2 provides an attractive platform for studying the novel physics of spin-triplet and topological superconductivity (SC) in bulk materials [1]. The compound displays bulk superconductivity below $T_c = 1.6\text{--}2.1$ K [2–4]. The formation of spin-triplet pairing is supported by a tiny reduction in the NMR Knight shift in the SC state [5,6], large upper critical field exceeding the ordinary Pauli paramagnetic limit [2,7–9], and multiple SC phases similar to superfluid ^3He [10–15]. The topological properties of the SC excitations, on the other hand, are suggested from measurements of the STM [16], Kerr effect [17,18], and London penetration depth [19,20] that have detected anomalous effects potentially originating from a chiral SC state but are still debated.

The upper critical field of the SC in UTe_2 increases significantly when a magnetic field (H) is applied exactly along the crystallographic b axis, which is perpendicular to the easy magnetic a axis along which the uranium $5f$ spin moments favor aligning with an Ising character [7–9]. With increasing the H , T_c initially decreases as in ordinary superconductors but starts to rise above $\mu_0 H^* \sim 15$ T. This increase continues up to 35 T, beyond which T_c suddenly drops to zero at higher magnetic fields. Such a field-reinforced SC behavior is closely related to the field-induced, first-order metamagnetic transition emerging at $\mu_0 H_m \approx 35$ T in $H\parallel b$ [7,8,21–25]. A phase boundary separating a low-field superconducting (LFSC) state from a high-field superconducting (HFSC) state has been recently discovered around $\mu_0 H^* \sim 15$ T [13–15]. The $\mu_0 H^*$ has been found to be insensitive to the sample quality and, thus, T_c at zero field. A possible change of the pairing mechanism has been discussed between the LFSC and

HFSC [13]. Moreover, the signature of another phase boundary has been detected inside the LFSC state around 13 T [15].

Several theoretical models have been proposed to explain such an anomalous SC phases diagram in $H\parallel b$ [26–30]. While these models predict different SC order parameters for the LFSC and HFSC states, they commonly require some mechanism to boost T_c in higher magnetic fields. In most cases, this is assumed to occur through enhanced spin fluctuations that may arise near H_m . This assumption is based on the similarities of the SC phase diagram to that for uranium-based ferromagnetic (FM) superconductors, URhGe [31] and UCoGe [32]. In these materials, the SC state is established within the FM state, and the pairing interaction is thought to be mediated by the exchange of FM spin fluctuations [33–37]. Under magnetic fields applied along the magnetically hard axis, the compounds exhibit field-induced (URhGe) or field-reinforced (UCoGe) SC, similar to UTe_2 . NMR experiments have revealed that the excitation spectrum of the spin fluctuations is modified strongly by an applied field, depending on its strength and direction [38–43]. This implies that the strength of the pairing interactions would also depend on magnetic fields, and, in fact, such a field-dependent pairing mechanism well explains many of the unconventional SC phenomena observed under magnetic fields in the FM superconductors [38–49].

In this Letter, we present the results of high-field ^{125}Te NMR experiments performed on high-quality single crystals of UTe_2 . Our NMR measurements with magnetic fields applied along the b of the crystal axis up to 36 T reveal the development of longitudinal ($\parallel b$) spin fluctuations above

$\mu_0 H^* \sim 15$ T and their divergence near the field-induced metamagnetic transition at $\mu_0 H_m \approx 35$ T. These findings offer valuable insights into the unique SC phase diagram for $H \parallel b$, where the diverging longitudinal spin fluctuations enhance the pairing interactions, resulting in the boost of T_c above H^* .

High-field NMR data were obtained using a 24 MW resistive magnet at the LNCMI-Grenoble. The experiment was performed on a ^{125}Te -enriched (99%) single crystal (#1) with $T_c = 2.0$ K grown by the molten salt flux (MSF) method [4]. ^{125}Te nuclei have the nuclear gyromagnetic ratio $\gamma_N = 13.454$ MHz/T ($I = 1/2$) with a natural abundance of 7%, so that the enrichment largely enhances the NMR signal intensity and the signal to noise ratio. NMR experiments at lower fields below 15 T were performed using a SC magnet on a single crystal (#2) with $T_c = 1.5$ K grown by the chemical vapor transport (CVT) method [2]. The H dependence of the NMR spectrum, the nuclear spin-lattice relaxation rate ($1/T_1$), and the nuclear spin-spin relaxation rate ($1/T_2$) were measured at a fixed temperature of 2.1 K for the #1 and 1.6 K for the #2 crystals; these temperatures are about 0.1 K above the zero-field T_c of each crystal.

In the orthorhombic structure of UTe_2 (space group No. 71, $Immm$, D_{2h}^{25}) [50,51], Te atoms have two crystallographically inequivalent sites, Te(1) and Te(2), in the unit cell. Thus, the ^{125}Te NMR spectrum consists of two distinct peaks arising from the two inequivalent sites in $H \parallel b$. In previous studies, we found no qualitative differences for $1/T_1$ and $1/T_2$ behaviors between the two peaks [52]. We, thus, focus on the NMR peak at the higher frequency, corresponding to the Te(2) site in this study.

In Fig. 1(b), we compared the NMR spectra observed in crystals #1 and #2 with different T_c in the field of 15 T. The NMR linewidth of the #1 crystal is about 20% narrower than that of the #2 crystal. The results support microscopically our expectation that crystals with a higher T_c have fewer crystal defects and/or less disorder than lower- T_c crystals [4,53,54]. As seen in Fig. 1(a), the NMR linewidth of both crystals increases linearly with increasing H , as expected when the distribution of the NMR shift determines the linewidth.

Figure 2 shows the field dependence of the NMR shift $\Delta f = f_{\text{NMR}} - f_0$, where f_{NMR} is the peak frequency of NMR spectrum determined by the peak position of each spectrum and $f_0 = \gamma_N H$. While the NMR linewidths are different between the two crystals, there is no significant change in $\Delta f(H)$, so their $\Delta f(H)$ are smoothly connected at 15 T. As expected, the $\Delta f(H)$ follows $M(H)$, and, hence, it is nearly proportional to H up to 32 T. This provides a nearly field-independent Knight shift at $K = \Delta f/f_0 \approx 5.5\% - 6\%$, as shown in the inset in Fig. 1, whereas the $K(H)$ presents only a very weak, gradual increase. The K values are consistent with those in previous reports [14,52]. Note that no NMR shift data are above

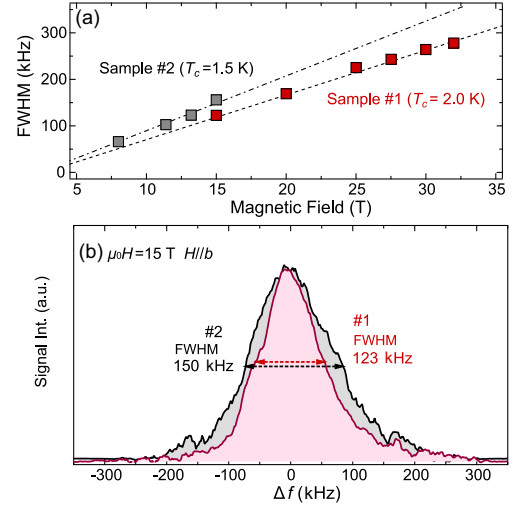


FIG. 1. (a) Field dependences of the NMR linewidth (full width at half maximum) from two different T_c samples, #1 ($T_c = 2.0$ K) and #2 ($T_c = 1.5$ K). (b) Comparison of the NMR spectra obtained in these two crystals in the field of 15 T.

32 T, although our experiment was performed up to 36 T. In this field region, we completely lost the NMR spin-echo signals due to extremely short T_2 , as discussed below.

We now turn to the spin dynamics for $H \parallel b$. Figures 3(a) and 3(b) show the field dependence of the $1/T_1$ and $1/T_2$ relaxation rates up to 32 T. Here, $1/T_1$ was measured by applying the saturation $\pi/2$ pulse at time t before the $\pi/2 - \tau - \pi$ spin-echo sequence used to record the recovery data $R(t)$ and was evaluated by fitting $R(t)$ to the exponential function for spin $I = 1/2$ nuclei (^{125}Te) [55]. $1/T_2$ was measured by monitoring the decay of the spin-echo intensity $I(\tau)$ as a function of the interval time τ

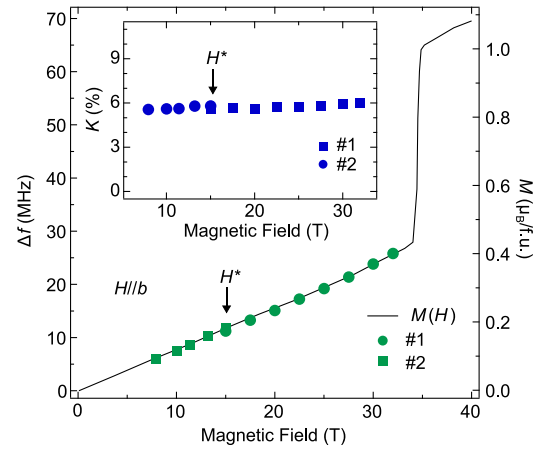


FIG. 2. The field dependence of the NMR shift $\Delta f = f_{\text{NMR}} - f_0$, where f_{NMR} is the peak frequency of NMR spectrum determined by the peak position of each spectrum at 2.1 (1.6) K for #1 (#2) crystals and $f_0 = \gamma_N H$. The inset shows the field dependence of the Knight shift $K = \Delta f/f_0$. The solid line is the magnetization curve at 1.4 K [22].

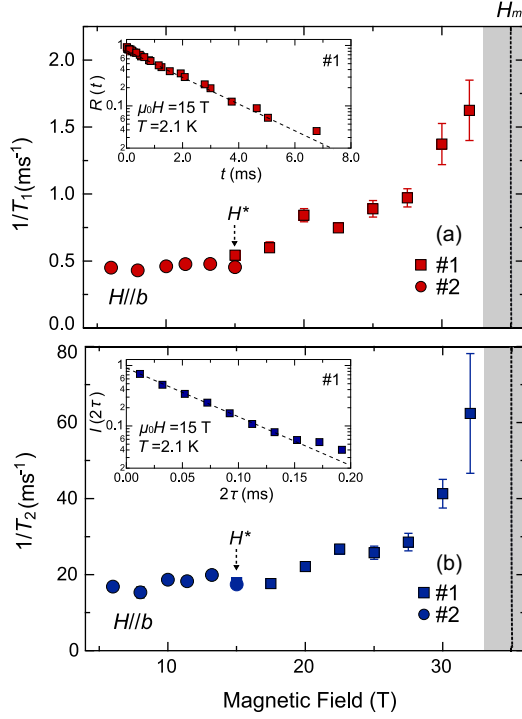


FIG. 3. The field dependence of (a) $1/T_1$ and (b) $1/T_2$ in $H\parallel b$. In the gray area above 32 T, we lost NMR spin-echo signals due to extremely short T_2 . Insets display examples of the measured relaxation curves (symbols) and the exponential fits (dotted lines).

between the $\pi/2$ and π pulses and was evaluated by fitting $I(\tau)$ to the exponential function. We obtained satisfactory fitting by a single component of T_1 [T_2] for $R(t)$ [$I(\tau)$] (insets in Fig. 3), showing that the spin fluctuations are homogeneous.

While the $1/T_1$ and $1/T_2$ are nearly field independent at lower fields, both quantities start to increase above ~ 15 T and show a tendency to diverge above 32 T. As mentioned, NMR spin-echo signals were not observed above 32 T (gray area in Figs. 3 and 4). In this region, T_2 values become extremely short, much shorter than 3 μ s, the dead time of our NMR spectrometer. This confirms the divergence of fluctuations in the vicinity of the field-induced metamagnetic transition at $H_m \approx 35$ T. Previous macroscopic studies have defined $\mu_0 H^* \sim 15$ T as the characteristic field above which H_{c2} shows an upturn as the HFSC phase emerges on top of the LFSC phase [13,15]. Interestingly, the $\mu_0 H^*$ has been found to remain unchanged between UTe₂ crystals with different qualities, even though the improvement of the crystal quality markedly increases both the onset T_c and the extrapolated $H_{c2}^{\text{LFSC}}(0)$ [15]. This implies that the H^* is not simply determined as the intersection of two SC phase boundaries but that there are some other sources to locate the H^* around 15 T. Our NMR results show that the H^* is defined as a characteristic field above which critical

fluctuations begin to develop toward the metamagnetic transition. It has been demonstrated [1,13] that even the rather sharp upturns observed on H_{c2} could be reproduced with a smooth continuous increase of the pairing strength $\lambda(H)$, whose field dependence is related to the growth of critical fluctuations.

In the following, we discuss the nature of fluctuations detected by $1/T_1$ and $1/T_2$. In general, $1/T_2$ is given by the sum of electronic and nuclear contributions, $1/T_2 = (1/T_2)^{\text{el}} + (1/T_2)^{\text{nu}}$. In the present case, we can safely assume that $(1/T_2)^{\text{el}} \gg (1/T_2)^{\text{nu}}$, since $1/T_2$ values are confirmed to be independent of the ¹²⁵Te isotope concentration. $1/T_1$ and $1/T_2$ are, thus, both determined by electronic contributions. For $I = 1/2$ nuclear spins in the magnetic field applied along the b direction, the electronic-spin-induced fluctuations of the local magnetic field $h(t)$ will induce the $1/T_1$ relaxation according to Moriya's formula [56],

$$1/T_{1,b} = [u_{aa}(\omega_{\text{NMR}}) + u_{cc}(\omega_{\text{NMR}})]/2, \quad (1)$$

where

$$u_{AB}(\omega) = \gamma^2 \int_{-\infty}^{+\infty} \langle h_A(0)h_B(t) \rangle e^{-i\omega t} dt \quad (2)$$

is the Fourier transform of the field-fluctuation correlation function. The same fluctuations also contribute to the so-called Redfield term in the T_2 relaxation [57–60]:

$$\begin{aligned} 1/T_{2,b} &= u_{bb}(0)/2 + [u_{aa}(\omega_{\text{NMR}}) + u_{cc}(\omega_{\text{NMR}})]/4 \\ &= u_{bb}(0)/2 + (1/T_{1,b})/2. \end{aligned} \quad (3)$$

The first term is determined by the *longitudinal* ($\parallel b$) fluctuations of $h(t)$ averaged over the timescale of T_2 itself. It is, thus, sensitive to *slow* fluctuations of electronic, magnetic moments as compared to those at the NMR frequency ω_{NMR} of the order of 100 MHz. On the other hand, the second term is caused by the transverse ($\perp b$) fluctuations of $h(t)$ probed at ω_{NMR} that define $1/T_1$.

In the absence of frequency dependence of the local field fluctuations, Eq. (3) tells us that for pure transverse, isotropic, or longitudinal fluctuations, we expect that the ratio $(1/T_{2,b})/(1/T_{1,b})$ respectively equals to 1/2, 1, or ∞ . The experimental value $(1/T_{2,b})/(1/T_{1,b}) \approx 36$ (Fig. 4) then clearly points to dominant longitudinal fluctuations. However, if we admit possible strong frequency dependence of fluctuations, the conclusion is less evident. Nevertheless, in this case, it is clear that $1/T_2$ and $1/T_1$ would present very different $\omega_{\text{NMR}} \propto H$ dependence, with $1/T_2$ being essentially field independent and $1/T_1$ *decreasing* with the field. This is in strong contrast to what is observed: Both quantities have essentially identical, *increasing* field dependence (Fig. 4). We, thus, conclude

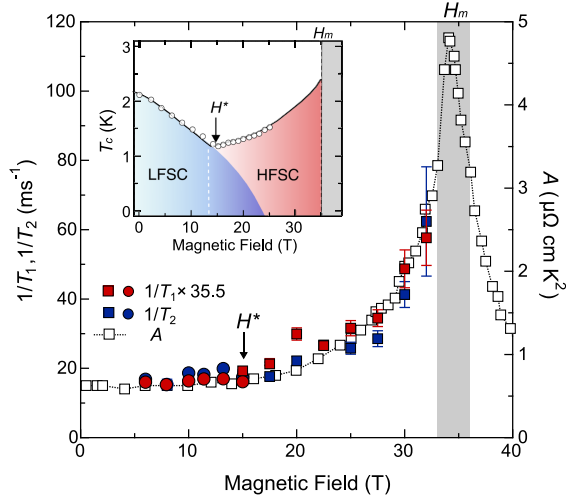


FIG. 4. The field-dependent quadratic coefficient $A(H)$ (with current $I \parallel a$) reported in Ref. [21] is compared with $1/T_2(H)$ and with $1/T_1(H)$ scaled by a factor of 35.5. The inset shows a schematic $H-T$ phase diagram for UTe_2 in the case of $H \parallel b$ [13–15]. The $T_c(H)$ data (white circle) are from Ref. [15].

that both quantities track the same dominant *longitudinal* spin fluctuations.

Precisely this will be realized if strong anisotropy of spin fluctuations overcomes the weakness of the (squared) off-diagonal terms of the hyperfine coupling tensor, $A_{ab}^2, A_{cb}^2 < A_{aa}^2, A_{cc}^2$. Here, we recall that the A tensor relates the electronic spin or moment to the local field felt by the nuclei [57,58]. In Supplemental Material [61], we show that in this case the *field-independent* ratio $(1/T_{2,b})/(1/T_{1,b}) = A_{bb}^2/(A_{ab}^2 + A_{cb}^2) + 1/2$ is observed only when the spin fluctuations are themselves frequency independent between $\omega = 0$ and $\omega = \omega_{\text{NMR}}$.

We also remark that a development of extremely slow fluctuations, observed in $1/T_2$ but not in $1/T_1$, was detected in our previous study [53] using a CVT crystal of UTe_2 at low fields, in $H \parallel a$ but not in $H \parallel b$. A low-temperature upturn in the spin susceptibility ($H \parallel a$) is attributed to disorder or defects that are intrinsically present in MSF crystals but do not exist in the presently studied CVT crystal [4].

In Fig. 4, we plot the H -dependent quadratic coefficient $A(H)$ together with our $1/T_1$ and $1/T_2$ data. Here, the $A(H)$ is extracted from a Fermi-liquid fit to the electrical resistivity at low temperatures (with current $I \parallel a$) by $\rho(T) = \rho_0 + AT^2$ in $H \parallel b$ [21,24,25]. The significant increase in $A(H)$ near H_m has been regarded as due to the enhancement of the effective mass m^* [21,24,25]. We found a reasonable scaling between the $A(H)$ and the NMR relaxation rates up to 32 T. A similar strong field dependence, accompanying a sharp maximum around H_m , has also been observed in the Sommerfeld coefficient $\gamma(H)$, which was derived in literature from the specific-heat data at low T [13,23] or the T dependence of magnetization using a

thermodynamic Maxwell relation [22,63]. Our NMR experiments demonstrate the importance of longitudinal spin fluctuations in the entire field range and that their development underlies these unconventional transport and thermal behaviors in $H \parallel b$. Remarkably, Miyake has predicted based on an extended Landau theory of the first-order metamagnetic transition that enhanced longitudinal spin fluctuations would give rise to the enhancement of the A coefficient and the γ around the metamagnetic field [64].

It should be noted that the diverging $1/T_2$ behavior observed here in UTe_2 is very similar to that in URhGe . The latter compound is a ferromagnet with a strong Ising-type spin anisotropy and exhibits a metamagnetic-like transition from a FM state to a high-field polarized state when a field of $H_m = 12$ T is applied along a hard-magnetization axis ($\parallel b$). The diverging $1/T_2$ has been observed near the critical fields, which is also attributed to the divergence of longitudinal spin fluctuations [39–41,43]. For URhGe , the divergence has been associated with the presence of a tricritical point (TCP). The TCP locates at 12 T and around 5 K [31,65–67], below which a second-order transition changes to a first-order transition. With regard to magnetic excitations, a characteristic feature of the TCP is that it can trigger a diverging susceptibility not only for the order parameter (M_c) but also for the physical quantity that is conjugate to the tuning parameter H_b driving the phase transition, that is, M_b in the case of URhGe [39,40,68–70]. Thus, we can expect a divergence of the longitudinal component of magnetic fluctuations, resulting in the diverging $1/T_2$ in $H \parallel b$. In URhGe , the reentrant SC has been found to occur in almost the same region as that where the longitudinal fluctuations are developing [39,40]. On the other hand, there is no TCP in UCoGe , and, hence, only a broad peak appears in $1/T_2$ around a critical field of 12–13 T [42].

For the metamagnetic transition in UTe_2 , there is no TCP, but a critical point (CP) exists at $T = 5\text{--}7$ K with $H_m = 35$ T [21,22,24]. Below the CP, a metamagnetic crossover at higher temperatures changes to a first-order transition. In this context, it is also important to notice that the diverging $1/T_2$ has also been observed in the vicinity of the CP (at 10 K, 1 T) in an itinerant paramagnet UCoAl [71,72]. This compound is located on the paramagnetic side in the vicinity of FM order, and a metamagnetic transition to a FM phase occurs under a small H (~ 1 T). In the case of UCoAl , however, the critical fluctuations are suppressed rapidly in the first-order region below 10 K, making clear contrast to the case of UTe_2 . This might be related to the fact that the CP of UCoAl occurs under H applied along the easy-magnetization axis [1,73]. It is also remarkable that both UTe_2 and UCoAl exhibit a broad maximum in $\chi(T)$ around 20–30 K [2,73].

Theoretically, spin fluctuations near a FM quantum critical point have been supposed to create a binding force between quasiparticles with equal (triplet) spin pairs [36,37], analogous to the mechanism of a superfluid pairing in ^3He [74]. After the discovery of the uranium-based FM superconductors, theoretical models of the spin-triplet SC under magnetic fields were developed by Mineev [75–77], Tada and Fujimoto [45,78], and Hattori and Tsunetsugu [46], independently. Those theories have pointed out the importance of field-dependent spin susceptibility and, thus, spin fluctuations. More recently, using the density matrix renormalization group, Suzuki and Hattori have analyzed SC correlations in the one-dimensional Kondo lattice models with Ising anisotropy under transverse magnetic fields [47,48]. They found that competitions between the transverse magnetic field and the Kondo singlet formation lead to both enhanced SC correlations and metamagnetic behaviors, where metamagnetic fluctuations play a crucial role [48]. The present NMR experimentally captures the fluctuations that well corroborate these theoretical models, although a more quantitative comparison between experiments and theories remains for future works. We will also extend the NMR experiments to higher magnetic fields above H_m , where the rapid suppression of the diverging fluctuations is expected, accompanied by the sudden decrease of $T_c(H)$.

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