Incommensurate Spin Fluctuations in the Spin-triplet Superconductor Candidate UTe$_2$

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Spin-triplet superconductors are of extensive current interest because they can host topological state and Majorana fermions important for quantum computation. The uranium based heavy-fermion superconductor UTe$_2$ has been argued as a spin-triplet superconductor similar to UGe$_2$, URhGe, and UCoGe, where the superconducting phase is near (or coexists with) a ferromagnetic (FM) instability and spin-triplet electron pairing is driven by FM spin fluctuations. Here we use neutron scattering to show that although UTe$_2$ exhibits no static magnetic order down to 0.3 K, its magnetism is dominated by incommensurate spin fluctuations near antiferromagnetic (AF) ordering wave vector and extends to at least 2.6 meV. We are able to understand the dominant incommensurate spin fluctuations of UTe$_2$ in terms of its electronic structure calculated using a combined density functional and dynamic mean field theory.

Superconductivity occurs in many metals when electrons form coherent Cooper pairs below the superconducting transition temperature $T_c$. In conventional and most unconventional superconductors, electron Cooper pairs in the superconducting state form anti-parallel spin-singlets with the total spin $S = 0$. However, electrons in the superconducting state can also form parallel spin-triplet Cooper pairs, analogous to the equal spin pairing state in the superfluid $^3$He. The Pauli’s exclusion principle can be fulfilled for both singlet and triplet Cooper pairs by adjusting the symmetry of the orbital part of the wave function. For the spin-singlet pairing state, the orbital wave function has even parity (symmetric) with orbital angular momentum $L = 0$ (s-wave), 2 (d-wave), etc. For spin-triplet state, the orbital wave function has odd parity (antisymmetric) with orbital angular momentum $L = 1$ (p-wave), 3 (f-wave), and etc. While most unconventional superconductors have spin-singlet pairing associated with antiferromagnetic (AF) spin fluctuations, spin-triplet superconductors are rare, and the superconductivity is believed to be driven by longitudinal ferromagnetic (FM) spin fluctuations. Since spin-triplet superconductors are intrinsically topological and can host Majorana fermions important for quantum computation, it is important to understand a candidate spin-triplet superconductor by determining the associated spin fluctuations.

In spin-triplet superconductors such as UGe$_2$, URhGe, and UCoGe, superconductivity arises through suppression of the static FM order or coexists with static FM order. Inelastic neutron scattering (INS) experiments find clear evidence of FM spin fluctuations in URhGe and UCoGe. For the spin-triplet candidate superconductor Sr$_2$RuO$_4$, where the material is paramagnetic at all temperatures and superconductivity does not coexist with static FM order, magnetism is dominated by incommensurate spin fluctuations arising from Fermi surface nesting of itinerant electrons, although weak FM spin fluctuations are also observed. Similarly, although considerable evidence exists for spin-triplet superconductivity in UPt$_3$, its superconductivity appears to be associated with AF order and spin fluctuations instead of FM spin fluctuations.

Recently, UTe$_2$ has been identified as a new spin-triplet superconductor with $T_c \approx 1.6$ K. UTe$_2$ has an orthorhombic unit cell with space group Immmm, where the U atoms form parallel ladders along the $a$-axis inside trigonal prisms of Te atoms [Fig. 1(a)]. The shortest U-U bond is along the rung of the ladder in the $c$-axis direction, while the easy-axis of the U spins is along the $a$-axis. The symmetry operation that connects one ladder to its nearest neighbor is the body-center ($\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$) translation. A Curie-Weiss fit to the magnetic susceptibility data reveals an effective moment per U atom close to the $5f^2$ or $5f^3$ free ion value at high temperature. No long-range magnetic order has been reported down to 0.25 K. Instead, a sudden increase in the magnetic susceptibility below 10 K in response to a magnetic field applied parallel to the $a$-axis resembles the quantum critical behavior of metallic ferromagnets, indicating strong FM spin fluctuations along the $a$-axis. This suggests that UTe$_2$ sits at the paramagnetic end of a series of FM heavy fermion superconductors including UGe$_2$, URhGe, and UCoGe. At the FM end, the compound UGe$_2$ is a pressure-induced superconductor with optimal $T_c \approx 0.5$ K at 1.2 GPa.
Moving from UGe$_2$ to URhGe, superconductivity occurs at ambient pressure below $T_c \approx 0.25\,K$ and coexists with static FM order below a Curie temperature $T_C \approx 9.5\,K$ [18]. Finally, UCoGe has coexisting superconductivity and FM order with increased $T_c \approx 0.425\,K$ and decreased $T_C \approx 3\,K$, respectively [19].

The scenario that UTe$_2$ is a candidate spin-triplet superconductor [31, 32] is supported by a growing list of observations. These include the upper critical fields $H_{c2}$ that exceed the Pauli limits along all crystallographic directions [33, 34]; temperature independent $^{125}$Te Knight shift across $T_c$ in nuclear magnetic resonance measurements [31]; coexisting FM spin fluctuations and superconductivity [36, 39]; signatures of chiral superconductivity [31]; coexisting FM spin fluctuations and superconductivity [30]; signatures of chiral superconductivity [40]; and breaking of time reversal symmetry expected for a spin-triplet superconductor [31]. There are also theoretical [12] and experimental [13] efforts to understand the underlying electronic structure of UTe$_2$.

In this Letter, we use INS to probe the wave vector and energy dependence of spin fluctuations in UTe$_2$. In addition to confirming that UTe$_2$ exhibits no static magnetic order down to 0.3 K, we discovered that the dominant spin fluctuations in UTe$_2$ are three-dimensional (3D) in reciprocal space, centered at the incommensurate wave vector $Q = (0, \pm (K + 0.57), 0) \ (K = 0, 1)$, and extend to energies of at least $E = 2.6\,meV$. FM spin fluctuations, if present, are much weaker than the incommensurate spin fluctuations. Based on density functional theory (DFT) in the generalized gradient approximation (GGA) [44], combined with dynamical mean-field theory (DMFT) calculations [45–48], we understand the dominant incommensurate fluctuations by showing that the associated wave vector is approximately consistent with the AF wave vector of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the $5f$ moments. Therefore, in addition to a FM instability, incommensurate (close to AF) spin fluctuations must also be considered to unveil the magnetic and superconducting properties of UTe$_2$.

Our INS experiments were carried out at the Cold Neutron Chopper Spectrometer (CNCS) at Oak Ridge National Laboratory. The momentum transfer $Q$ in 3D reciprocal space is defined as $Q = H\vec{a}^* + K\vec{b}^* + L\vec{c}^*$, where $H, K$ and $L$ are Miller indices and $\vec{a}^* = \vec{a}2\pi/a$, $\vec{b}^* = \vec{b}2\pi/b$, $\vec{c}^* = \vec{c}2\pi/c$ with $a = 4.16\,\text{Å}$, $b = 6.12\,\text{Å}$ and $c = 13.95\,\text{Å}$ of the orthorhombic lattice [35]. Single crystals of UTe$_2$ were prepared using the chemical vapor transport method with I$_2$ as the transport media [39]. The crystals are naturally cleaved along the $ab$-plane and form small flakes of about 0.5 to 1 mm thick. The typical dimension of the crystals in the $ab$-plane is from 1 to 2 mm and the mass is in the range of 10 to 30 mg.

We co-aligned 61 pieces (total mass 0.7-g) of single crystals on oxygen-free Cu-plates using an X-ray Laue machine to check the orientation of each single crystal. The sample assembly is aligned in the $[0, K, L]$ scattering plane as shown in the black frame of Fig. 1(b) and mounted inside a He$^3$ refrigerator. Most of our measurements were carried out with $E_i = 3.37\,meV$ on CNCS at different temperatures.

Figure 1(c) shows a map of reciprocal space in the $[0, K, L]$ scattering plane at 0.3 K and elastic position. We see nuclear Bragg peaks at the expected $(0, \pm 1, \pm 1)$, $(0, \pm 2, 0)$, and $(0, \pm 2, \pm 2)$ positions. The spread of the Bragg peaks along the $L$ direction indicates a broad sample mosaic of $\sim 15$ degrees. To search for possible static FM or AF magnetic order, we show in Fig. 1(d) the temperature difference plot between 0.3 K and 2 K, and find no evidence of intensity gain anywhere within the probed reciprocal space. We therefore conclude that UTe$_2$ does not exhibit static FM or AF order down to 0.3 K, consistent with earlier work [31, 34, 36].

Despite the absence of long range magnetic order, INS experiments on UTe$_2$ reveal clear evidence for excitations at finite energy transfers. Figures 2(a-d) show 2D images of constant-energy cuts in the $[0, K, L]$ plane at different energies below $T_c$ ($T = 0.3\,K$). At $E = 0.4 \pm 0.1\,meV$, we see clear excitations at incommensurate wave vectors $Q_{IC} = (0, \pm 0.57, 0)$ and a possible signal at FM wave vectors $Q_{FM} = (0, \pm 1, \pm 1)$. On increasing energies to $E = 1 \pm 0.1, 1.3 \pm 0.1$, and $1.9 \pm 0.1\,meV$, we see excitations at $Q_{IC} = (0, \pm (K + 0.57), 0)$ with $K = 0, 1$ and no scattering at $Q_{FM}$ [Figs. 2(b,c,d)].

Figure 2(e) shows the energy dependence of excitations along the $[0, K, 0]$ direction, which reveals two dispersionless excitations at $Q_{IC} = (0, K + 0.57, 0)$ with $K = 0, 1$ and a dispersive excitation stemming from the $(0, 2, 0)$ nuclear Bragg peak. The dispersionless excitations at in-
FIG. 2: Images of 2D constant-energy cuts in the \((0,K,L)\) scattering plane at \(T = 0.3\) K. Along the \(H\) direction, the integration range is from \(-0.1\) to \(0.1\) r.l.u., while along energy, the ranges are (a) \(0.3\) to \(0.5\) meV, (b) \(0.9\) to \(1.1\) meV, (c) \(1.2\) to \(1.4\) meV, and (d) \(1.8\) to \(2.0\) meV. The BZs are indicated by white solid lines. In subplot (a), black squares labeled from 1 to 6 indicate the \(Q\) ranges of the 1D cuts along energy plotted in Fig. 3(a-f), respectively. 

\[\chi''(E, Q)\] integrated in (e) \(-0.5 < L < 0.5\) and (f) \(1.5 < L < 2.5\) r.l.u. (g,h) 1D cuts along the \(K\) direction at different \(E\) and \(Q\) across \(Q_{IC}\). Their corresponding integration ranges are marked by dashed rectangles of the same color in (e). Backgrounds fitted by linear functions are subtracted in the 1D cuts, and the curves are artificially separated along the \(y\)-axis for clarity. By fitting the peaks with Gaussian functions, we obtain \(K = 0.57 \pm 0.01\) and \(1.56 \pm 0.01\) r.l.u.

Commensurate wave vectors \(Q_{IC}\) starting from \(E = 0.2\) meV must be spin fluctuations since low-energy acoustic phonons must be dispersive and originate from nuclear Bragg peak positions. To test if the dispersive excitation from \((0,2,0)\) is indeed an acoustic phonon mode, we plot in Fig. 2(f) the 2D image of excitations along the \([0,K,2]\) direction. While the \(Q_{IC}\) excitations are no longer present, one can see a similar dispersive mode stemming from the nuclear Bragg peak \((0,2,2)\). By comparing the scattering intensity of these modes with nuclear structure factors, we conclude that the dispersive mode is the longitudinal acoustic phonon mode with a sound velocity of \(\sim 1000\) m/s comparable to the sound velocity of UTe measured by Brillouin light scattering [50]. Figures 2(g) and 2(h) show constant-energy cuts

FIG. 3: (a-f) Constant-\(Q\) cuts correspond to the \(Q_{1}-Q_{6}\) positions marked in Fig. 2(a) at \(T = 0.3, 2,\) and \(12\) K. The insets shows the incoherent or Bragg intensity from \(E = -0.2\) to \(0.2\) meV depending on whether a Bragg peak exists at each \(Q\) position. Dashed lines on the neutron energy loss side (\(E > 0\) side) are obtained by fitting the \(T = 0.3\) K data, which are then scaled up based on the Bose population factor for the 2 K and 12 K data. On the neutron energy gain side (\(E < 0\)), dashed lines are obtained by fitting the 12 K data, which are then scaled down based on the Bose population factor. (g,h) The \(\chi''(E)\) within \(Q_{1}\) and \(Q_{3}\), respectively. The pink shadow region highlights the energy range of \(3k_{B}T_{c}\) to \(5k_{B}T_{c}\), where a spin resonance appears below \(T_{c}\) in many spin-singlet unconventional superconductors [54-57].
along the [0, K, 0] direction at different energies marked in Fig. 2(c). At all energies probed, we see dispersionless incommensurate spin excitations centered around \(Q_{IC} = (0, 0.57 \pm 0.02, 0)\) with the dynamic spin correlation length of \(\sim 12 \, \text{Å}\). This is close to the commensurate AF wave vector of \((0, 0.5, 0)\), thus indicating that spin fluctuations in UTe\(_2\) are predominately AF in nature.

To see how incommensurate spin fluctuations around \(Q_{IC}\) change across \(T_c\) and determine if there are strong FM spin fluctuations, we carried out energy scans at wave vectors \(Q_1, Q_5\) as marked in Fig. 2(a) at temperatures \(T = 0.3, 2,\) and 12 K. Figures 3(a) and 3(b) summarize the key results at the incommensurate wave vectors \(Q_1 = (0, 0.57, 0)\) and \(Q_2 = (0, 1.57, 0)\), respectively. To accurately determine the nuclear incoherent scattering backgrounds from the UTe\(_2\) sample and the Cu sample holder, we chose \(Q_3 = (0, 1.1, 2)\) [Fig. 3(c)] and \(Q_4 = (0, 2, 1)\) [Fig. 3(d)] since these positions are near the incommensurate and FM positions, respectively, but are sufficiently away from the nuclear Bragg peak positions. For possible FM spin fluctuations, we consider nuclear Bragg peak positions \(Q_5 = (0, 1, 1)\) [Fig. 3(e)] and \(Q_6 = (0, 2, 0)\) [Fig. 3(f)].

Figures 3(c) and 3(d) show the nuclear incoherent scattering background at \(Q_3\) and \(Q_4\), respectively. As expected, the scattering is weakly wave vector and temperature dependent between 0.3 K and 12 K. The green dashed lines in Figs. 3(a) and 3(b) are measured incoherent background scattering. The incommensurate spin fluctuations are clearly above the background scattering and follow the Bose population factor \([6]\) and the Bose population factor \([6]\). \(\chi''(E)\) at both incommensurate wave vectors increase with increasing energy, but show no dramatic temperature dependence on warming from 0.3 K to 2 K across \(T_c\), and to 12 K. This is reminiscent of the temperature dependent \(\chi''(E)\) in Sr\(_2\)RuO\(_4\) \([22, 24]\), but clearly different from spin-singlet unconventional heavy Fermion superconductors such as CeCoIn\(_5\) \([51, 52]\), CeCu\(_2\)Si\(_2\) \([53]\), and etc., where there is a strong enhancement of \(\chi''(E)\), termed neutron spin resonance \([54, 57]\), in the pink marked energy region below \(T_c\).

Figures 3(c) and 3(f) summarize our attempt to extract FM spin fluctuations in UTe\(_2\), where the green dashed lines are incoherent scattering backgrounds measured at \(Q_1\) and \(Q_4\), respectively. Compared with Figs. 3(a) and 3(b) at \(Q_{IC}\), we see that FM spin fluctuations, if present, are much smaller in magnitude and essentially vanish above \(\sim 0.7\,\text{meV}\) within the errors of our measurements. Although temperature dependence of the scattering suggests the presence of FM spin fluctuations, they do not dominate the spin fluctuations spectra and neutron polarization analysis \([25]\) may be necessary to conclusively identify FM spin fluctuations in UTe\(_2\).

To understand these results, we have also performed the electronic structure calculations of UTe\(_2\) using the DFT+DMFT method \([45, 48]\). In a heavy fermion metal such as UTe\(_2\), there are two potential origins for the wave vector of incommensurate spin fluctuations. One is the RKKY interaction between the 5\(f\) moments, as appearing in a Kondo lattice model, which is determined by the electronic structure of the spd conduction electrons. For this electronic structure, U-5\(f\) electrons are treated as open core states (similarly via DFT to ThTe\(_2\) \([49]\)), the calculated Fermi surface is shown in Figs. 4(a,c,e) \([49]\). Noticeably, the electron momentum transfer across the two purple Fermi pockets is about 0.61\(b^*\), close to \(Q_{IC}\) observed in the INS experiments. Another potential source for the wave vector of incommensurate spin fluctuations is the Fermi-surface nesting of the U-5\(f\) heavy bands, which we have determined using the DFT+DMFT method \([49]\). The Fermi surface of the heavy bands is shown in Figs. 4(b,d,f). Specifically, at \(k_x = 2\pi/c\), the Fermi surface exhibits a rectangular shape, and the ele-
tron momentum transfer across the short edges of the rectangular shape is about 0.72b* , which is slightly away from the observed QIC. Therefore, the RKKY interaction of the 5f moments is likely driving the incommensurate spin fluctuations of UTe2, although the nesting of the strongly renormalized f-electron bands at the Fermi energy cannot be ruled out.

In summary, we have discovered that the dominant spin fluctuations in UTe2 are incommensurate near AF wave vector and extend to at least 2.6 meV. These results are consistent with DFT+DMFT calculations, indicating that incommensurate spin fluctuations in UTe2 may arise from the Q-dependence of the RKKY interaction between the U-5f moments. We expect these incommensurate spin fluctuations to play an important role in the development of the unconventional superconductivity.

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