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Absence of Spontaneous Magnetic Fields Due to Time-Reversal Symmetry Breaking in Bulk Superconducting UTe₂

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We have investigated the low-temperature local magnetic properties in the bulk of molten salt-flux (MSF) grown single crystals of the candidate odd-parity superconductor UTe₂ by zero-field muon spin relaxation (μ SR). In contrast to previous μ SR studies of UTe₂ single crystals grown by a chemical vapour transport (CVT) method, we find no evidence of magnetic clusters or electronic moments fluctuating slow enough to cause a discernible relaxation of the zero-field μ SR asymmetry spectrum. Consequently, our measurements on MSF-grown single crystals rule out the generation of spontaneous magnetic fields in the bulk that would occur near impurities or lattice defects if the superconducting state of UTe₂ breaks time-reversal symmetry. This result suggests UTe₂ is characterized by a single-component superconducting order parameter.

A superconducting phase results from the condensation of electron (Cooper) pairs into a coherent quantum state characterized by a pair wavefunction or complex order parameter consisting of an amplitude and a phase. All superconductors break gauge symmetry, which means their order parameter adopts a well defined phase below the superconducting transition temperature (T_c). Conventional superconductors have a spin-singlet pairing state with an isotropic spatial component (s -wave) mediated by electron-phonon coupling. By contrast, unconventional superconductors deviate from this pairing state and may be characterized by more elaborate superconducting order parameters due to the breaking of additional symmetries [1].

Recently-discovered UTe₂ is believed to be a rare example of an unconventional odd-parity superconductor based on an abnormally large upper critical magnetic field (H_{c2}) [2, 3], as well as the small change in the nuclear-magnetic resonance (NMR) Knight shift as a function of temperature in the superconducting state [4–6]. The observation of a nonzero polar Kerr effect (PKE) in the superconducting state of CVT-grown UTe₂ single crystals that exhibit two phase transitions in the specific heat has been taken as evidence of a time-reversal symmetry (TRS) breaking order parameter [7, 8]. Broken TRS is a defining property of chiral superconductivity [9], and hence the appearance of the PKE below T_c lends support to other signatures of chiral superconductivity detected by scanning tunneling microscopy (STM) [10] and magnetic penetration depth [11, 12] measurements of UTe₂. Chiral odd-parity superconductivity is of much current interest, because certain topological non-trivial Cooper pairing states can host Majorana zero modes with potential applications for topological quantum computing [9, 13].

The superconducting order parameter at a second order phase transition is restricted to an irreducible repre-

sentation of the total symmetry group [1]. The possible superconducting phases in UTe₂ are therefore classified by the crystalline point group symmetry D_{2h} . Order parameters that transform under the one-dimensional irreducible representations (1-D irreps) of this group do not break TRS. Consequently, to explain the signature of spontaneous TRS breaking in the polar Kerr measurements and the chiral surface states detected by STM, an odd-parity superconducting order parameter having two nearly degenerate components with a relative phase belonging to different 1-D irreps of the D_{2h} crystalline point group has been proposed [7, 10]. However, this characterization of the superconducting order parameter for UTe₂ presents a number of challenges. The PKE was observed in a sample showing two phase transitions in the specific heat at ambient pressure, as expected if the two components of the order parameter are nearly degenerate. However, as the quality of the samples improved, only a single superconducting transition was observed [14, 15]. A recent study of the PKE in CVT-grown and MSF-grown UTe₂ single crystals that exhibit a single superconducting phase transition in the specific heat found no evidence for TRS breaking superconductivity [16]. Furthermore, pulse-echo ultrasound measurements of the changes in elastic moduli across T_c in single and double phase transition CVT-grown samples [17] and recent NMR Knight shift measurements on MSF-grown UTe₂ single crystals [6] both favor a single-component odd parity superconducting order parameter.

Zero-field muon spin relaxation (ZF- μ SR) is an ideal tool for independently determining whether TRS symmetry is spontaneously broken in a superconducting state. In the bulk of a TRS breaking superconductor, inhomogeneities of the order parameter that occur near impurities, lattice defects, or around domain walls generate spontaneous currents [18]. The corresponding weak spontaneous local magnetic fields have been detected by

ZF- μ SR in numerous unconventional superconductors — most notably UPt_3 [19] and Sr_2RuO_4 [20], two compounds in which TRS breaking has been confirmed by polar Kerr measurements [21, 22]. Both superconductors have been considered as likely chiral superconductors, although Sr_2RuO_4 is no longer believed to be a candidate for odd-parity superconductivity [23].

As for UTe_2 , the relaxation rate of the ZF- μ SR signal from CVT-grown single crystals exhibiting single or double transitions in the specific heat was found to be dominated by inhomogeneous freezing of magnetic clusters [24, 25], which thwarted sensitivity to the potential onset of weak spontaneous fields at T_c . As discussed in Ref. [25], the magnetic clusters are likely responsible for the residual linear term in the temperature dependence of the specific heat (C) below T_c and the low-temperature upturn in C/T versus T that are ubiquitous in UTe_2 samples grown via the CVT method. Moreover, a saturation in the growth of the total volume of the magnetic clusters and an abrupt slowing down of their fluctuation rate was observed near T_c . By contrast, current UTe_2 single crystals grown by the MSF method have less disorder, and correspondingly a much larger residual resistivity ratio (RRR) as well as a substantially smaller residual T -linear term in the specific heat compared to CVT-grown single crystals [15]. A potential origin of the magnetic clusters is discussed later, but they are presumably induced by defects that disrupt long-range electronic correlations, as suggested in Ref. [26]. Hence while MSF-grown UTe_2 single crystals may contain trace amounts of the ferromagnetic (FM) impurities U_7Te_{12} and U_3Te_5 [15], magnetic clusters are expected to be sparse.

Here we present results of a ZF- μ SR study of MSF-grown UTe_2 single crystals. The single crystals all come from the same growth batch and exhibit bulk superconductivity below $T_c = 2.01$ K, as determined from the midpoint of the specific heat jump shown in Fig. 1(a). The crystals have an RRR value of 200 and the coefficient of the residual T -linear term in the specific heat below T_c is approximately 9 mJ/mol-K². Figure 1(b) shows a comparison of the temperature dependence of the bulk magnetic susceptibility (χ) for a magnetic field of 1 kOe applied along the three principal crystallographic axes. In contrast to the low-field behavior of $\chi(T)$ along the a -axis in CVT-grown UTe_2 single crystals [2, 14], $\chi_a(T)$ does not exhibit an upturn below $T \sim 10$ K.

For the ZF- μ SR measurements a mosaic of 24 single crystals was mounted on a 5×17 mm pure silver (Ag) backing plate thermally anchored to the Ag sample holder of an Oxford Instruments top-loading dilution refrigerator at the end of the M15 surface muon beamline at TRIUMF, Vancouver, Canada. The MSF-grown UTe_2 single crystals covered 83 % of the Ag backing plate. The c axis of each single crystal was aligned within 2° of the normal of the Ag backing plate. For the zero-field measurements, stray external magnetic fields at the sample position were reduced to $\lesssim 41$ mG using field compensation coils and the precession signal of muonium (Mu

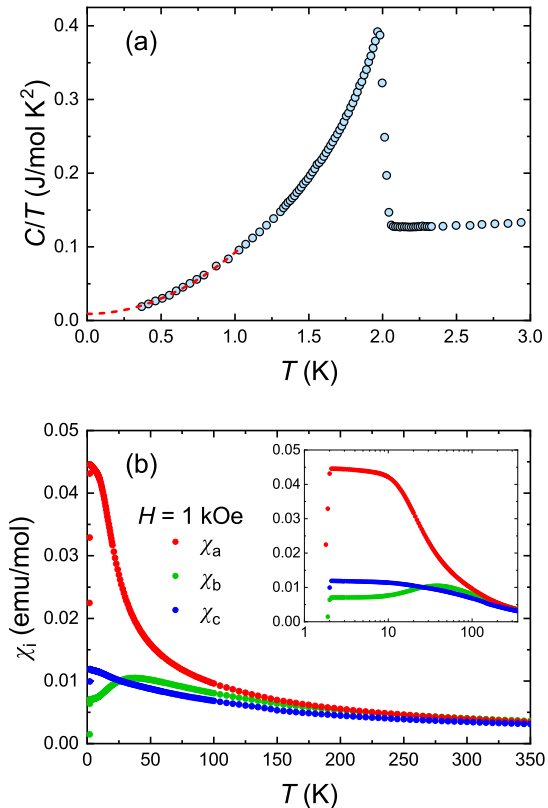


FIG. 1. (a) Temperature dependence of the specific heat (C) for one of the MSF-grown UTe_2 single crystals, plotted as C/T vs. T . The dashed curve is a fit of the data below $T = 1$ K to $C/T = \gamma^* + \beta T^2$, which yields $\gamma^* = 9.1(6)$ mJ/mol-K² and $\beta = 83(1)$ mJ/mol-K³. (b) Temperature dependence of the bulk magnetic susceptibility of the single crystal for a magnetic field $H = 1$ kOe applied parallel to the three different principal crystallographic axes. The inset shows the same data as a semi-logarithmic plot.

$\equiv \mu^+ e^-$) in intrinsic Si as a sensitive magnetometer [27]. The ZF- μ SR measurements were performed by implanting nearly 100 % spin-polarized positive muons (μ^+) into the sample with the initial muon spin polarization $\mathbf{P}(0)$ antiparallel to the muon beam direction (defined as the z -axis direction) and parallel to the crystalline c -axis. The time evolution of the muon spin polarization $P_z(t)$ was determined by detecting the muon decay positrons in a pair of opposing detectors positioned outside of the dilution refrigerator in front and behind the sample.

The ZF- μ SR asymmetry spectra are well described by the following equation

$$A(t) = a_0 P_z(t) = a_1 G_z^{\text{GKT}}(\Delta, t) e^{-\lambda_1 t} + a_2 e^{-\lambda_2 t}, \quad (1)$$

where $G_z^{\text{GKT}}(\Delta, t)$ is a static Gaussian Kubo-Toyabe (GKT) function, characterized by the linewidth Δ/γ_μ ($\gamma_\mu/2\pi$ is the muon gyromagnetic ratio) of a Gaussian distribution of local magnetic fields [28]. Figure 2 shows

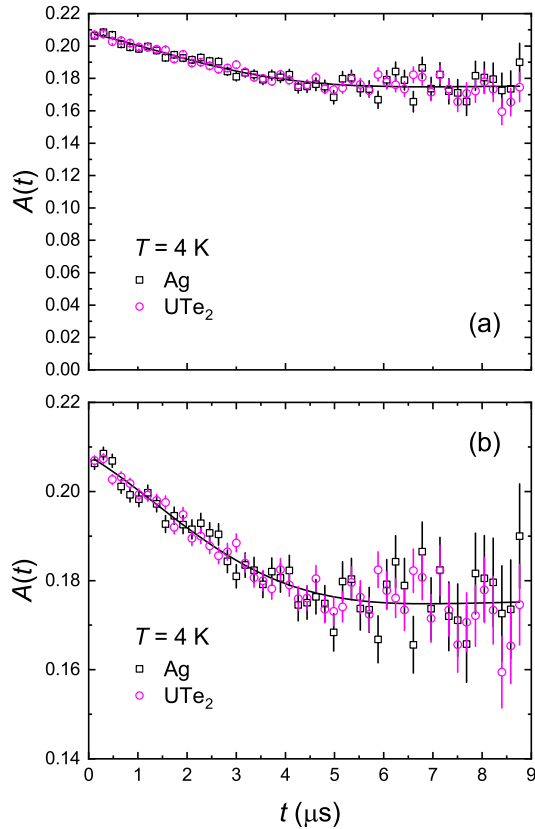


FIG. 2. (a) Comparison of the ZF- μ SR asymmetry spectra for the Ag backing plate with (open circles) and without (open squares) the MSF-grown UTe_2 single crystals. The solid curve is a fit of the ZF- μ SR spectrum for the Ag plate to Eq. (1). (b) Same as (a), but showing a zoom in on the vertical axis.

a comparison of ZF- μ SR asymmetry spectra for $T=4$ K recorded for measurements of the Ag backing plate with and without the MSF-grown UTe_2 single crystals. The observed relaxation of the ZF- μ SR signal with time for the Ag plate without the sample is due to muons landing in material elsewhere within the dilution refrigerator. This contribution is described by the first term in Eq. (1), which makes up ~ 16 % of the total ZF- μ SR signal ($a_1/a_0 \sim 16$ %). The remaining 84 % of the ZF- μ SR asymmetry spectrum is due to muons stopping in the Ag plate (~ 64 %) and a portion of the Ag sample holder (~ 20 %). This component of the ZF- μ SR signal is essentially non-relaxing ($\lambda_2 \sim 0$), as Ag does not possess electronic moments and has only very small nuclear moments that do not cause an appreciable muon spin depolarization in the data time window. The relaxation of the ZF- μ SR signal by randomly oriented nuclear moments is also negligible in UTe_2 , because the only stable uranium isotope with non-zero nuclear spin is depleted ^{235}U , which has a natural abundance of 0.20 %, and the natural abundance of the tellurium isotopes with nuclear

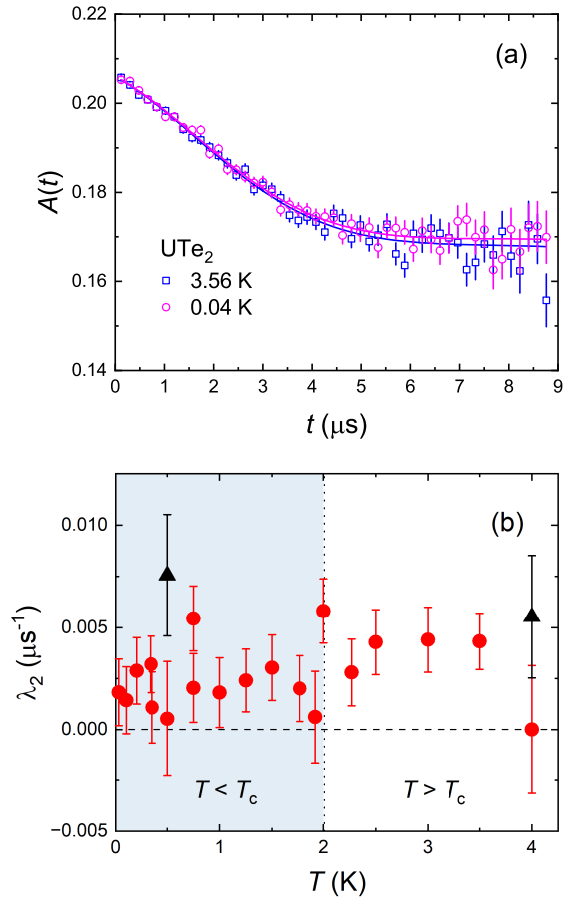


FIG. 3. (a) Comparison of the ZF- μ SR asymmetry spectra recorded for the MSF-grown UTe_2 single crystals at $T=3.56$ K and $T=0.04$ K. The solid curves are fits of the ZF- μ SR spectra to Eq. (1) obtained from a global fit of the ZF- μ SR spectra at all temperatures with a_1 , a_2 , λ_1 and Δ as shared fitting parameters. (b) Temperature dependence of λ_2 obtained from the global fit described in (a). The solid black triangles are values of λ_2 for the Ag backing plate without the sample from fits assuming the same values of λ_1 and Δ generated by the global fit.

spin, ^{123}Te and ^{125}Te , are only 0.89 % and 7 %, respectively. No discernible difference is observed between the ZF- μ SR spectra for the Ag plate and the UTe_2 single crystals mounted on the Ag plate, indicating there are no electronic moments fluctuating in the UTe_2 sample at this temperature that are slow enough to cause additional relaxation of the ZF- μ SR signal. With the sample in place, the non-relaxing component is due to muons stopping in the UTe_2 single crystals, in the Ag backing plate, and in a portion of the Ag sample holder. These contributions are separable in a transverse-field (TF) μ SR measurement, due to a sizeable muon Knight shift in UTe_2 [29]. The contribution of the UTe_2 sample to the non-relaxing part of the asymmetry spectrum is estimated to be at least 63 % (or 53 % of the total ZF- μ SR signal) from

TF- μ SR measurements for an applied magnetic field of 20 kOe.

The ZF- μ SR asymmetry spectrum for the UTe₂ sample was recorded for 19 different temperatures between $T = 0.03$ K and $T = 4.0$ K. A global fit of the corresponding 19 different ZF- μ SR spectra to Eq. (1) was carried out with the fit parameters a_1 , a_2 , λ_1 and Δ being shared parameters for all temperatures. The global fit yielded the values $a_1/a_0 = 16.1(7)$ %, $a_2/a_0 = 83.9(7)$ %, $\lambda_1 = 0.158(7)$ μs^{-1} , and $\Delta = 0.280(7)$ μs^{-1} . Representative fits to ZF- μ SR asymmetry spectra included in the global fit are shown in Fig. 3(a) and the temperature dependence of the exponential relaxation rate λ_2 generated from the global fit is shown in Fig. 3(b). There is no systematic increase in λ_2 with decreasing temperature, as previously observed in CVT-grown single crystals [24, 25]. Hence, there are no electronic moments fluctuating slow enough to cause a detectable relaxation of the ZF- μ SR spectrum. More importantly, there is no increase in λ_2 near T_c , and hence no evidence of spontaneous magnetic fields in the bulk associated with a TRS broken superconducting state.

Extremely slow intra U-ladder FM fluctuations along the a axis have been inferred from NMR experiments on CVT-grown single crystals [26, 30, 31] that are far below the rate of spin fluctuations probed in inelastic neutron scattering studies [32, 33]. The low-energy FM spin fluctuations may be a consequence of vacancies in the U-ladder structure associated with a small U deficiency in some CVT-grown UTe₂ samples [15]. In particular, spins next to U vacancies may couple more strongly to the remaining neighboring spins [34], creating slowly fluctuating

magnetic clusters that are detectable on the time scales of the NMR and μ SR measurements. However, it is clear from the current findings that the FM-like fluctuations observed in our initial ZF- μ SR study of CVT-grown single crystals [24] are not detectable in the absence of significant disorder. Consequently, it is an open question as to whether FM fluctuations are an intrinsic property of UTe₂.

To summarize, the main result of our ZF- μ SR study of MSF-grown single crystals is the absence of spontaneous local magnetic fields in the bulk, which are expected for a superconducting state that breaks TRS. This is in agreement with recent polar Kerr [16], ultrasound [17] and NMR [6] measurements that do not support a two-component superconducting order parameter.

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