Coexistence of ferromagnetic fluctuations and superconductivity in the actinide superconductor UTe$_2$

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We report low-temperature muon spin relaxation/rotation ($\mu$SR) measurements on single crystals of the actinide superconductor UTe$_2$. Below 5 K we observe a continuous slowing down of magnetic fluctuations that persists through the superconducting (SC) transition temperature ($T_c = 1.6$ K), but we find no evidence of long-range or local magnetic order down to 0.025 K. The temperature dependence of the dynamic relaxation rate down to 0.4 K agrees with the self-consistent renormalization theory of spin fluctuations for a three-dimensional weak itinerant ferromagnetic metal. Our $\mu$SR measurements also indicate that the superconductivity coexists with the magnetic fluctuations.

The unusual physical properties of intermetallic uranium-based superconductors are primarily due to the U-5f electrons having both localized and itinerant character. In a subclass of these compounds, superconductivity coexists with ferromagnetism. In URhGe and UCoGe [1,2] this occurs at ambient pressure, whereas superconductivity appears over a limited pressure range in UGe$_2$ and UIr [3,4]. With the exception of UIr, the Curie temperature of these ferromagnetic (FM) superconductors significantly exceeds $T_c$, and the upper critical field $H_{c2}$ at low temperatures greatly exceeds the Pauli paramagnetic limiting field. These observations indicate that the SC phases in these materials are associated with spin-triplet Cooper pairing, and likely mediated by low-lying magnetic fluctuations in the FM phase [4-8]. The triplet state is specifically non-unitary, characterized by a non-zero spin-triplet Cooper pair magnetic moment due to alignment of the Cooper pair spins with the internal field generated by the pre-existing FM order.

Very recently, superconductivity has been observed in UTe$_2$ at ambient pressure below $T_c \sim 1.6$ K [9]. The superconductivity in UTe$_2$ also seems to involve spin-triplet pairing, as evidenced by a strongly anisotropic critical field $H_{c2}$ that exceeds the Pauli limit, and by the lack of any temperature dependence of the $^{125}$Te nuclear magnetic resonance (NMR) Knight shift through and below $T_c$. Furthermore, a large residual value of the Sommerfeld coefficient $\gamma$ is observed in the SC state, which is nearly 50 % of the value of $\gamma$ above $T_c$ [9,10]. This suggests that only half of the electrons occupying states near the Fermi surface participate in spin-triplet pairing, while the remainder continue to form a Fermi liquid. While this is compatible with UTe$_2$ being a non-unitary spin-triplet superconductor (in which the spin of the Cooper pairs are aligned in a particular direction), unlike URhGe, UCoGe and UGe$_2$, there is no experimental evidence for ordering of the U-5f electron spins prior to the onset of superconductivity. Instead, the normal-state a-axis magnetization exhibits scaling behavior indicative of strong magnetic fluctuations associated with metallic FM quantum criticality [9].

Little is known about the nature of the magnetism in UTe$_2$ below $T_c$, including whether it competes or coexists with superconductivity. Specific heat measurements show no anomaly below $T_c$ [9,10], but like other bulk properties may be insensitive to a FM transition with little associated entropy (such as small-moment itinerant ferromagnetism). NMR experiments indicate the development of low-frequency longitudinal magnetic fluctuations along the a-axis, but the corresponding NMR signal vanishes below 20 K [11]. Here we report $\mu$SR experiments on UTe$_2$ single crystals that confirm the absence of FM order below $T_c$ and demonstrate the presence of magnetic fluctuations consistent with FM quantum criticality that coexist with superconductivity.

The UTe$_2$ single crystals were grown by a chemical vapor transport method. Powder x-ray diffraction (XRD), neutron scattering and Laue XRD measurements indicate that the single crystals are of high quality. The details of the sample growth and characterization are given in Ref. [9]. Zero-field (ZF), longitudinal-field (LF), transverse-field (TF), and weak transverse-field (wTF) $\mu$SR measurements were performed on a mosaic of 21 single crystals. Measurements over the temperature range $0.02 \leq T \leq 5$ K were achieved using an Oxford Instruments top-loading dilution refrigerator on the M15 surface muon beam line at TRIUMF. The UTe$_2$ single crystals covered $\sim 70 \%$ of a $12.5 \text{ mm} \times 14 \text{ mm}$ silver (Ag) sample holder. For the ZF-$\mu$SR experiments, stray external magnetic fields at the sample position were re-
duced to $\lesssim 20$ mG using the precession signal due to muonium ($\mu \equiv \mu^+ e^-$) in intrinsic Si as a sensitive magnetometer \footnote{Reference}. The TF and LF measurements were performed with a magnetic field applied parallel to the linear momentum of the muon beam (which we define to be in the $z$-direction). The wTF experiments were done with the field applied perpendicular to the beam (defined to be the $x$-direction). The initial muon spin polarization $P(0)$ was directed parallel to the $z$-axis for the ZF, LF and wTF experiments, and rotated in the $x$-direction for the TF measurements. The $c$- or $a$-axis of the single crystals were arbitrarily aligned in the $z$-direction. All error bars herein denote uncertainties of one standard deviation.

Representative ZF-$\mu$SR asymmetry spectra for UTe$_2$ at $T=0.04$ K and 4.9 K are shown in the inset of Fig. \ref{fig:ZF}. No oscillation indicative of magnetic order is observed in any of the ZF-$\mu$SR spectra, which are well described by a three-component function consisting of two exponential relaxation terms plus a temperature-independent background term due to muons stopping outside the sample

$$A(t) = A(0)P_z(t) = A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} + A_B e^{-\sigma^2 t^2}.$$ \hspace{1cm} (1)

The sum of the sample asymmetries $A_1 + A_2$ is a measure of the recorded decay events originating from muons stopping in the sample. A global fit of the ZF spectra for all temperatures assuming common values of the asymmetry parameters, yielded $A_1/A(0) = 24\%$, $A_2/A(0) = 29\%$ and $A_B/A(0) = 47\%$. A previous $\mu$SR study of UGe$_2$ identified two muon stopping sites, with site populations of $\sim 45\%$ for one site and $\sim 55\%$ for the other \footnote{Reference}, in excellent agreement with the results here. The temperature variation of the ZF relaxation rates $\lambda_1$ and $\lambda_2$ are shown in Fig. \ref{fig:ZF}. The monotonic increase in $\lambda_1$ and $\lambda_2$ with decreasing temperature indicates that the local magnetic field sensed at each muon site is dominated by a slowing down of magnetic fluctuations, as explained below. The difference in the size of the relaxation rates reflects a difference in the dipolar and hyperfine couplings of the U-$5f$ electrons to the muon at the two stopping sites.

To confirm the dynamic nature of the magnetism, LF-$\mu$SR measurements were performed for various longitudinal applied fields $H_{LF}$. Representative LF-$\mu$SR asymmetry spectra for $T=2.5$ K and 0.25 K are shown in Fig. \ref{fig:LF}. The LF signals are reasonably described by Eq. \ref{eq:lambda}. Figure \ref{fig:lambda} shows the dependence of the fitted relaxation rates $\lambda_1$ and $\lambda_2$ on $H_{LF}$. Also shown in Fig. \ref{fig:lambda} are fits of the field dependence of the larger relaxation rate $\lambda_1$ to the Redfield equation \footnote{Reference}.

$$\lambda_1(H_{LF}) = \frac{\lambda_1(H_{LF} = 0)}{1 + (\gamma H_{LF} \tau)^\alpha},$$ \hspace{1cm} (2)
where \( \lambda_1(H_{LF} = 0) = 2\gamma_\mu^2 \langle B^2_{loc} \rangle \tau \), \( \langle B^2_{loc} \rangle \) is the mean of the square of the transverse components of the time-varying local magnetic field at the muon site, and \( \tau \) is the characteristic fluctuation time. The fit for 2.5 K yields \( \lambda_1(H_{LF} = 0) = 0.065(5) \mu s^{-1} \), \( \tau = 8(3) \times 10^{-10} \) s and \( B_{loc} = 76(22) \) G, whereas the fit for 0.25K yields \( \lambda_1(H_{LF} = 0) = 0.70(9) \mu s^{-1} \), \( \tau = 9(2) \times 10^{-8} \) s and \( B_{loc} = 23(4) \) G. We could not confirm similar fluctuation rates at the second muon site, because \( \lambda_2 \) is much smaller and not well resolved for most fields.

Above \( \sim 150 \) K, the magnetic susceptibility \( \chi(T) \) of UTe\(_2\) is described by a Curie-Weiss law with an effective magnetic moment \( \mu_{\text{eff}} \) that is close to the expected value \( (3.4\mu_B/U) \) for localized U-5\( f \) electrons and a Weiss temperature \( \theta \sim -100 \) K [15]. However near \( \sim 35 \) K, \( \chi(T) \) for \( H \parallel b \)-axis exhibits a maximum that suggests the U-5\( f \) electrons may become more itinerant at lower temperatures. Figure 3 shows the temperature dependence of \( \lambda_1/T \), where \( \lambda_1 \equiv 1/T_1 \) is the larger of the two dynamic ZF exponential relaxation rates. The phenomenological self-consistent renormalization (SCR) theory for itinerant ferromagnetism [16], predicts that \( 1/T_1 \propto T^{-4/3} \) near a FM quantum critical point (QCP) in a three-dimensional metal [17]. As shown in Fig. 3, this behavior is observed down to \( T = 0.4 \) K. The deviation below \( \sim 0.3 \) K suggests a breakdown in SCR theory close to the presumed FM QCP. The inset of Fig. 4 shows that \( T_1 \) (which is proportional to the inverse of the imaginary part of the dynamical local spin susceptibility) goes to zero as \( T \rightarrow 0 \), which provides evidence for the ground state of UTe\(_2\) being close to a FM QCP.

Figure 4 shows \( \lambda_1/T \) for zero field. The solid blue line is a fit of the data over \( 0.4 \leq T \leq 4.9 \) K to the power-law equation \( 1/T_1 \propto T^{-n} \), which yields the exponent \( n = 1.35(\pm 0.04) \). The dashed line is a similar fit over \( 0.037 \leq T \leq 0.3 \) K, yielding \( n = 1.12(\pm 0.14) \). The inset shows a plot of \( T/\lambda_1 \) verses \( T^{1.12} \) with a linear fit that yields the \( \lambda = 0 \) intercept \( T/\lambda_1 = (0.7(\pm 4.2) \times 10^{-3} \) K \( \mu s.\)

FIG. 3. (Color online) Field dependence of the relaxation rates \( \lambda_1 \) and \( \lambda_2 \) from the fits of the LF-\( \mu \)SR asymmetry spectra at (a) 2.5 K, and (b) 0.25 K. The solid red curves are fits of \( \lambda_1(H_{LF}) \) to Eq. 2.

FIG. 4. (Color online) Temperature dependence of \( \lambda_1/T \) (\( \equiv 1/T_1 T \)) for zero field. The solid blue line is a fit of the data over \( 0.4 \leq T \leq 4.9 \) K to the power-law equation \( 1/T_1 T \propto T^{-n} \), which yields the exponent \( n = 1.35(\pm 0.04) \). The dashed line is a similar fit over \( 0.037 \leq T \leq 0.3 \) K, yielding \( n = 1.12(\pm 0.14) \). The inset shows a plot of \( T/\lambda_1 \) verses \( T^{1.12} \) with a linear fit that yields the \( \lambda = 0 \) intercept \( T/\lambda_1 = (0.7(\pm 4.2) \times 10^{-3} \) K \( \mu s.\)
signals were fit to the sum

\[ A(t) = A(0) P_z(t) = \sum_{i=1}^{2} A_i e^{-\Lambda_i t} \cos(2\pi \nu_i t + \psi) + A_B e^{-\Lambda_B t} \cos(2\pi \nu_B t + \psi), \]

where \( \psi \) is the initial phase of the muon spin polarization \( P(0) \) relative to the \( z \)-direction. The exponentially-damped terms account for muons stopping at the two sites in the sample, and the Gaussian-damped term accounts for muons that missed the sample. The precession frequencies \( \nu_i \) are a measure of the local field \( B_{\mu,i} \) sensed by the muon at the two stopping sites, where \( \nu_i = (\gamma_i/2\pi) B_{\mu,i} \) and \( \gamma_i/2\pi \) is the muon gyromagnetic ratio. The applied 1 kOe field induces a polarization of the \( 5f \) moments and a corresponding relative muon precession frequencies, and (c) TF relaxation rates.

The temperature dependence of \( \nu_1 \) and \( \nu_2 \) are shown in Fig. 6(b). Below \( T \sim 1.6 \) K there is a decrease in \( \nu_1 \) and \( \nu_2 \) compatible with the estimate of \( \pm 0.2\% \) for the SC diamagnetic shift from the relation \( 18 \rightarrow -4\pi M = (H_{c2} - H)/[1.18(2\kappa^2 - 1) + n] \), with \( H_{c2} = 200 \) kOe, \( H = 1 \) kOe, \( \kappa = 200 \), and \( n \leq 1 \). However, the temperature dependence of the TF relaxation rates \( \Lambda_1 \) and \( \Lambda_2 \) [see Fig. 6(c)] do not exhibit a significant change in behavior at \( T_c \). This indicates that \( \Lambda_1 \) and \( \Lambda_2 \) are dominated by the internal magnetic field distribution associated with the magnetic fluctuations and the London penetration depth \( \lambda_L \) is quite long — as is the case for other uranium-based superconductors in which \( \lambda_L \geq 10,000 \lambda \). The magnetic fluctuations may also contribute to \( \nu_1(T) \) and \( \nu_2(T) \) by adding or opposing the SC diamagnetic shift. Interestingly, the SC diamagnetic shift is not observed in the NMR Knight shift data for a powder sample [9], although this may be a consequence of anisotropic averaging of the NMR interactions.

In conclusion, we observe a gradual slowing down of magnetic fluctuations with decreasing temperature below 5 K, consistent with weak FM fluctuations approaching a magnetic instability. However, we find no evidence for magnetic order down to 0.025 K. Hence there is no phase transition to FM order in UTe2 preceding or coinciding with the onset of superconductivity. The magnetic volume fraction is not significantly reduced below \( T_c \), indicating that the superconductivity coexists with the fluctuating magnetism. Lastly, we note that because the relaxation rate of the ZF-\( \mu \)SR signal below 5 K is dominated by dynamic local fields, it is not possible to determine whether spontaneous static magnetic fields occur below \( T_c \) due to time-reversal symmetry breaking in the SC state.

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[1] D. Aoki, A. Huxley, E. Ressouche, D. Braithwaite, J. Flouquet, J.P. Brison, E. Lhotel, C. Paulsen, Nature 413, 613 (2001).
[2] N. T. Huy, A. Gasparini, D. E. de Nijs, Y. Huang, J. C. P. Klaasse, T. Gortenmulder, A. de Visser, A. Hamann, T. Görlach, H. v. Löhnseysen, Phys. Rev. Lett. 99, 067006 (2007).
[3] S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Sheikin, D. Braithwaite, and J. Flouquet, Nature 406, 587 (2000).
[4] T. Akazawa, H. Hidaka, T. Fujiwara, T. C. Kobayashi, E. Yamamoto, Y. Haga, R. Settai, and Y. Onuki, J. Phys.: Condens. Matter 16, L29-L32 (2004).
[5] D. Fay and J. Appel, Phys. Rev. B 22, 3173 (1980).
[6] R. Roussev and A. J. Millis, Phys. Rev. B 63, 140504(R) (2001).
[7] T. R. Kirkpatrick, D. Belitz, T. Vojta, and R. Narayanan, Phys. Rev. Lett. 87, 127003 (2001).
[8] N. Tateiwa, Y. Haga, and E. Yamamoto, Phys. Rev. Lett. 121, 237001 (2018).
[9] S. Ran, C. Eckberg, Q.-P. Ding, Y. Furukawa, T. Metz, S. R. Saha, I.-L. Liu, M. Zic, H. Kim, J. Paglione, and N. P. Butch, arXiv:1811.11808.
[10] D. Aoki, A. Nakamura, F. Honda, D. Li, Y. Homma, Y. Shimizu, Y. J. Sato, G. Knebel, J.-P. Brison, A. Pourret, D. Braithwaite, G.0 Lapertot, Q. Niu, M. Vališka, H. Harima, and J. Flouquet, J. Phys. Soc. Jpn. 88, 043702 (2019).
[11] Y. Tokunaga, H. Sakai, S. Kambe, T. Hattori, N. Higa, G. Nakamine, S. Kitagawa, K. Ishida, A. Nakamura, Y. Shimizu, Y. Homma, D. Li, F. Honda, and D. Aoki, J. Phys. Soc. Jpn. 88, 073731 (2019).
[12] G. Morris and R. Heffner, Physica B 326, 252 (2003).
[13] S. Sakarya, P. C. M. Gubbens, A. Yaouanc, P. Dalmas de Réotier, D. Andreica, A. Amato, U. Zimmermann, N. H. van Dijk, E. Brück, Y. Huang, and T. Gortenmulder, Phys. Rev. B 81, 024429 (2010).
[14] A. Schenck, Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics (Adam Hilger Ltd., Bristol and Boston, 1985).
[15] S. Ikeda, H. Sakai, D. Aoki, Y. Homma, E. Yamamoto, A. Nakamura, Y. Shiokawa, Y. Haga, and Y. Onuki, J. Phys. Soc. Jpn. 75, 116 (2006).
[16] T. Moriya, J. Magn. Magn. Mater. 100, 261 (1991).
[17] A. Ishigaki and T. Moriya, J. Phys. Soc. Jpn. 65, 3402 (1996).
[18] A. A. Abrikosov, J. Phys. Chem. Solids 2, 199-208 (1957).
[19] F. Gross, K. Andres, and B. S. Chandrasekhar, Physica C 162, 419 (1989).