Evidence for Weak Itinerant Long-Range Magnetic Correlations in UGe$_2$


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(Received 12 September 2001; published 11 September 2002)

Positive muon spin relaxation measurements performed on the ferromagnet UGe$_2$ reveal, in addition to the well-known localized 5f-electron density responsible for the bulk magnetic properties, the existence of itinerant quasistatic magnetic correlations. Their critical dynamics is well described by the conventional dipolar Heisenberg model. These correlations involve small magnetic moments.

DOI: 10.1103/PhysRevLett.89.147001 PACS numbers: 74.70.Tx, 76.75.+i

The discovery of superconductivity below 1 K within a limited pressure range in the ferromagnet UGe$_2$ [1–4] provides an unanticipated example of coexistence of superconductivity and strong ferromagnetism. The electronic pairing mechanism needed for superconductivity is believed to be magnetic in origin. However, it is amazing that ferromagnetically ordered uranium magnetic moments with so large magnitude (~1.4 $\mu_B$ at ambient pressure as deduced from magnetization measurements) are directly involved. Since the pairing must involve the conduction electrons, it is important to characterize their magnetic properties. Because of the restrictions imposed by their magnetic form factor, this cannot be done by diffraction techniques. As the muons localize in interstitial sites, they have the potentiality to yield information on the conduction electrons. Here we show, using the muon spin relaxation technique, that UGe$_2$ is actually a dual system where two substrates of f electrons coexist. We indeed report the existence at ambient pressure of itinerant long-range magnetic correlations with magnetic moments of ~0.02 $\mu_B$ and a spectral weight in the megahertz range. A quantitative understanding of this state is moreover reached assuming that these correlations involve only long wavelength fluctuation modes.

UGe$_2$ is a ferromagnet with a Curie temperature $T_C \approx 52$ K which crystallizes in the orthorhombic ZrGa$_2$ crystal structure (space group Cmmm) [5,6]. Magnetic measurements indicate a strong magnetocrystalline anisotropy [3,7,8] with easy magnetization axis along the a axis.

We present results obtained by the muon spin relaxation ($\mu$SR) technique. Fully polarized muons are implanted into the studied sample. Their spin (1/2) evolves in the local magnetic field, $B_{loc}$, until they decay into positrons. Since the positron is emitted preferentially in the direction of the muon spin at the decay time, it is possible to follow the evolution of the muon spin polarization [9,10]. The measured physical parameter is the so-called asymmetry which characterizes the anisotropy of the positron emission. Below $T_C$, if $B_{loc}$ has a component perpendicular to the initial muon beam polarization, $S_\mu$ (taken parallel to Z), we expect the asymmetry to display spontaneous oscillations with an amplitude maximum for $B_{loc} \perp S_\mu$. On the other hand, if $B_{loc} \parallel S_\mu$, the asymmetry can be written as the product of an initial asymmetry related to sample, $a_s$, and the muon spin relaxation function, $P_Z(t)$, which monitors the dynamics of $B_{loc}$.

UGe$_2$ crystals were grown from a polycrystalline ingot using a Czochralski tri-arc technique [7]. We present results for two samples. Each consists of pieces cut from the crystals, put together to form a disk and glued on a silver backing plate. They differ by the orientation (either parallel or perpendicular) of the a axis relative to the normal to the sample plane. The measurements were performed at the EMU spectrometer of the ISIS facility, from 5 K up to 200 K, mostly in zero field. Additional $\mu$SR spectra were recorded with a longitudinal field.

We found that the temperature dependence of $a_s$ for $S_\mu \parallel a$ is consistent with $B_{loc} \parallel a$. In agreement with that conclusion, a spontaneous muon spin precession resulting in wiggles in the asymmetry is observed for $S_\mu \perp a$. Defining $T_C$ as the temperature at which the wiggles disappear, we found $T_C = 52.49(2)$ K. This value coincides with the maximum of the relaxation rate (to be evidenced below) for $S_\mu \parallel a$ and $S_\mu \perp a$.

In this Letter, we focus on the description of data taken around the Curie point.

All the spectra were analyzed as a sum of two components: $a[P_Z(t) + a_{bg}]$. The first component describes the $\mu$SR signal from the sample and the second accounts for the muons stopped in the background, i.e., the cryostat walls and sample holder. In zero field, for all relevant temperatures and for the two orientations of $S_\mu$ relative to a, $P_Z(t)$ is well described by an exponential function: $P_Z(t) = \exp(-\lambda_Zt)$, where $\lambda_Z$ measures the spin-lattice relaxation rate at the muon site. An example is shown in Fig. 1. $a_{bg}$, which is basically temperature independent, was measured for $S_\mu \perp a$ and $T < T_C$ as the constant background signal. We got $a_{bg} = 0.077$ [11]. For $S_\mu \parallel a$, it could be estimated only from the sample size...
B Lorentzian distribution for measurements. The magnetic signal that we observe has dependence at small fields.

Scaling laws from mode coupling theory [16]. The two scaling variables at play depend on two material parameters determining, respectively, the amount of longitudinal (L) and transverse (T) fluctuations probed by parameters: $\xi_0$, the magnetic correlation length at $T = 2T_C$, and $q_D$, the dipolar wave vector which is a measure of the strength of the exchange interaction relative to the dipolar energy. This model initially derived for the paramagnetic phase applies also below $T_C$ [14].

Specifically, the model predicts that $\lambda_2(T) = W[a_L I_L(T) + a_T I_T(T)]$, where $I_L$ [17] are scaling functions obtained from mode coupling theory and $a_{L,T}$ are parameters determining, respectively, the amount of longitudinal (L) and transverse (T) fluctuations probed by the measurements. The L,T indices denote the orientation relative to the wave vector of the fluctuation mode. $a_{L,T}$ depend only on muon site properties. The result of the fit of $\lambda_2(T)$ is shown in the insets of Fig. 2. The divergence of $\lambda_2$ at $T_C$ is strongly reduced by the effect of the dipolar interaction [16]. The temperature scale gives the product $q_D \xi_0^D$ [13]. For $S_\mu \perp a$, we get $q_D \xi_0^D = 0.021(2)$, and for $S_\mu \parallel a$, $q_D \xi_0^D = 0.043(2)$ and $q_D \xi_0^D = 0.020(2)$. The index + (-) on $\xi_0$ specifies that we consider the paramagnetic phase.
(ferromagnetic) state. \( \xi_0(S_{\mu} \parallel a) > \xi_0(S_{\mu} \perp a) \) in the paramagnetic state, suggesting that the magnetic correlations are somewhat anisotropic. The fact that \( \xi_0^p > \xi_0 \) is an expected feature \[9\]. The relaxation rate scale yields \( \mathcal{W}^+ a_L = 0.140(4) \) MHz and \( \mathcal{W}^- a_L = 0.20(2) \) MHz for \( S_{\mu} \parallel a \). The transverse contribution to \( \lambda_2 \) for both \( T < T_C \) and \( T > T_C \) is more difficult to estimate since \( a_T \) is found much lower than \( a_L \). Reasonable fits are obtained with \( a_T/a_L = 0.036(14) \). We have computed \( a_L \) and \( a_T \) for different possible muon sites and found only one site satisfying \( a_T < a_L/2 \). This is site 2b (in Wyckoff notation) of coordinates (0, 1/2, 0) for which \( a_L = 1.2486 \), \( a_T = 0.0386 \). We then deduce \( \mathcal{W}^+ = 0.112(3) \) MHz and \( \mathcal{W}^- = 0.161(16) \) MHz. The scale deduced from the measurements with \( S_{\mu} \perp a \) is about twice as large, pointing again to the weak anisotropy of the magnetic correlations.

In order to further characterize the relaxation near \( T_C \), we performed at a given temperature longitudinal field measurements for the two orientations of \( S_{\mu} \) relative to \( a \). The field responses for the two geometries are similar. An illustration is given in Fig. 1. Surprisingly, the spectra are field dependent at extremely low external field, \( B_{\text{ext}} \), prov- ing that the probed magnetic fluctuations are quasistatic (fluctuation rate in the MHz range) and, since \( \lambda_2 \) is small, the associated magnetic moment must be small as well. Quantitatively, the field dependence of \( P_{\lambda_2}(t) \) cannot be described consistently either by a simple exponential relaxation form (see the lower panel of Fig. 1) or by a relaxation function computed with the strong collision model assuming an isotropic Gaussian component field distribution \[18\]. On the other hand, the relaxation is well explained if we assume that the distribution of \( B_{\text{loc}} \) is squared Lorentzian \[19\]. We write \( P_{\lambda_2}(t) = P_{\lambda_2}(\Delta_{\text{Lor}}, \nu_L, t) \), where \( \Delta_{\text{Lor}} \) characterizes the width of the field distribution and \( \nu_L \) its fluctuation rate \[20\]. A global fit of the spectra (\( B_{\text{ext}} = 0, 0.2, 0.4, 0.6, 0.8, 1.0, \) and \( 2.0 \) mT) taken at a given temperature is possible for \( S_{\mu} \parallel a \) at \( T = 52.59(2) \) K, the description of the seven spectra is done with \( \Delta_{\text{Lor}} = 70 \) \( \mu \)T and \( \nu_L = 0.10 \) MHz. For \( S_{\mu} \parallel a \) at \( T = 52.47(2) \) K, the two parameters are \( \Delta_{\text{Lor}} = 40 \) \( \mu \)T and \( \nu_L = 0.50 \) MHz: the zero-field spectra have therefore been recorded in the motional narrowing limit \( \nu_L/\sqrt{\gamma_{\mu}\Delta_{\text{Lor}}} > 1 \), where \( \gamma_{\mu} \) is the muon gyromagnetic ratio; \( \gamma_{\mu} = 851.6 \) Mrad s \(^{-1} \) T \(^{-1} \). This justifies the formalism used to treat \( \lambda_2(T) \) close to \( T_C \).

We now present an interpretation of our results.

We first note that the detected fluctuations cannot arise directly from the localized uranium 5f electrons since \( \nu_L \) would then be in the THz window as estimated from \( \nu_L \approx k_B T_C/h \), rather than in the MHz range as measured. We also already mentioned that the observed \( \mu \)SR signal has not the properties expected from the known macroscopic properties. These apparently conflicting results can be understood if the 5f electrons are viewed as two electron subsets. This picture has already been argued for UCu5 [21] and UPd2Al3 [22–26]. However, for UGe2 the sig-
\[ \Omega = 18 \text{ meV A}^{-2.5} \] at criticality and for small \( q_D \) (see Eq. 4.14b of Ref. [13]). Since the measured dynamics is mainly driven by the fluctuations at \( q_D \) [17], we estimate \( \nu_f \approx \Gamma(q_D) = 0.87 \text{ MHz}, \) not far from the measured value.

We now discuss the magnitude of \( \Delta_{\text{loc}} \). If the distribution of \( B_{\text{loc}} \) was Gaussian, the zero-field width of the distribution would be \( \Delta_{\text{Gauss}} = 1.7 \text{ mT} \) for muon at site 2, and \( m_U = 0.02 \mu_B \) computed using the Van Vleck-type formalism of Ref. [18]. However, the distribution is squared Lorentzian rather than Gaussian. Such a distribution is observed in systems with diluted and disordered magnetic moments. This has already been seen in UAs et al. [24] N. Metoki et al. [25], and characterized by a very slow spin dynamics. A quantitative picture for that subset is achieved by assuming

that only fluctuations at long wavelength are at play. It would be of interest to follow the small moment itinerant state as a function of pressure to determine whether the Cooper’s pairs arise from it. However, it seems difficult to perform that task with \( \mu_{\text{SR}} \), unless the spin-lattice relaxation rate increases appreciably at high pressure.

[11] In a previous short report [A. Yauanc et al., Physica (Amsterdam) 259B–261B, 126 (1999)], \( \Delta(T) \) was presented for \( s_{\parallel \perp} \). The values were slightly smaller, reflecting the uncertainty on \( a_{bg} \) which was estimated and not measured directly.
[12] When present, the anisotropy of the critical magnetic fluctuations can be observed; see, for example, P. C. M. Gubbens et al., Hyperfine Interact. 85, 245 (1994).