Evidence for Weak Itinerant Long-Range Magnetic Correlations in UGe_2

A. Yaouanc,[1] P. Dalmas de Réotier,[1] P. C. M. Gubbens,[2] C. T. Kaiser,[2] A. A. Menovsky,[3] M. Mihalik,[3] and S. P. Cottrell[4]

[1]*Commissariat à l'Energie Atomique, Département de Recherche Fondamentale sur la Matière Condensée, F-38054 Grenoble Cedex 9, France*

[2]*Interfacultair Reactor Instituut, Delft University of Technology, 2629 JB Delft, The Netherlands*

[3]*Van der Waals-Zeeman Instituut, Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands*

[4]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom*

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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.

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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 ($\sim 1.4\mu_B$ 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 the 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 substates of f electrons coexist. We indeed report the existence at ambient pressure of itinerant long-range magnetic correlations with magnetic moments of $\sim 0.02\mu_B$ 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 $T_C \simeq 52$ 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 (μSR) technique. Fully polarized muons are implanted into the studied sample. Their spin (1/2) evolves in the local magnetic field, $\mathbf{B}_{loc}$, 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 $\mathbf{B}_{loc}$ has a component perpendicular to the initial muon beam polarization, $\mathbf{S}_\mu$ (taken parallel to Z), we expect the asymmetry to display spontaneous oscillations with an amplitude maximum for $\mathbf{B}_{loc} \perp \mathbf{S}_\mu$. On the other hand, if $\mathbf{B}_{loc} \parallel \mathbf{S}_\mu$, 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 $\mathbf{B}_{loc}$.

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 μSR spectra were recorded with a longitudinal field.

We found that the temperature dependence of a_s for $\mathbf{S}_\mu \parallel \mathbf{a}$ is consistent with $\mathbf{B}_{loc} \parallel \mathbf{a}$. In agreement with that conclusion, a spontaneous muon spin precession resulting in wiggles in the asymmetry is observed for $\mathbf{S}_\mu \perp \mathbf{a}$. 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 $\mathbf{S}_\mu \parallel \mathbf{a}$ and $\mathbf{S}_\mu \perp \mathbf{a}$.

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: $aP_Z^{\exp}(t) = a_s P_Z(t) + a_{bg}$. The first component describes the μSR 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 $\mathbf{S}_\mu$ relative to **a**, $P_Z(t)$ is well described by an exponential function: $P_Z(t) = \exp(-\lambda_Z t)$, where λ_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 $\mathbf{S}_\mu \perp \mathbf{a}$ and $T < T_C$ as the constant background signal. We got $a_{bg} = 0.077$ [11]. For $\mathbf{S}_\mu \parallel \mathbf{a}$, it could be estimated only from the sample size

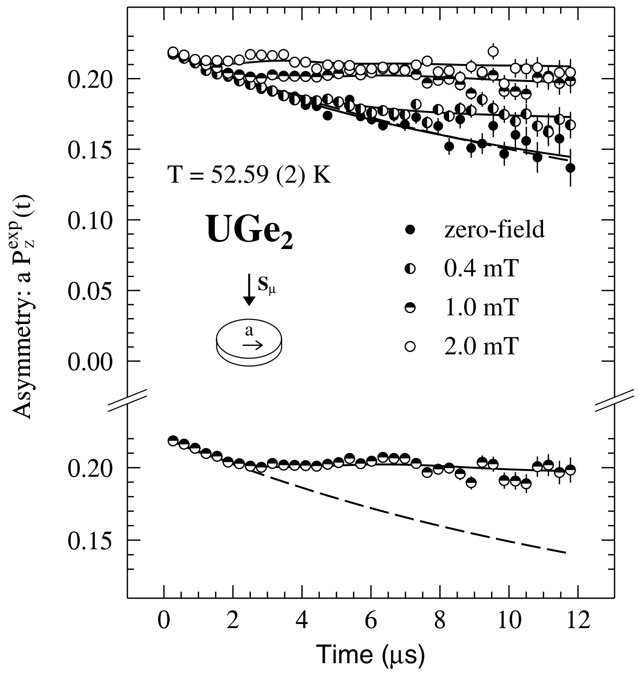

FIG. 1. Upper: examples of μSR spectra recorded in zero and longitudinal fields at $T = 52.59(2)$ K [above $T_C = 52.49(2)$ K] for $\mathbf{S}_\mu \perp \mathbf{a}$. The solid lines are fits assuming a squared-Lorentzian distribution for B_{loc}. The dashed line, which is the result of the fit of the zero-field spectrum with an exponential relaxation function, cannot be distinguished from the solid line except above $\sim$11 μs. Lower: the comparison of the 1.0 mT spectrum with the prediction of an exponential fit shows that this model is not valid in longitudinal fields. The field dependence at small field of $P_Z(t)$ proves that the field distribution at the muon site is quasistatic.

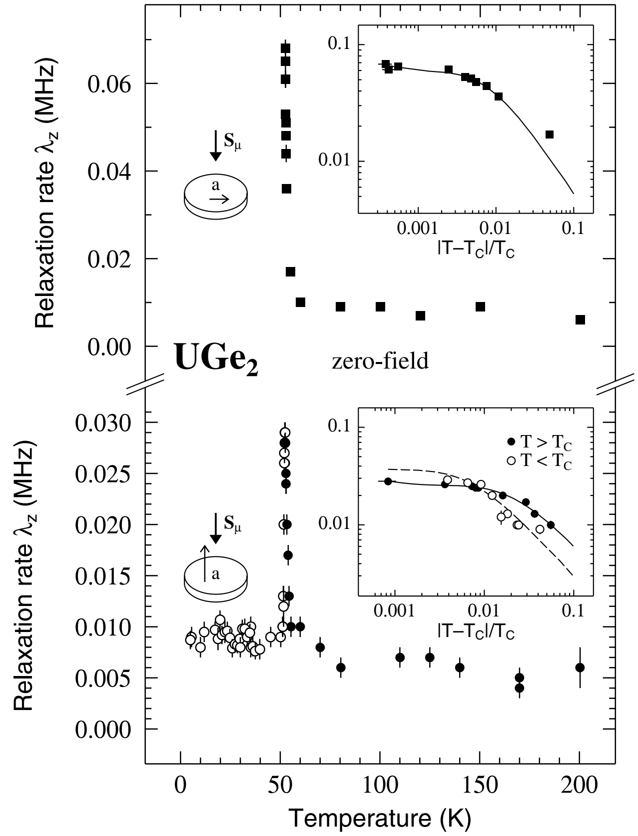

FIG. 2. Temperature dependence of λ_Z measured in zero field for $\mathbf{S}_\mu \perp \mathbf{a}$ and $\mathbf{S}_\mu \parallel \mathbf{a}$ in the upper and lower parts, respectively. The insets display $\lambda_Z(T)$ near T_C. The solid and dashed lines are the results of fits for a dipolar Heisenberg ferromagnet as explained in the main text. Since $\mathbf{B}_{loc} \parallel \mathbf{a}$, we cannot observe a spin-lattice relaxation process for $\mathbf{S}_\mu \perp \mathbf{a}$ in the ordered state. The point for $\mathbf{S}_\mu \perp \mathbf{a}$ at $(T - T_C)/T_C = 0.05$ does not fit the critical description, pointing out that it was recorded outside the critical region for that geometrical configuration.

since the relaxation was never strong enough to measure it directly. We took $a_{bg} = 0.064$. The uncertainty on this a_{bg} leads to an uncertainty on the absolute value of $\lambda_Z(\mathbf{S}_\mu \parallel \mathbf{a})$ of $\sim$10%.

In Fig. 2, we display $\lambda_Z(T)$ measured in zero field for $\mathbf{S}_\mu \perp \mathbf{a}$ and $\mathbf{S}_\mu \parallel \mathbf{a}$. For both geometries, $\lambda_Z(T)$ exhibits a maximum at T_C. It is due to the critical slowing down of the spin dynamics. Surprisingly, the anisotropy between the orientations is very weak although UGe_2 is known to be extremely anisotropic [12]. Furthermore, we show in the following lines that $\lambda_Z(T)$ near T_C is quantitatively understood in the framework of the Heisenberg model with dipolar interactions, whereas UGe_2 is considered as an Ising system. The magnetic signal that we observe has therefore a different origin from the well documented uranium magnetic state observed, e.g., by macroscopic measurements.

$\lambda_Z(T)$ was computed several years ago [13] for the critical regime of dipolar Heisenberg ferromagnets and has been successfully compared to experiments [13–15]. It is based on the derivation of the static and dynamicalscaling laws from mode coupling theory [16]. The two scaling variables at play depend on two material parameters: ξ_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 $\lambda_Z(T) = \mathcal{W}[a_L I^L(T) + a_T I^T(T)]$, where $I^{L,T}$ [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 $\lambda_Z(T)$ is shown in the insets of Fig. 2. The divergence of λ_Z at T_C is strongly reduced by the effect of the dipolar interaction [16]. The temperature scale gives the product $q_D\xi_0$ [13]. For $\mathbf{S}_\mu \perp \mathbf{a}$, we get $q_D\xi_0 = 0.021(2)$, and for $\mathbf{S}_\mu \parallel \mathbf{a}$, $q_D\xi_0^+ = 0.043(2)$ and $q_D\xi_0^- = 0.020(2)$. The index $+$ $(-)$ on ξ_0 specifies that we consider the paramagnetic (ferromagnetic) state. $\xi_0(\mathbf{S}_\mu \parallel \mathbf{a}) > \xi_0(\mathbf{S}_\mu \perp \mathbf{a})$ in the paramagnetic state, suggesting that the magnetic correlations are somewhat anisotropic. The fact that $\xi_0^+ > \xi_0^-$ is an expected feature [9]. The relaxation rate scale yields $\mathcal{W}^+ a_\mathrm{L} = 0.140(4)$ MHz and $\mathcal{W}^- a_\mathrm{L} = 0.20(2)$ MHz for $\mathbf{S}_\mu \parallel \mathbf{a}$. The transverse contribution to λ_Z for both $T < T_\mathrm{C}$ and $T > T_\mathrm{C}$ is more difficult to estimate since a_T is found much lower than a_L. Reasonable fits are obtained with $a_\mathrm{T}/a_\mathrm{L} = 0.036(14)$. We have computed a_L and a_T for different possible muon sites and found only one site satisfying $a_\mathrm{T} < a_\mathrm{L}/2$. This is site $2b$ (in Wyckoff notation) of coordinates (0, 1/2, 0) for which $a_\mathrm{L} = 1.2486$, $a_\mathrm{T} = 0.0386$. We then deduce $\mathcal{W}^+ = 0.112(3)$ MHz and $\mathcal{W}^- = 0.161(16)$ MHz. The scale deduced from the measurements with $\mathbf{S}_\mu \perp a$ 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 $\mathbf{S}_\mu$ relative to $\mathbf{a}$. 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_ext, proving that the probed magnetic fluctuations are quasistatic (fluctuation rate in the MHz range) and, since λ_Z is small, the associated magnetic moment must be small as well. Quantitatively, the field dependence of $P_Z(t)$ 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_loc is squared Lorentzian [19]. We write $P_Z(t) = P_Z(\Delta_\mathrm{Lor}, \nu_f, t)$, where Δ_Lor characterizes the width of the field distribution and ν_f its fluctuation rate [20]. A global fit of the spectra ($B_\mathrm{ext} = 0$, 0.2, 0.4, 0.6, 0.8, 1.0, and 2.0 mT) taken at a given temperature is possible. For $\mathbf{S}_\mu \perp \mathbf{a}$ at $T = 52.59(2)$ K, the description of the seven spectra is done with $\Delta_\mathrm{Lor} = 70\ \mu$T and $\nu_f = 0.10$ MHz. For $\mathbf{S}_\mu \parallel \mathbf{a}$ at $T = 52.47(2)$ K, the two parameters are $\Delta_\mathrm{Lor} = 40\ \mu$T and $\nu_f = 0.50$ MHz: the zero-field spectra have therefore been recorded in the motional narrowing limit [$\nu_f/(\gamma_\mu \Delta_\mathrm{Lor}) > 1$, where γ_μ is the muon gyromagnetic ratio; $\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}]. This justifies the formalism used to treat $\lambda_Z(T)$ 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 ν_f would then be in the THz window as estimated from $\nu_f \simeq k_\mathrm{B} T_\mathrm{C}/\hbar$, rather than in the MHz range as measured. We also already mentioned that the observed μSR 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 UCu_5 [21] and UPd_2Al_3 [22–26]. However, for UGe_2 the signatures of both subsets are found at a single temperature, the Curie temperature, whereas for UCu_5 and UPd_2Al_3 the temperatures at which the two subsets are detected are far apart. So UGe_2 presents a novel variant of the two electron subset model. Within this picture, the anisotropy of the magnetization arises from the localized $5f$ spectral density and the magnetic fluctuations probed by μSR is a signature of the bandlike electrons. We do not detect the signature of the localized $5f$ electrons, because of the strong motional narrowing of the related relaxation rate.

The effect of the dipolar interaction on the quasielastic linewidth, $\Gamma(q)$, of the fluctuations has already been observed for the weak itinerant ferromagnet Ni_3Al [27]. In particular, at criticality $\Gamma(q) \propto q^{5/2}$, as expected from scaling [16]. Thus, it is not completely surprising to detect its influence on $\lambda_Z(T)$ for the bandlike electrons of UGe_2. Quantitatively, the data have been described in the established framework of critical dynamics [16]. We shall now prove that the detected magnetization density arises entirely from long wavelength, i.e., small q, fluctuations. The magnetic properties of weak itinerant ferromagnets are explained with the latter hypothesis [28,29]. In our model, the values of $\mathcal{W}$ and ν_f and of the magnitude of the bandlike uranium magnetic moment, m_U, are controlled by two wave vectors: q_D, already introduced, and the cutoff wave vector, q_c, which sets the upper bound for the wave vector of the fluctuations involved in the buildup of the magnetization density. For simplicity, we consider that the magnetic properties of this electronic subset are isotropic. We shall detail the analysis of the data taken with $\mathbf{S}_\mu \parallel \mathbf{a}$. The same approach works equally well for the data recorded with $\mathbf{S}_\mu \perp \mathbf{a}$. As explained below, we get an overall consistent picture setting $q_\mathrm{D} = 1.0 \times 10^{-3}$ Å^{-1} and $q_c = 0.1$ Å^{-1}.

The magnetization arising from the conduction electrons can be viewed as a stochastic variable with a variance $\langle(\delta\mathcal{M})^2\rangle$. From the fluctuation-dissipation (Nyquist's) theorem, $\langle(\delta\mathcal{M})^2\rangle$ obeys the sum rule,

$$\langle(\delta\mathcal{M})^2\rangle = \frac{3 k_\mathrm{B} T}{2\pi^2 \mu_0} \int_0^{q_c} \chi(q) q^2 dq, \qquad (1)$$

if the energy of the magnetic fluctuations is smaller than the thermal energy. μ_0 is the permeability of free space. Assuming an Ornstein-Zernike form for the wave vector dependent susceptibility, $\chi(q)$, and since q_D is very small, $\langle(\delta\mathcal{M})^2\rangle \simeq 3 k_\mathrm{B} T q_\mathrm{D}^2 q_c/(2\pi^2\mu_0)$. Since $m_\mathrm{U} = v_0\sqrt{\langle(\delta\mathcal{M})^2\rangle}$, where v_0 is the volume per uranium atom ($v_0 = 61.6$ Å^3), we infer $m_\mathrm{U} = 0.02\mu_\mathrm{B}$ at T_C. Interestingly, the analysis of polarized neutron scattering data suggests for the conduction electrons a magnetic moment of $0.04(3)\mu_\mathrm{B}$ at low temperature [30].

The scale $\mathcal{W}$ for λ_Z can then be computed within the framework presented above. Numerically, from Eq. 5.10c of Ref. [13], we get $\mathcal{W} = 0.16$ MHz, close to the measured values. With the same theory, $\hbar\Gamma(q) = \Omega q^{5/2}$ with

$\Omega = 18$ meV Å$^{2.5}$ 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 $\nu_f \simeq \Gamma(q_D) = 0.87$ MHz, not far from the measured value.

We now discuss the magnitude of Δ_{Lor}. If the distribution of B_{loc} was Gaussian, the zero-field width of the distribution would be $\Delta_{Gauss} = 1.7$ mT for muon at site $2b$, 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 [19]. According to Uemura *et al.* [20], $\Delta_{Lor} = \sqrt{\pi/2}\, c\, \Delta_{Gauss}$, where c is the concentration of moments at the origin of the distribution. This relation leads to $c = 1.9\%$, consistent with the usual fact that a tiny fraction of the total number of valence electrons is able to contribute to the magnetic susceptibility.

From the q_D value, we derive the exchange interaction. We obtain $2\mathcal{J} = k_B T_C/4.2$ (see Eq. 4.4b of Ref. [13] and Ref. [16]). For comparison, the same method gives $2\mathcal{J}/k_B T_C = 1/11$ and $1/20$ for metallic Fe and Ni, respectively. Therefore the evaluation of the exchange energy is quite reasonable. From the measured product $q_D \xi_0^+$, we get $\xi_0^+ \simeq 43$ Å. This means that the correlation between the itinerant magnetic moments is relatively long range, even far outside the critical regime. Although about an order of magnitude larger than for conventional ferromagnets, ξ_0^+ compares favorably with the neutron result for Ni_3Al: $\xi_0^+ = 24(9)$ Å [27]. For the same compound, we derive from [28] that $q_c = 0.2$ Å^{-1}, a value twice as large as found for UGe_2. The moment carried by the itinerant electrons is about 4 times smaller for UGe_2 than for Ni_3Al [31]. Nearest neighbor U atoms form zigzag chains parallel to **a** [3]. This may lead to magnetic frustration and, thus, explains the disordered nature of the distribution of B_{loc}.

One may question the uniqueness of our interpretation. The observed λ_Z could arise from an impurity phase. This is unlikely since this phase would have the same critical temperature as UGe_2. It could be argued that the observed signal is the signature of a weak disorder in the uranium magnetic moments. This has already been seen in UAs [32] where the μSR signal below the Néel temperature has been attributed to a diluted source of small magnetic moments. Their quasistatic nature is related to the absence of spin excitations. However, the moments we observe in UGe_2 are quasistatic even above T_C.

In conclusion, we have shown that at ambient pressure UGe_2 is a dual system where an electronic subset of itinerant states coexist with the subset of localized $5f$ electrons responsible for the well-known bulk magnetic properties. Its associated magnetic moment is quite small 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 μSR, unless the spin-lattice relaxation rate increases appreciably at high pressure.

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