Magnetic Excitations in the Ferromagentic Superconductor UGe_2

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We report that the uniform magnetization is not conserved in the magnetic excitation spectrum of UGe_2. The measured spectrum is therefore different from that in d-electron ferromagnetic metals in a way that would facilitate magnetically mediated superconductivity.

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Theoretically unconventional spin-triplet superconductivity can arise from the exchange of longitudinal magnetic excitations, but is suppressed by the exchange of magnetic excitations transverse to the magnetic axis [1]. Ferromagnetic or nearly ferromagnetic Ising metals are then ideal candidates for the formation of such states and the first material in which superconductivity was discovered to coexist with ferromagnetism as a noncompeting order, UGe_2 [2], is indeed a strongly uniaxial ferromagnet. Our present finding that there is a strong relaxation of the uniform magnetization in UGe_2 identifies an additional feature that is probably generic to a wider class of f-electron metals, and would additionally favor the formation of magnetically mediated superconductivity.

In UGe_2 there are two different magnetic phases, FM1 and FM2, with different moments and electronic specific heat capacities [3–5]. The pressure-temperature phase diagram is schematically summarized in Fig. 1. The 1st order phase line separating the FM1 and FM2 phases is shown to end at a critical point situated at low temperature, although this is not well established. Such a behavior would be analogous with that of a phase line separating a liquid from its vapor. Within this interpretation at zero pressure there is a crossover between the FM2 and FM1 behaviors at around 30 K [6] rather than a phase transition. Our measurements that probe the magnetic excitation spectrum in the range 30–70 K at zero pressure therefore apply more ostensibly to a discussion of the FM1 phase. The discussion splits naturally into a description of the results found above and below T_C.

Above T_C the static properties are found to be consistent with the predictions of the standard spin fluctuation theory, previously also found to apply to d-metal magnets such as Ni [7] and Ni_3Al [8]. However, there is a substantial difference in the dynamic energy scale of the fluctuations compared to the d metals. Above T_C the relaxation rate for the magnetization density ($\Gamma_\mathbf{q}$) in the d metals vanishes with $q \to 0$ as $\Gamma \propto q^z$ where $z = 1$ for clean metals, $z = 2$ in the hydrodynamic limit [9], and sufficiently close to T_C the critical renormalization group analysis yields [10] $z \to 2.5$. In contrast with these results we find that for UGe_2 there is a finite relaxation rate as $q \to 0$, except exactly at T_C. This indicates that the magnetization is not a conserved quantity. Above T_C the dynamic energy scale of the fluctuations $\Gamma_\mathbf{q}$ multiplied by the static susceptibility $\chi(\mathbf{q})$, which gives a measure of the intrinsic relaxation process, is found to be independent of temperature. Below the Curie temperature, however, we find a sharp decrease in this product, although $\Gamma_\mathbf{q}$ in the limit of $q \to 0$ remains large.

The measurements were performed on the IN14 spectrometer at the Institute Laue Langevin, France. The single crystal of UGe_2 (approximately $\phi 6$ mm $\times$ 10 mm parallel c) was grown by the Czochralski technique and mounted with its b axis perpendicular to the scattering plane (UGe_2 is orthorhombic and orders with magnetic moments aligned to the crystal a axis). Energy scans were made at constant $\mathbf{q}$ with a fixed scattered energy ($k_f =$ 1.3 Å^{-1}), with collimation 40′-open-open and a cold Be filter in the incident beam. The number of scattered neutrons detected as a function of energy at a representative scattering vector normalized to a fixed number of

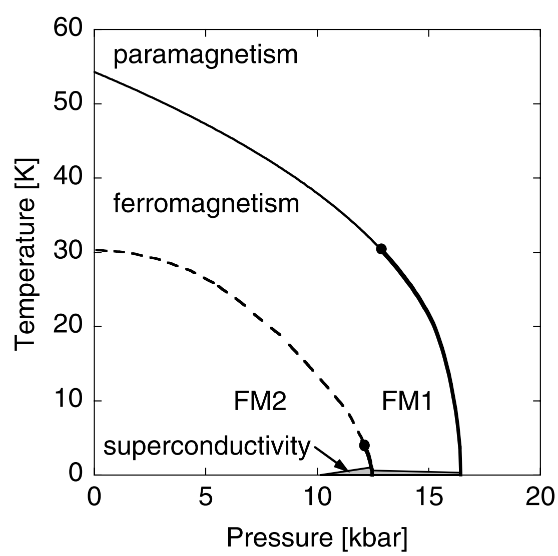

FIG. 1. The schematic phase diagram of UGe_2. Thick lines denote first order transitions and fine lines second order transitions. The dashed line is a crossover. Dots mark the positions of critical (tricritical) points. The region where superconductivity occurs is shaded.

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neutrons detected by a neutron flux monitor in the incident beam, is plotted in Fig. 2. The energy resolution has a width (FWHM) of 0.10 meV determined from the background incoherent scattering (the sharp peak at 1.5 K in Fig. 2). The absolute scattering cross section was determined by remeasuring the magnetic scattering at an additional value of $k_f = 1.97$ Å^{-1} (with no Be filter) at which the energy-integrated intensity of a longitudinal phonon at low wave-vector transfer could also be measured close to a (002) lattice point; the neutron scattering cross section for the phonon was calculated assuming the phonon wave vector to be sufficiently small for the mode to be hydrodynamic. The accuracy in determining absolute intensities by this procedure is typically of the order of 10% [11]. The magnetic scattering was in general measured near to the (001) reciprocal lattice point at which the nuclear structure factor (and phonons) make a negligible contribution due to a fortuitous cancellation of the coherent scattering from uranium and germanium nuclei. A small almost **q** independent but temperature dependent "background" signal was seen in complementary lower resolution measurements with the IN22-CRG instrument that we attribute to multiple scattering inelastic in both the phonon and magnetic systems. This contribution corresponds to less than one count in Fig. 2 and the background scattering was therefore taken to be that measured at 1.5 K at each energy and **q** for all temperatures in the analysis of the IN14 data.

For the case of ferromagnetic fluctuations, the inelastic magnetic neutron scattering cross section is related to the imaginary part of the dynamic susceptibility $\chi''_{\alpha\beta}(\mathbf{q}, \omega)$ by

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k_f}{k_i}\frac{4}{\pi}\left(\frac{m_n\mu_n}{\hbar^2}\right)^2(\delta_{\alpha,\beta} - \hat{Q}_\alpha\hat{Q}_\beta)|F_{\mathbf{Q}}|^2 \times \frac{\chi''_{\alpha\beta}(\mathbf{q}, \omega)}{1 - e^{-\beta\omega}}, \tag{1}$$

where the total wave-vector transfer is $\mathbf{Q} = \mathbf{q} + \boldsymbol{\tau}$, $\boldsymbol{\tau}$ is a reciprocal lattice vector and **q** lies in the first Brillouin zone. $\hat{Q}_\alpha$ [$\hat{Q}_\beta$] is the direction cosine of **Q** along coordinate axis α [β]. $F_{\mathbf{Q}}$ is the magnetic form factor assumed to be the same as that determined in static measurements [12], and attributed to uranium f electrons. Terms for other electronic orbits would be multiplied by much smaller values of $F_{\mathbf{Q}}$ and therefore not contribute significantly to the measured scattering. Measurements with **Q** parallel to the crystal a axis revealed no extra scattering relative to the background while for **Q** parallel to the c axis a strongly temperature dependent contribution to the scattering was found of a form characteristic of critical magnetic scattering. The dynamic susceptibility detected is thus due to excitations polarized parallel to the easy magnetic a axis at the temperatures we examined (30–70 K), i.e., $\chi_{aa} \gg \chi_{bb}, \chi_{cc}$ as expected for an Ising system. In the following we refer only to this component of the susceptibility and drop the subscripts referring to the crystal axis. As can be seen in Figs. 2 and 3, the wave vector and energy dependence of the scattering for the range of parameters studied is well described by

$$\frac{\chi''(\mathbf{q}, \omega)}{\omega} = \chi(\mathbf{q})\frac{\Gamma_{\mathbf{q}}}{\omega^2 + \Gamma_{\mathbf{q}}^2}, \tag{2}$$

$$\chi(\mathbf{q}) = \frac{\chi_0}{1 + (\xi q)^2}, \tag{3}$$

where susceptibilities are given in c.g.s. units throughout this Letter. The f-electron contribution to the static uniform susceptibility, χ_0, deduced from our measurements is shown in the inset of Fig. 3. Also shown is the total uniform static differential susceptibility measured in a magnetometer. The near coincidence of the two susceptibilities shows that the observed f-electron component accounts for the total magnetic spectral weight within the experimental accuracy of 10%.

The ordered moment, M, has previously been found to vary as $M \propto (1 - T/T_C)^\beta$ below T_C with β close to the renormalization group three-dimensional Ising (RG) value 0.33 [13] down to a temperature of the order 30 K [5,12]. In contrast immediately above T_C a Curie-Weiss law equivalent to a mean-field critical exponent describes the susceptibility (Fig. 3, inset). This suggests that there is either a large asymmetry in the temperature range to either side of T_C where non–mean-field critical scaling applies or that the scaling behavior is simply different above and below T_C. Such an asymmetry has been reported for Ni_3Al [8]. For measurements in a magnetometer below T_C the field dependence of the ordered moment

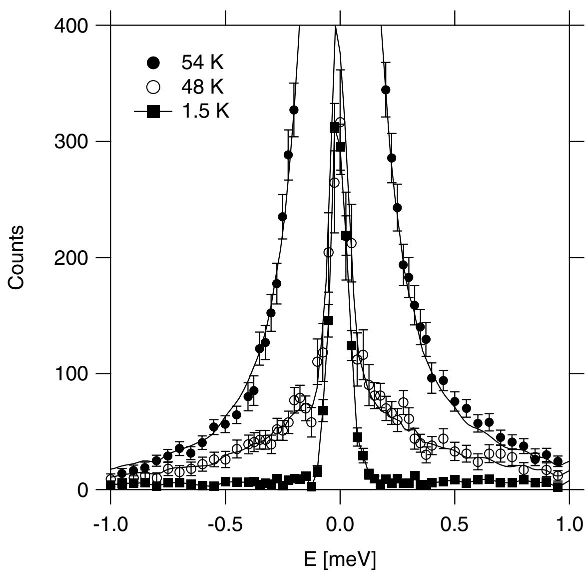

FIG. 2. The detected neutron scattering (normalized to a fixed incident beam monitor count) is shown as a function of energy transfer at $\mathbf{Q} = (0, 0, 1.04)$ reciprocal lattice units, just above T_C, at 48 K and at 1.5 K. The lines through the data points at 48 and 54 K are fits to Eqs. (1) and (2) convoluted with the instrument's resolution in energy, relative to the (background) signal at 1.5 K.

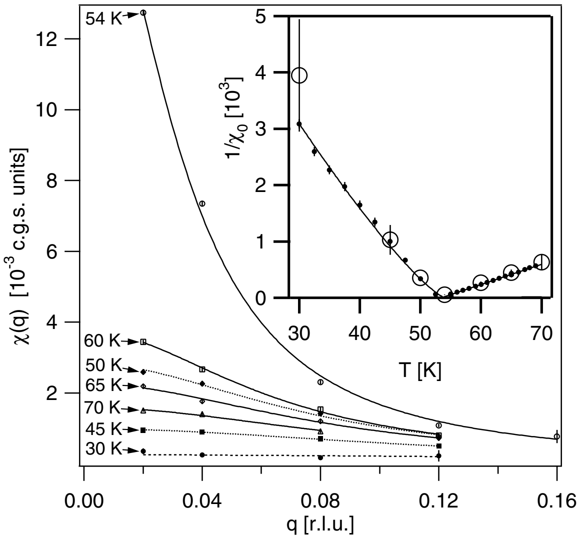

FIG. 3. The static susceptibility as a function of q parallel to the c axis (in reciprocal lattice units, 1 r.l.u. $= 1.53$ Å^{-1}) at various temperatures. The lines through the data are fits to Eq. (3). The deduced uniform static susceptibility is shown in the inset (large symbols) along with the easy axis differential susceptibility (small symbols) determined in a SQUID magnetometer. The latter is taken as M/H at $H = 0.08$ T above T_C. Below T_C, dM/dH at $H = 0$ was inferred from fitting the field dependence of the magnetization to $M \propto (H + H_0)^{1/\delta}$ in the field range 0.08–2 T. Above T_C the points lie on a straight line (Curie Weiss law), whereas below T_C one cannot distinguish between a RG critical exponent $\gamma_- = 1.24$ (line shown) and a mean-field behavior $\gamma_- = 1$ (see text).

has to be distinguished from changes in the magnetic domain structure. The low field differential susceptibility has therefore to be estimated by extrapolating data from high fields where the sample is monodomain and is therefore not determined in a model independent way. The neutron measurements are not subject to this difficulty. Whereas above T_C a mean-field exponent, $\gamma_+ = 1$, is unambiguously found with $1/\chi_0 \propto |1 - T/T_C|^{\gamma_\pm}$, below T_C, based on our data, it is not possible to distinguish directly between the RG value of $\gamma_- = 1.24$ and a mean-field value $\gamma_- = 1$ for the critical exponent. Nevertheless a simple mean-field behavior can be ruled out below T_C since the coefficient of proportionality necessary to fit the differential susceptibility with $\gamma_- = 1$ is close to 3 times the value required above T_C, whereas it should only be 2 times the value above T_C theoretically.

The product $\chi_0 \xi^{-2}$ is found experimentally to be independent of temperature both above and below T_C. Above T_C where χ_0 obeys a Curie-Weiss law we find $\xi^{-2} = \kappa_+^2 \mid 1 - T/T_C \mid$ with $\kappa_+ = 0.19$ r.l.u (0.29 Å^{-1}) (measurements for $\mathbf{q} \parallel \mathbf{a}$ show that the correlation length is isotropic to within 10% in the **a**-**c** plane). The magnitude of κ_+ is similar to that in the weak itinerant d-metal magnet MnSi, while the observed temperature independence of $\chi_0 \xi^{-2}$ does not distinguish between mean-field and RG behaviors since no temperature dependence is predicted for the former while the predicted temperature dependence for the latter is too weak to be discernible.

We now discuss the dynamics of the magnetic fluctuations. In Fig. 4(a) the $\mathbf{q}$ dependence of $\Gamma_{\mathbf{q}}$ is shown at several temperatures for $\mathbf{q} \parallel \mathbf{c}$. The main result is that $\Gamma_{\mathbf{q}}$ does not vanish as $q \rightarrow 0$ for temperatures different from T_C. The temperature dependence of $\Gamma_{\mathbf{q}}$ over the full temperature range studied is shown in Fig. 4(b) at a conveniently small value of $\mathbf{q} = (0, 0, 0.04)$ at which there is absolutely no possibility of contamination from the Bragg reflection at $q = 0$. In the standard theory for itinerant ferromagnetism [14,15], that has been applied successfully to the d metals, the relaxation of the magnetization is attributed to Landau damping. For a clean metal, not too close to T_C, the product $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ is given by the Lindhard dependence $\chi(\mathbf{q})\Gamma_{\mathbf{q}} = (2/\pi) v_f \chi_p q$, where χ_p is the noninteracting Pauli susceptibility and v_f the Fermi velocity, both determined from the band structure. The modification to consider anisotropic Fermi surfaces does not change the form of this result [14], while very close to T_C or for dirty materials a higher leading power of q is predicted for the q dependence of $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$. This product is plotted at different temperatures for $\mathbf{q} \parallel \mathbf{c}$ above T_C for UGe_2 in Fig. 4(c). As for the d metals this quantity is independent of temperature, but in contrast with the above theory $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ remains large in the limit $q \rightarrow 0$ instead of vanishing. The q dependence is almost flat with $\chi(\mathbf{q})\Gamma_{\mathbf{q}} = 0.70(4)$ μeV as $q \rightarrow 0$ [and a slope of 0.5(5) μeV Å estimated at 54 K]. For comparison in Ni_3Al [16] and $ZrZn_2$ [17] $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ depends linearly on q with a coefficient of proportionality in the range 2–3 μeV Å.

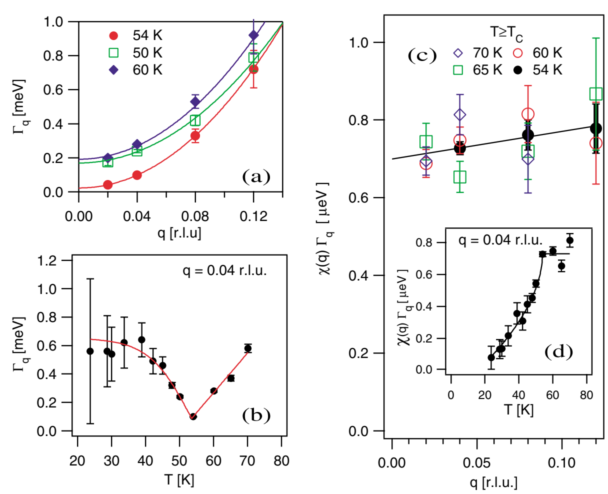

FIG. 4 (color online). Panel (a) shows the q dependence ($\mathbf{q} \parallel \mathbf{c}$) of $\Gamma_{\mathbf{q}}$ at three different temperatures. Panel (b) shows the temperature dependence of $\Gamma_{\mathbf{q}}$ at $\mathbf{q} = (0, 0, 0.04)$. Panel (c) shows the q dependence of the product $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ at different temperatures above T_C. Panel (d) shows the temperatures dependence of $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ at $\mathbf{q} = (0, 0, 0.04)$ above and below T_C. The lines in the various figures serve to guide the eye.

The large finite value of $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ as $q \to 0$ implies that a spatially uniform fluctuation of the magnetization relaxes rapidly and that the average magnetization density can no longer be considered to be a conserved quantity. The dipole interaction [18] between moments is one example of a non–spin-conserving interaction that could give such damping, albeit very weak. Although long range forces such as dipole interactions can also lead to a recovery of mean-field–like exponents very close to T_C [19] the general weak nature of dipole forces means that dipole interactions are unlikely to explain the observed damping. Instead it is more likely that significant non–spin-conserving terms arise from the strong spin-orbit interactions associated with the f electrons [20] while the mean-field behavior observed above T_C simply indicates that the temperature is outside the critical scaling region.

A weaker homogeneous relaxation of a similar form has previously been found in the anisotropic itinerant ferromagnet MnP [21], although it was not studied below T_C. In general for the d-electron metals longitudinal excitations are difficult to study below T_C in the ferromagnetic state where they must be distinguished from dominant transverse excitations (spin waves). This difficulty is not present in a strong Ising magnet, such as UGe_2, where there are no low energy spin waves. The temperature dependence of $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ over the full temperature range studied is shown in Fig. 4(d) at $\mathbf{q} = (0, 0, 0.04)$. The observation that $\Gamma_{\mathbf{q}}\chi(\mathbf{q})$ decreases below T_C is contrary to the behavior expected for dynamical critical scaling for an Ising magnet with a nonconserved order parameter; the latter theory predicts that $\chi(\mathbf{q})\Gamma_{\mathbf{q}}$ should be almost independent of temperature both above and below T_C [10]. The unusual behavior below T_C might, however, relate in a simple way to the spin splitting of the Fermi surfaces in the magnetic ground state. Such a splitting between the pseudospin majority and minority Fermi surfaces removes the degeneracy between the opposite spin polarities. The phase space for transitions from one spin surface to the other that respect energy and momentum conservation is then reduced, potentially partially suppressing the relaxation mechanism.

We now discuss the consequences of the damping we have observed. First, we note that the large value of $\Gamma_{\mathbf{q}}$ at small q clarifies why muon spin relaxation failed to detect critical fluctuations corresponding to the full Curie-Weiss moments [22], although the origin of the damping that was seen in the muon experiments and attributed to small moments of 0.02 μ_B is not explained.

Finally, we discuss the impact of our finding for superconductivity. We have attributed the damping of the uniform magnetization to spin-orbit interactions related to the presence of f electrons. Our measurements show that this damping remains strong at low temperatures at least 25 K below T_C at zero pressure. Superconductivity occurs only under pressure where T_C is reduced to of the order of 30 K. The low temperature anisotropy of the differential magnetic susceptibility is, however, much higher at these pressures than at zero pressure [23], indicating that spin-orbit interactions are likely to be more important. It is therefore likely that the relaxation process we observe at zero pressure would be strong at the high pressures and low temperatures necessary for superconductivity. Since the relaxation rate, $\Gamma_{\mathbf{q}}$, determines the energy scale for magnetically mediated pairing any increase in the damping of the magnetic fluctuations promotes magnetically mediated superconductivity. This is theoretically supported by numerical calculations [24] that show that uniform damping, such as we have observed, can significantly enhance the superconducting critical temperature above the value calculated in the presence of Landau damping alone.

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