Field-Dependent Energy Scales in URu_2Si_2

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At low temperature, macroscopic properties of URu_2Si_2 display a characteristic energy scale $\Delta_0(B)$ which decreases when a magnetic field is applied, and eventually vanishes at an extrapolated value of the field of about 40 T. We have performed inelastic neutron scattering measurements of the magnetic dynamics of URu_2Si_2 in applied fields along the c axis of intensities up to 12 T. We show that $\Delta_0(B)$ is not related to gaps in the magnetic fluctuations spectra. This provides direct evidence of the fact that two distinct energy scales govern the physics of this compound.

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The unique properties of URu_2Si_2 have attracted so much attention in the past that it has now become one of the most experimentally characterized actinide compounds. Although it is often classified as a heavy fermion system, there is at present no evidence, neither direct (i.e., from de Haas–van Alphen or positron annihilation studies of the Fermi surface) nor indirect, that (heavy) f-type quasiparticle bands actually exist. The moderate enhancement of the linear term in the low-T specific heat can indeed be interpreted as due to the scattering of conduction electrons by the measured fluctuations of the $5f$ magnetic moments [1]. Inelastic neutron scattering (INS) experiments [2,3] show that the spectrum of these fluctuations is gapped, with no trace of the overdamped, quasielastic response typical of heavy fermion compounds. A single, dispersive excitation frequency, sharply defined over most of the Brillouin zone, is observed. The characteristics of this mode are those expected for a transition between two crystal field (CEF) singlets [3]. In addition to the magnetic dynamics, many other properties of URu_2Si_2 are difficult to frame into the phenomenology of heavy fermion compounds, while they are strongly reminiscent of that of localized f-electrons compounds with a singlet CEF ground state, such as, for example, $PrNi_5$. Indeed, a semiquantitative interpretation of a great part of the phenomenology of URu_2Si_2 may be given by a model of localized f electrons in a CEF [4,5]. It is clear that the CEF framework is to be seen only as an approximation for the complex physics of this compound. This framework may remain approximately valid, even in the presence of coupling to conduction electrons, when the ground state of the CEF Hamiltonian is a singlet.

At $T_N = 17.5$ K URu_2Si_2 undergoes a phase transition, marked by sizable anomalies in macroscopic observables. The strong increase of T_N with applied pressure suggests again the physics to be governed by local degrees of freedom of the U ions rather than itinerant electrons. The transition has been attributed to a type-I antiferromagnetic ordering along the c axis, with an ordered moment $\mu \sim 0.03\mu_B$ at saturation [3,6]. However, the correlation length is finite and sample dependent [7]. No way has been found so far to reconcile the tiny value of the moment with the large macroscopic anomalies observed at T_N. For example, the anomaly in the specific heat coefficient is comparable to that of UPd_2Al_3 whose ordered moment, however, is 30 times larger. On the other hand, in UPt_3 a magnetic state extremely similar to that of URu_2Si_2 exists, but no sign of this state is seen in macroscopic measurements. Moreover, the anomalies observed at T_N in macroscopic observables are not dependent on the sample quality, while the temperature variation of the ordered magnetic moment μ changes from sample to sample [7]. Also, when a magnetic field is applied, T_N and μ appear to vanish at different values of the field [8]. The most obvious way out of these inconsistencies is to assume that macroscopic anomalies are not associated with μ, but rather with a hidden order parameter (OP) not yet observed directly in scattering experiments. Various ideas have been put forward [9–11]. Within the model of Ref. [4], the characteristics of the anomalies observed at T_N, as well as their changes in an applied magnetic field [5], are consistent with staggered ordering of one of the electric quadrupoles Q_{xy} or $Q_{x^2-y^2}$. The c_{66} and c_{11}-c_{12} elastic constants, which *selectively* sense the uniform quadrupolar susceptibility conjugated to these two quadrupoles, display temperature [12] and applied-magnetic-field [13] dependencies consistent with the model. However, the question of which theoretical framework is the most appropriate is still open [14].

While static properties in zero and applied field, and the magnetic dynamics in zero field are now known in great detail, a fundamental piece of information is still missing, i.e., how the field modifies the dynamics. This aspect is very important because various static quantities display the existence of a characteristic energy scale $\Delta_0(B)$ *decreasing* with a field applied along the c axis, and eventually

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vanishing at an extrapolated value of the field of about 40 T. For example, magnetoresistivity [15], thermal expansion [16], and specific heat [17] measurements show that by increasing B from 0 to 12 T this energy decreases by 10%. INS spectra in zero field show a single ($\mathbf{q}$-dependent) energy scale $E_{\mathbf{q}}$. If the compound were governed uniquely by such an energy scale, $E_{\mathbf{q}}$ (or, at least, the associated density of states) should then move downwards as B increases.

In this Letter, we present measurements of the magnetic dynamics in applied fields along the c axis of intensities up to 12 T. The results show that macroscopic gap $\Delta_0(B)$ cannot be identified with $E_{\mathbf{q}}$, and this suggests the existence of two separate energy scales in the system. Experiments were performed on a well-characterized single crystal of URu_2Si_2, grown in a tri-arc furnace with the Czochralski method and annealed for 8 days at 1223 K [17]. The sample, with a mass of 4.5 g, was mounted with the c axis of the tetragonal crystal structure along the vertical direction, parallel to the magnetic field $\mathbf{B}$ of a 12 T cryomagnet. Neutron inelastic scattering measurements were made on the IN14 (cold) and IN8 (thermal) triple-axis spectrometers at the Institut Laue Langevin, Grenoble. On IN14, graphite (002) was used for the vertically focusing monochromator and horizontally focusing analyzer, at fixed $k_f = 1.295$ Å^{-1}, and a Be filter was used to remove high-order contamination. The energy resolution was 0.13 meV full width at half maximum (FWHM). The maximum energy transfer of IN14 equipped with the cryomagnet was 4.7 meV. Measurements at higher energy transfers were done on IN8, using a Cu(111) monochromator and a horizontally focusing graphite analyzer with $k_f = 2.662$ Å^{-1}, giving an energy resolution of about 1.6 meV FWHM. On both instruments, data were collected at 5 K for different values of B, between 0 and 12 T.

The results of constant-$\mathbf{Q}$ scans taken at 5 K with IN14 at the antiferromagnetic zone center, $\mathbf{Q} = (1, 0, 0)$, and at $\mathbf{Q} = (1.4, 0, 0)$, are shown in Fig. 1. The asymmetric shape of the peak at the magnetic zone center is a resolution effect that arises from the strong curvature of the dispersion of the magnetic excitations. In zero field, at $\mathbf{Q} = (1, 0, 0)$ the magnetic excitation has minimum energy and maximum intensity. Its behavior in an applied field is opposite to that expected from macroscopic behavior, since an energy *increase* is observed, up to about 30% for $B = 12$ T. The field dependence of the energy gap and of the magnetic Bragg peak intensity at $\mathbf{Q} = (1, 0, 0)$ is shown in Fig. 2. By assuming a quadratic field dependence of the magnetic Bragg peak intensity (shown as a solid line in Fig. 2), extrapolation gives a critical field for the vanishing of μ of 15.0 ± 0.4 T, close to that estimated in Ref. [8]. The magnetic correlation length is found to be essentially field independent, and of the order of 200–300 Å. By moving along the $(1 + \zeta, 0, 0)$ direction $E_{\mathbf{q}}(B = 0)$ increases, but the shift $D_{\mathbf{q}} = [E_{\mathbf{q}}(B = 12\text{ T}) - E_{\mathbf{q}}(B = 0)]/E_{\mathbf{q}}(B = 0)$ decreases. At the edge of the accessible energy window, e.g., at $\mathbf{q} = (1.4, 0, 0)$, it is slightly but unambiguously negative, $D_{\mathbf{q}} \sim (-2.2 \pm 1.4)\%$ (see Fig. 1).

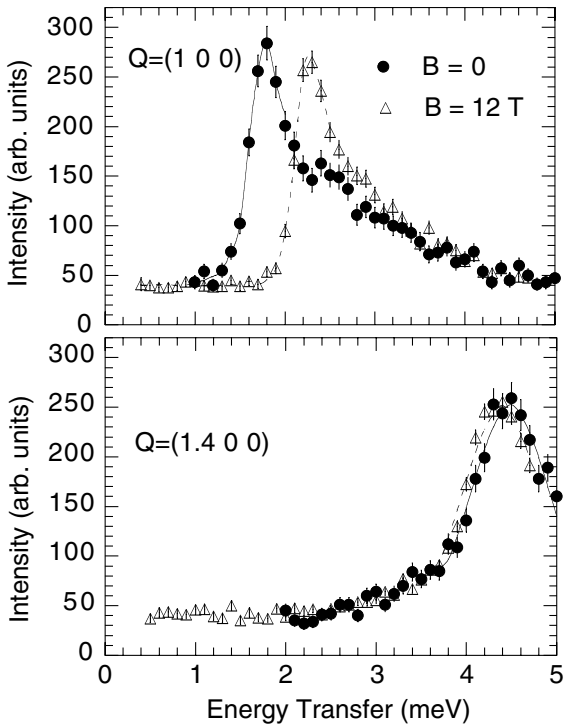

FIG. 1. Constant-$\mathbf{Q}$ spectra at $T = 5$ K on IN14 with $B = 0$ (dots) and with a field $B = 12$ T applied along the c axis (triangles). Lines are guides to the eye.

Excitations at higher energies, measured on IN8, do not show any appreciable field-induced energy shift, $|D_{\mathbf{q}}| \lesssim 3\%$. The results for $\mathbf{q} = (1.5, 0, 0)$ are shown in Fig. 3 as an example. For this wave vector $E_{\mathbf{q}}$ lies on the peak of the magnetic density of states [2,3]. If the macroscopic Δ_0 were associated with magnetic excitations, we should have observed a downwards shift $D_{\mathbf{q}} \sim -10\%$ in this energy

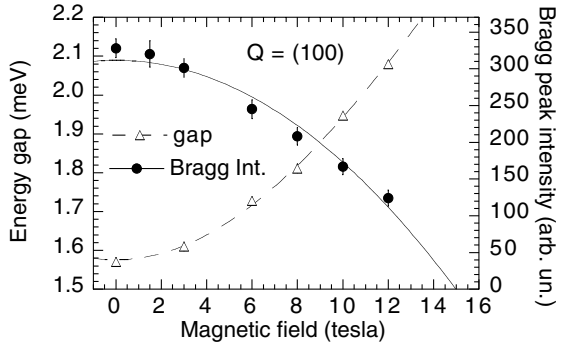

FIG. 2. Field dependence of the integrated magnetic Bragg peak intensity (circles) and of the energy gap (triangles) at the magnetic zone center. The solid line is a quadratic fit, while the dashed line is a fit of $E(B) = [E^2(0) + AB^2]^{1/2}$ to the gap energy.

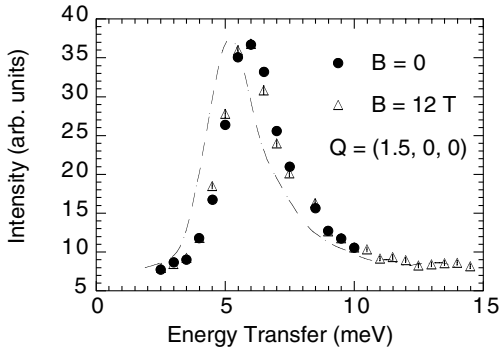

FIG. 3. Constant-**Q** spectra at $T = 5$ K on IN8 with $B = 0$ (dots) and with a field $B = 12$ T applied along the c axis (triangles). The broken line represents the expected position of the excitation at 12 T if the magnetic gap could be identified with the macroscopic $\Delta_0(B)$.

window [18]. However, we do not observe such strong negative shifts at any energy, which indicates that Δ_0 is not related to $E_{\mathbf{q}}$. This conclusion is strengthened by the fact that, besides being inconsistent with static properties, the hypothesis of a single energy scale appears to be in *qualitative* disagreement with observed effects of the field on the dynamics. To illustrate this, let us assume that there are only two states (say, $|0\rangle$ and $|1\rangle$) at low energy (these can be either two singlets or a doublet split by the OP

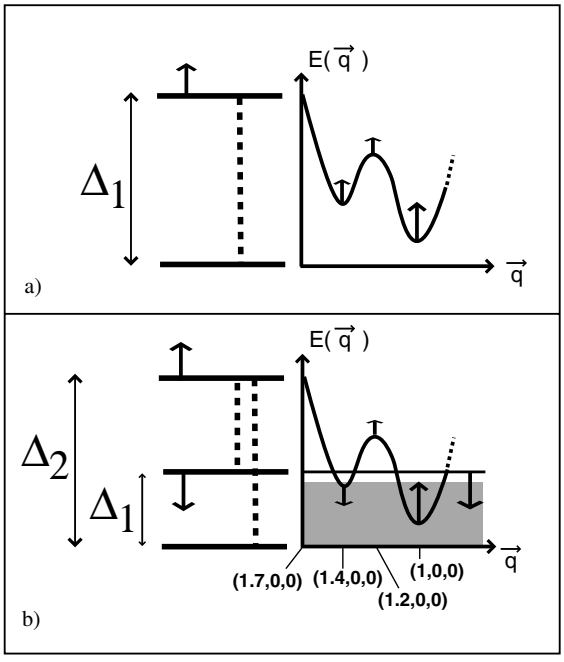

FIG. 4. (a) Situation with two low-lying CEF states. The right-hand side shows schematically how the excitation disperses along the $(1 + \zeta, 0, 0)$ direction [3]. Arrows indicate the direction of the shift expected for a magnetic field applied along z. (b) Situation for the model of Ref. [4]. The shaded area corresponds to the energy window which was analyzed in the IN14 experiment. Dashed vertical lines in a and b represent nonzero J_z matrix elements when $B = 0$.

molecular field). To interpret INS in zero field, one must assume [3] a matrix element $\alpha = \langle 0|g\mu_B J_z|1\rangle \sim 1.2\mu_B$, and a splitting $\Delta_1 = E_1 - E_0 \sim 10$ meV, which is the single energy scale of the system (see Fig. 4a). Although the INS results in zero field can be fitted to this type of models [3], there are two problems: one is the inconsistency with static properties for $B \neq 0$, discussed above, the second is that it is difficult to explain why the energy shifts produced by the field at $\mathbf{q} = (1, 0, 0)$ and $\mathbf{q} = (1.4, 0, 0)$ are of opposite sign. The random phase approximation, which works well at $B = 0$ [3], gives for the dispersion of the magnetic excitation

$$E_{\mathbf{q}}(B) = \Delta_1(B)[1 - 4\alpha^2(B)J(\mathbf{q})/\Delta_1(B)]^{1/2}, \quad (1)$$

with $\alpha(B)$ and $\Delta_1(B)$ the field-dependent matrix element and gap, and $J(\mathbf{q})$ the Fourier transform of the RKKY magnetic couplings, which can be determined by fitting Eq. (1) to the measured dispersion $E_{\mathbf{q}}(B = 0)$ [3]. Although the precise way α and Δ_1 vary with B is model dependent, Eq. (1) implies that the sign of the energy shift $D_{\mathbf{q}}$ produced by the field can be positive at $(1, 0, 0)$ and negative at $(1.4, 0, 0)$ only if the signs of $J(1, 0, 0)$ and $J(1.4, 0, 0)$ are opposite [19]. However, there is no question that these signs are the same. In fact, the observed dispersion $E_{\mathbf{q}}(B = 0)$ implies a positive $J(\mathbf{q})$ for all values of $\mathbf{q}$ such that $E_{\mathbf{q}}(B = 0) \lesssim 10$ meV [3], i.e., including $(1.4, 0, 0)$ with ample margin. Assuming a single energy scale leads therefore to qualitative disagreement with our results, since the signs of $D_{\mathbf{q}}$ at $(1, 0, 0)$ and $(1.4, 0, 0)$ should be the same, while the measured signs are opposite.

We do not see any good candidate mechanism to explain this inconsistency in a two-level framework. For example, a variation of $J(\mathbf{q})$ with the field seems very unlikely. The fact that at $B = 12$ T the staggered moment is not zero (see Fig. 2) implies that the microscopic field at the U sites is actually the sum of B and of the staggered molecular field B_m due to the moment (i.e., the U ions are in a weakly *ferrimagnetic* state). However, the resulting corrections to Eq. (1) are very small, since the value of B_m corresponding to the tiny moment is of the order of less than 1 T. Being much smaller than B, B_m is not expected to modify the energy shifts qualitatively.

In the following, we propose a qualitative interpretation of our results in terms of the model of Refs. [4,5]. In this model (see Fig. 4b), there are three CEF singlets ($|0\rangle$, $|1\rangle$, and $|2\rangle$) at low energy with gaps Δ_1 and Δ_2. Below T_N and at $B = 0$, $\alpha_{01} = \langle 0|g\mu_B J_z|1\rangle = 0$, and $\alpha_{02} = \langle 0|g\mu_B J_z|2\rangle \neq 0$; thus magnetic excitations $E_{\mathbf{q}}$ are associated with the gap to the second excited state Δ_2 (magnetic energy scale), while excitations to the first excited state correspond to a quadrupolar branch (drawn flat in Fig. 4b), not visible in the neutron spectra (nonmagnetic energy scale). The macroscopic Δ_0 is identified with Δ_1. Indeed, when $B \neq 0$, Δ_1 decreases in much the same way as observed for Δ_0 (e.g., the decrease is about 10% at

$B = 12$ T). In addition, Δ_2 increases with B, and a small nonzero matrix element α_{01} is induced by the combined effect of B and of the quadrupolar OP [5].

When $B \neq 0$, magnetic excitations whose energy $E_{\mathbf{q}}$ is far from Δ_1 are unaffected by the presence of the nonmagnetic branch, and move upwards with the field just as in the model of Fig. 4a. This is the case, for example, at $(1, 0, 0)$, where the shift with B is by far the greatest. However, because of the small α_{01} matrix element, when $B \neq 0$ the magnetic and quadrupolar branches weakly anticross, rather than cross. So, as the quadrupolar branch moves down with B, it repels and pushes downwards magnetic excitations whose energy $E_{\mathbf{q}}$ is only slightly smaller than Δ_1, thus making a change of sign of $D_{\mathbf{q}}$ possible. This is just what is expected to occur at $(1.4, 0, 0)$.

While this qualitative picture for the change of sign is easily understood, quantitative calculations of the shifts $D_{\mathbf{q}}(B)$ are not very meaningful, since these appear to depend critically on details of the model. $D_{\mathbf{q}}(B)$ can be calculated by the generalization of Eq. (1) to the case of three levels and by including all relevant ion-ion couplings. In particular, one has to specify the Fourier transform of magnetic and quadrupole couplings. The former is known, but the latter is not. Even if quadrupole couplings are neglected, $D_{\mathbf{q}}(B)$ appears to be very sensitive to the precise functional forms of $\Delta_1(B)$, $\Delta_2(B)$, $\alpha_{02}(B)$, and, particularly, $\alpha_{01}(B)$. Any unavoidable uncertainties or inaccuracies in these individual quantities combine nonlinearly to give strong variations of $D_{\mathbf{q}}(B)$ in the anticrossing region. We know that the value of the matrix element $\alpha_{02}(B)$ is not reproduced precisely by the model, since it is underestimated by about 25% already at $B = 0$ [4] [the measured value is $\alpha_{02}(B = 0) \sim 1.2\mu_B$ [3]]. Tentative fittings of $D_{\mathbf{q}}(B)$ indicate an inaccuracy also in $\alpha_{01}(B = 12$ T), which appears, instead, to be overestimated. It is calculated in the range $0.2\mu_B$–$0.3\mu_B$, while the observed shifts $D_{\mathbf{q}}(B)$ are rather consistent with $\alpha_{01}(B = 12 \text{ T}) \lesssim 0.1\mu_B$. The latter value of α_{01} is large enough to be sensed indirectly through the change of sign in $D_{\mathbf{q}}(B)$, but it is too weak for the quadrupolar branch to be directly observed in our INS spectra as a second peak.

In conclusion, we have performed inelastic neutron scattering measurements of the magnetic dynamics of URu_2Si_2 in applied fields of intensities up to 12 T. Macroscopic measurements display the existence of an energy scale $\Delta_0(B)$ decreasing by 10% at 12 T. This energy scale is usually assumed to be associated with the spectrum of magnetic fluctuations. We have shown that this assumption is incorrect: we do not find evidence of such scale in the magnetic excitations, which indicates that $\Delta_0(B)$ is nonmagnetic. This is also supported by the peculiar behavior of the magnetic excitations with an applied field.

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[18] Note that within the RPA, excitations with different values of $\mathbf{q}$ but the same $E_{\mathbf{q}}$ behave in the same way when the field is applied. Thus, the behavior at $\mathbf{q} = (1.5, 0, 0)$ is representative of that of all modes with $E_{\mathbf{q}}(B = 0) \sim 6$ meV, i.e., of the peak of the magnetic density of states.
[19] We assume $\Delta_1(B)$ to increase with B, which is by far the most realistic situation. Anyway, we have checked that even in the opposite hypothesis it would not be possible to account for the observed behavior of $D_{\mathbf{q}}(B)$.