Observation of low-energy magnetic response in the heavy-fermion compound UBe_{13}

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Inelastic-neutron-scattering experiments have been used to examine the low-frequency (<2 meV) response of polycrystalline samples of the heavy-fermion UBe_{13}. A quasielastic response with a characteristic linewidth of 1.5 meV for $T>1$ K is found. Surprisingly we found no anomalous dependence of this response on q, the reduced reciprocal lattice vector. The strength of the response corresponds to $\sim 60\%$ of the bulk susceptibility, and it scales with the latter for $1.5<T<30$ K. Taking into account earlier experiments the total response function for UBe_{13} thus contains two components, one with a linewidth of 1.5 meV and the other [A. I. Goldman *et al.*, Phys. Rev. B **33**, 1627 (1986)] with a linewidth of ~ 13 meV. The weight is approximately evenly divided between the two. UBe_{13} is therefore similar to UPt_3 in that the magnetic response functions exist on two quite different energy scales. This appears different from Ce-based heavy fermions, and is not yet understood.

I. INTRODUCTION

The study of heavy fermions (HF's) is passing into its second decade, and they are far from understood. A fundamental problem is the interplay between magnetic correlations and superconductivity. The most important characteristic of HF systems is their high value of γ, the coefficient of their electronic specific heat at low temperature. For uranium compounds, values of γ range from ~ 200 mJ/(mol K^2) for URu_2Si_2 to 1100 mJ/(mol K^2) for UBe_{13}.[1] At the same time, the magnetic susceptibility is also large at low temperature. The combination of these two experimental observations leads to the obvious suggestion that both are caused by magnetic fluctuations.[1] If the spectrum of these fluctuations is purely relaxational in form, then the frequency dependence ($\hbar=1$ throughout this paper) of the magnetic response may be related to the dynamical susceptibility by

$$\mathrm{Im}X(\mathbf{Q},\omega)=[\mathrm{Re}X(\mathbf{Q})]P(\mathbf{Q},\omega)\omega\Pi \ , \tag{1}$$

where

$$\mathrm{Re}X(\mathbf{Q})=f^2(\mathbf{Q})\mathrm{Re}X_{\mathrm{loc}}(\mathbf{Q}) \ . \tag{2}$$

$f(\mathbf{Q})$ is the magnetic form factor, which arises from the spatial extent of the unpaired electrons, and $X_{\mathrm{loc}}(\mathbf{Q})$ allows for variations in the local susceptibility as a function of wave vector (or momentum transfer $\mathbf{Q}$). Such variations could occur because of correlation effects. In the limit of $Q\rightarrow 0$,

$$\mathrm{Re}X_{\mathrm{loc}}(\mathbf{Q})\rightarrow \mathrm{Re}X_{\mathrm{loc}}(0) \ , \tag{3}$$

which is the bulk susceptibility.

Returning to Eq. (1), the spectral weight function is

$$P(\mathbf{Q},\omega)=\frac{I}{\Pi}\frac{\Gamma(\mathbf{Q})}{\Gamma^2(\mathbf{Q})+\omega^2} \ , \tag{4}$$

where $\Gamma(\mathbf{Q})$ defines the half width of the quasielastic Lorentzian centered about zero frequency. $P(\mathbf{Q},\omega)$ is a normalized function when integrated over energy. If, furthermore, $\Gamma(\mathbf{Q})$ is independent of $\mathbf{Q}$, we may readily derive that there is a direct inverse proportionality between Γ and γ. We see this clearly in the Ce HF systems[2] as shown in Table I, where we give the product $\gamma\Gamma$. Although important $\mathbf{Q}$-dependent effects are seen,[2] the bulk of the low-temperature susceptibility $\mathrm{Re}X(0)$ clearly corresponds to fluctuations that are responsible for the large value of γ.

The situation is much more complex for uranium-based HF systems.[3] In UPt_3, for example, *two* energy scales have been found, each with its own $\mathbf{Q}$ dependence. The low-energy part "drives" the short-range magnetic ordering,[4] so that we would not expect the simple relationship between Γ and γ to hold. UBe_{13}, on the other hand, does *not* show magnetic order[5] and yet has the largest γ of any uranium-based HF material. Our expectation is then that a narrow line ($\Gamma\leq 1$ meV) should be seen in the neutron scattering corresponding to these long-lived magnetic fluctuations. In the early work on polycrystalline material, Goldman *et al.*[6] reported a very

TABLE I. Values at low temperature of γ, the electronic specific heat, and Γ (the quasielastic Lorentzian half width) of various heavy fermions. The γ values are taken from Ref. 1; the Γ values are from the references quoted.

	γ [J/mol K^2)]	Γ (meV)	Product $\gamma\Gamma$	Ref.
$CeRu_2Si_2$	0.35	2.0	0.70	2
$CeCu_6$	1.60	0.42	0.67	2
UPt_3	0.45	~ 10	4.5	3
		~ 0.20	0.1	4
UBe_{13}	1.10	13	14	6
		~ 1.5	1.7	this work

wide magnetic response ($\Gamma \sim 13$ meV). They also looked for, and failed to find, any low-energy response. Subsequent single-crystal efforts have been hampered by the weakness of the signal, as well as by the uncertainty of which Q value to select. The object of our present experiments has therefore been *to search for the low-energy excitations* in polycrystalline UBe_{13} with an appropriate neutron spectrometer and *determine the Q and energy dependence* of such a response. (An earlier study by Neumann *et al.*[7] had insufficient energy resolution, in addition to poor statistics, to answer these questions.)

II. EXPERIMENTAL DETAILS

For these experiments we have used polycrystalline material together with a time-of-flight spectrometer (IN6) at the Institut Laue-Langevin, Grenoble. With its large detector coverage and intense beam of cold neutrons, this configuration maximizes the possibility of observing a magnetic signal. In addition, since UBe_{13} is *cubic* ($a_0 = 10.25$ Å at 5 K), magnetic interactions are expected to be isotropic and so should be a function of the magnitude of Q rather than having any directional dependence. Thus the region $0 < Q < 1$ Å^{-1}, where Q is the total-momentum transfer, represents an understandable average over the Brillouin zone; this is different in the case of hexagonal UPt_3 or the tetragonal URu_2Si_2. (Despite these caveats about noncubic systems, Holland-Moritz *et al.*[8] did extract some Q-dependent information from experiments on polycrystalline URu_2Si_2, which was later confirmed and extended by single-crystal measurements by Broholm *et al.*[9] Nevertheless, we are on firmer ground with cubic URe_{13}.)

The sample was the *same* polycrystalline sample (powdered material sieved through 420-μm mesh) as used by Goldman *et al.*[6] in the Brookhaven experiments. A special holder $51 \times 33 \times 5$ mm^3 was constructed from 7049A-T6 aluminum alloy. The sample (20.2 g) was loaded in a helium-filled container. Calibrated resistors rested within the powder material. Cadmium sheets were installed in the cell in the plane perpendicular to the neutron beam and parallel to the plane of scattering to reduce multiple-scattering effects. The sample was placed in a ^{3}He cryostat in transmission geometry with the thin direction 45° to the neutron beam. Experiments were also performed on polycrystalline (powder) $ThBe_{13}$ (16.9 g) in the same cell. Runs with the empty cell and vanadium (11.6 g) in the same cell allowed the scattering to be placed on an absolute scale. Experiments were performed at 0.6, 4.0, and 10 K in the ^{3}He cryostat. In a later experiment, in a conventional ILL "orange" ^{4}He cryostat, experiments were performed at 1.5, 4.0, 9.7, and 30 K. The powder nature of our samples introduces some uncertainty as to whether or not the material was actually at ~0.6 K. Although He gas was in the sample cell, the thermal conductivity of superconducting powder material is known to be very poor. A better choice would be solid pieces as used in the muon experiments.

The IN6 time-of-flight spectrometer consists of a neutron guide from the ILL cold source, a series of focusing pyrolitic graphite monochromators, and a Fermi chopper producing a pulsed monochromatic beam at the sample position. There are then ~300 detectors subtending a large solid angle after the sample. Scattered neutrons are characterized by the detector into which they are scattered and their time of arrival. From these [ϕ (angle) and t (time)] coordinates, a spectra in (Q,ω) may be readily constructed. Detectors were grouped to give a Q resolution of ~10%. Those detectors at Q positions corresponding to Bragg peaks were not used in subsequent analysis. For all the experiments reported in this study, we have used an incident neutron energy of 3.12 meV (5.12 Å), giving a resolution (Gaussian) at the elastic position of ~50 μeV [half width half maximum (HWHM)].

Typical spectra are shown in Fig. 1. Since these spectra are taken at low temperature, there is no appreciable

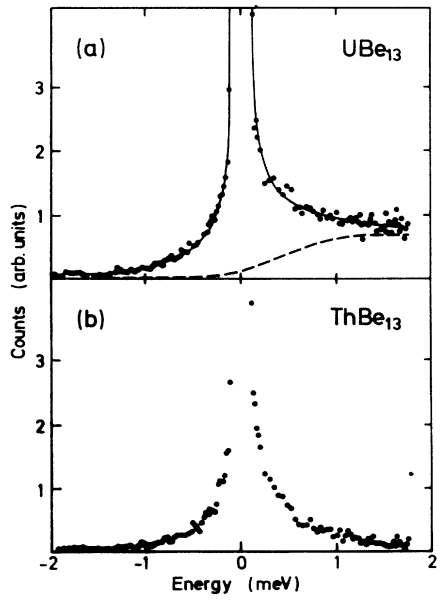

FIG. 1. Representative $S(Q,\omega)$ spectra for (a) UBe_{13} and (b) $ThBe_{13}$. Both are at $T = 1.5$ K and correspond to $Q = 0.95$ Å^{-1}. To make the comparison, the spectra have been normalized to the same peak values, which is 60 times the scale shown. A clear difference may be seen between the spectra, indicating a magnetic response function present at low-energy transfers in UBe_{13}. The solid line is a fit to the total spectrum, whereas the dashed line shows the form of the magnetic response function.

scattering on the neutron-energy-gain side (see Sec. II A), but there is a clear difference between the signal from UBe_{13} and $ThBe_{13}$. However, the addition scattering in UBe_{13} is weak.

A. Analysis procedure

Following Goldman *et al.*,[16] the neutron-scattering cross section may be written as

$$\frac{d^2\sigma}{d\Omega\, d\omega}=\gamma_0^2\frac{k_f}{k_i}(g^2/4)2S(Q,\omega)e^{-2W}\ , \tag{5}$$

where

$$S^{xx}(Q,\omega)=S^{yy}(Q,\omega)=S^{zz}(Q,\omega)=S(Q,\omega)\ ,$$

since we have a cubic system. In the above, $\gamma_0^2=0.291$ b, g is the Landé factor,and e^{-2W} the Debye-Waller factor, which, at small Q and low temperature, is taken as unity. The dynamical structure factor $S(Q,\omega)$ is the Fourier transform of the magnetic correlations in the system[10] and can be related to the imaginary part of the susceptibility through the fluctuation-dissipation theorem

$$S(Q,\omega)=(\pi g^2\mu_B^2)^{-1}[n(\omega)+1]\mathrm{Im}X(Q,\omega)\ , \tag{6}$$

where

$$[n(\omega)+1]=(1-e^{-\omega/kT})^{-1}\ . \tag{7}$$

It is this latter factor which ensures that at low temperature ($\omega\gg kT$) there is no signal on the neutron-energy-gain side ($\omega<0$) of the spectra (see Fig. 1).

The imaginary part of the susceptibility may be expressed in terms of the real part of the static susceptibility, $\mathrm{Re}X(Q)$, and a spectral weight function, which we have already defined in Eqs. (1)–(4). We note that this definition of the spectral weight function has its peak at $\omega=0$, and the presence of the detailed balance factor $n(\omega)+1$ can drastically change the appearance of the spectra. In the high-temperature limit $kT\gg\omega$, $n(\omega)+1\approx kT/\omega$, and $S(Q,\omega)$ will peak at $\omega=0$ [see Eqs. (4) and (6)]. In the low-temperature regime, $n(\omega)+1\approx 1$, and $\mathrm{Im}(Q,\omega)$ is measured directly. $S(Q,\omega)$ then shows a peak at $\omega\approx\Gamma$ for the spectral function $P(Q,\omega)$ of Eq. (4). During our analysis of the observed spectra as a function of Q and T, we have found no evidence for a truly inelastic component to $S(Q,\omega)$ (recall that in neutron-energy loss at low temperature our maximum-energy transfer corresponds to $\omega\sim 2$ meV), and we use throughout the Eqs. (1)–(7) to characterize $S(Q,\omega)$ and extract $\mathrm{Re}X(Q)$ from the neutron measurements to compare with the bulk susceptibility data. For this comparison the scattering is put on an absolute scale with the vanadium reference.

The analysis program treats the corrected data (detector efficiency, empty subtraction, etc.) and convolutes a Gaussian (instrumental) resolution function with the cross section as defined by Eqs. (2)–(5). The least-squares fit then gives two parameters A, the total strength of the scattering which is proportional to $\mathrm{Re}X(Q)$, of Eq. (2), and $\Gamma(Q)$, the half width at half maximum of the Lorentzian of Eq. (4). Both these are determined for various values of Q and T.

One unusual aspect of the data analysis is worth commenting on. This material is, of course, mostly Be, which has a very large coherent scattering length and results in very strong phonon scattering. At low energies we expect to see only the acoustic modes which are centered on Bragg points. At low Q their intensity will be small, since phonon intensity varies as Q^2, and we have not seen any phonons at low temperature (they appear clearly at higher temperature when the Bose population factor increases). However, we have seen in *both* the spectra of UBe_{13} and $ThBe_{13}$ evidence for multiphonon contributions near the Q values corresponding to the strongest Bragg reflection (200). Normally, the instrumental resolution function is taken from the vanadium spectra to model the strong elastic incoherent signal, but the effects observed in the $ThBe_{13}$ spectra prompted us to use the $ThBe_{13}$ data itself to model the resolution function. The fits to the UBe_{13} spectra were much improved as a result, especially near the (200) Bragg peak. The (111) Bragg peak is weak and the effects are small at that position.

III. RESULTS

We present first in Fig. 2(a) the HWHM parameter $\Gamma(Q)$ of Eq. (2) as a function of Q at different temperatures. Because the magnetic scattering is so weak, the least-squares analysis does not always converge satisfactorily, and so some values are missing. To avoid confusion we have shown values determined at 0.6 K from the first run and those at 1.5, 4, and 9.7 K from the second run. The values (of both Γ and A) obtained in

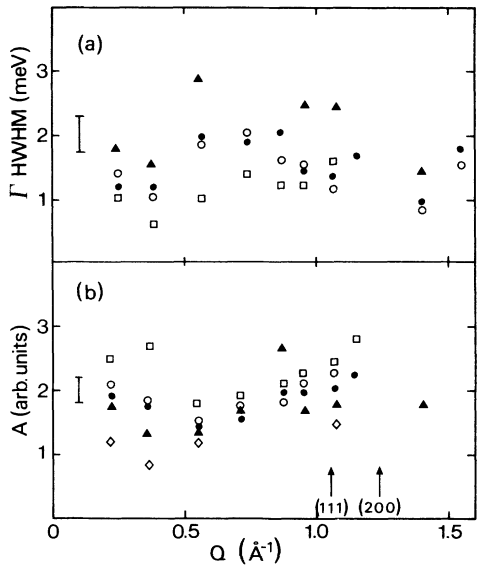

FIG. 2. Parameters (a) Γ, the HWHM of Eq. (4), and (b) the amplitude A of the magnetic response function plotted vs Q, the momentum transfer, for a series of temperature. The positions of the Bragg peaks are also marked on the abscissa. Representative error bars ($\sim 10\%$ for A, somewhat more for Γ) are shown on the left-hand side. Temperatures are -0.6 K (□), -1.5 K (○), -4 K (●), -10 K (▲), and -30 K (◇).

different runs for $T=4$ and 10 K were in excellent agreement. For $T=30$ K the Γ values obtained were not realistic; this is because by this temperature appreciable amounts of the high-energy response ($\Gamma\sim 13$ meV) are beginning to enter the spectrometer window and a more complicated analysis would be required. This is not warranted in view of the limited energy range available. However, an approximate value of A (see below) can be extracted.

The values of A are presented in Fig. 2(b). Recall that apart from a scale factor, which we determine absolutely by calibrating against the known vanadium cross section, A is $\mathrm{Re}X(Q)$ of Eq. (2).

Earlier work by Goldman *et al.*[6] showed that the response in the energy region $\sim 2<E<30$ meV has an energy width of $\Gamma\sim 13$ meV. Because of the detailed balance factor at low temperature, the scattering from this signal peaks at Γ (13 meV). The question arises as to whether the signal we see in our spectrometer window ($0.4<E<1.8$ meV) is not a small part of this higher-energy response function. To examine this hypothesis, we have analyzed the data of Fig. 1(a) by keeping Γ fixed at 13 meV and allowing A to vary. The resulting fits are relatively poor. There is an underestimate of the scattering at ~ 0.6 meV and a compensating overestimate at the highest energies (~ 1.8 meV) in the spectrometer window. However, more important than the quality of the fits is the very large value of A required for the fits in this procedure. A rises to a value of 9.9(4), i.e., by a factor of more than 4 [see Fig. 2(b)]. As discussed below, A is related to the static bulk susceptibility. A value as high as 9.9 signifies a bulk susceptibility more than twice that actually measured and can be totally excluded. Thus our observed signal at low energy cannot simply be fit as the tail of the high-energy response.

IV. DISCUSSION

The most striking feature of Fig. 2 is that in neither Γ nor A is there any statistically significant variation with Q. Given the situation in UPt_3,[4] this is most surprising, particularly since the Curie-Weiss constant Θ in UBe_{13} is strongly negative (between -70 and -100 K for various samples[1]), which indicates strong antiferromagnetic interactions. The zone boundaries are at Q values corresponding to (100), (110), and $(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, which all lie in range $0.5<Q<0.9$ Å^{-1}, and it is noticeable in Fig. 2(b) that, if anything, the signal is in fact slightly weaker in this region. An earlier report [H. Mook (unpublished), but quoted by Liu[11]] shows evidence for a peak at ~ 2 meV in the scattering that has a maximum near (110). This work was on single crystals, but further details are not available with which to compare the sensitivities of the two experiments. The error bars in both our work and that quoted by Liu (see Figs. 11 and 12 of that paper) are large, so that there is no substantial disagreement if the statistical uncertainties are taken into account. Clearly, more precise experiments with larger crystals and a triple-axis spectrometer will be necessary to observe any Q dependence of the low-frequency response. In general, there is a slight tendency for $\Gamma(T)$ to decrease with temperature, but the only statistically significant reduction is from 1.5 to 0.6 K; here, Γ reduces from ~ 1.6 to ~ 1.0 meV.

For the A parameter, there is a significant reduction with increasing temperature, but no noticeable Q dependence at any T. Note that A should show a form-factor dependence [see Eq. (2)], but across this Q range $f^2(Q)$ varies by no more than 15%. The form factor of UBe_{13} appears like[12] a "standard" U $f(Q)$, which may be well approximated[13] for small Q as $f^2(Q)=\exp(-0.07Q^2)$. We should emphasize that our results definitely relate to the magnetic response of $5f$ electrons. Contributions from plane-wave conduction states have a form factor $f(Q)$ that drops rapidly to zero (by $Q\sim 0.5$ A^{-1}), and so would give a quite different variation of A in Fig. 2(b). This is consistent with the form-factor measurements,[12] which show that the susceptibility arises from electrons with $5f$ character.

Naively, one might expect the magnetic response to decrease or even disappear in the superconducting regime (<0.9 K). However, the diamagnetic susceptibility occurs at $Q=0$ only, and theory does not tell us anything about X ($Q>0$) in the superconducting state. Indeed, the antiferromagnetic (i.e., $Q\neq 0$) correlations still exist in UPt_3 (Ref. 4) and URu_2Si_2 (Ref. 9) below T_c, and so in that sense our results are not different. The results quoted by Liu[11] also show magnetic quasielastic scattering at 0.5 K ($T<T_c$), so that this experiment is consistent with our finding. Our results are consistent also with muon spectroscopy on UBe_{13} by Heffner *et al.*,[14] in which they report no change in the zero-field μsr rate as UBe_{13} is cooled from 10 to <0.1 K. The time scale of the muon experiment is μs, whereas the fluctuations probed by neutrons are in the ps range.

Finally, we can discuss the temperature variation of A. After normalizing with the vanadium cross section and taking account of the different masses used, we find that $\mathrm{Re}X(0)=(4.9\pm 0.1)A\times 10^{-3}$ emu/mol. In Fig. 3 we plot $\mathrm{Re}X(0)$ from the bulk susceptibility and neutron measurements. The latter is an average over the first four (most reliable) values of Q. both show a weak T dependence: The values of $\mathrm{Re}X(0)$ derived from A are $\sim 60\%$ of those established by bulk susceptibility, and the latter scaled by 0.6 is shown as a dashed line. Clearly, the two are consistent. Thus the response function measured in these experiments corresponds to (0.6 ± 0.1) of the total magnetic susceptibility. This may be compared with the contribution of Goldman *et al.*[6] (with $\Gamma\sim 13$ meV) that was found to contribute (0.7 ± 0.2) of the total susceptibility at 10 K. Within the uncertainties of the respective measurements of the absolute-scale factor, it is clear that the bulk susceptibility is equally divided between the low- ($\Gamma\sim 1.5$ meV) and high- ($\Gamma\sim 13$ meV) energy components. The fact that no Q dependence is readily apparent in either component lends weight to the recent arguments by Kim *et al.*,[15] in which they claim from a large number of bulk measurements on alloyed samples that single-ion effects are of major importance in UBe_{13}. We might conclude from this that correlation effects occur at Q values too small (i.e., their wavelengths are very long) for us to probe.

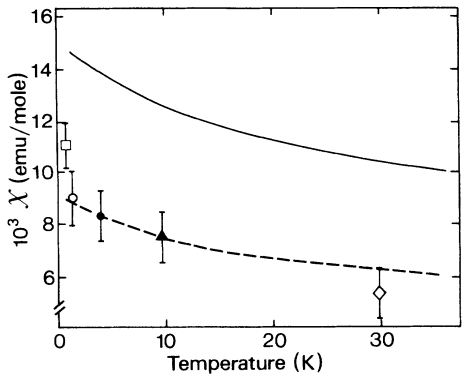

FIG. 3. Variation of susceptibility as deduced from bulk and neutron measurements. The bulk susceptibility (solid line) is taken from Ott and Fisk (Ref. 1, Fig. 21). The dashed line represents these measurements scaled by 0.6. The neutron values are deduced from the first four Q values of A in Fig. 2(b), together with the determined absolute-scale factor.

Our results do exclude the arguments of Cox,[16] who proposed a nonmagnetic doublet crystal-field state as the ground state of UBe_{13}. This would give rise to no quasielastic magnetic scattering, whereas the present work demonstrates the presence of such scattering at low energy and temperature.

Rietschel *et al.*,[17] on the other hand, have proposed that the ground state is a Γ_6 (magnetic) doublet with a $J=\frac{5}{2}$ ($5f^3$) configuration. We should then expect a narrow quasielastic line, consistent with our experimental observation. Furthermore, if single-ion effects dominate,[15] then one would not expect any strong Q dependence. A weakness of their model is that one would expect to see a strong *inelastic* signal corresponding to the Γ_6-Γ_8 transition. They propose that this is the origin of the high-energy scattering seen by Goldman *et al.*[6] However, the latter authors have clearly fit their data to a spectral response function centered on $\omega=0$. (It is the detailed balance factor which makes it appear with a peak at ~ 13 meV at low temperature.) To be more certain of this point, experiments need to be performed with $T > 100$ K, but strong phonon effects then make the interpretation of unpolarized-neutron experiments difficult.

In UBe_{13} the increasing intensity (see Fig. 3) and (perhaps) decreasing width below T_c are very unusual. Furthermore, we had anticipated a Γ of ~ 0.7 meV (hence the use of 3.12 meV as the incident of energy) to give some consistency in the product $\Gamma\gamma$ in Table I. This is not the case. At present it seems difficult to fit the U compounds into any simple scheme.

In conclusion, our experiment has fulfilled its objectives in that we have observed a low-frequency response in UBe_{13}. A similar situation in which both low- and high-energy responses exist in $S(Q,\omega)$ was found in UPt_3 and may also exist in USn_3.[18] The latter is not a HF system in the true sense since $\gamma \sim 170$ mJ/mol K^2), but these similarities do raise the general question of how widespread this "two-level" response is in actinide systems.

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