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Evidence of a magnetic gaplike excitation in URu_2Si_2

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We report the observation of a sharp gaplike magnetic excitation at low temperatures by inelastic neutron scattering in the heavy-fermion $5f$ system URu_2Si_2. At $T=10$ K this sharp excitation occurs at an energy transfer of 5.5 meV and has a Lorentzian relaxational-type tail at higher-energy transfer. The gap rapidly disappears as the sample is heated and at $T=50$ K the response function consists of only one quasielastic line. The Q dependence of the low-temperature peak suggests that antiferromagnetic correlations are important.

The study of the dynamical magnetic response function of $5f$ metallic systems[1] has become a field of much current activity since the discovery of heavy-fermion behavior in a variety of uranium compounds. This is because the energy and the wave-vector range probed by neutron spectroscopy is exactly the regime in which the correlated $5f$ electrons are expected to show unusual properties.

A compound of much present interest is URu_2Si_2. Recent *macroscopic* measurements by Schlabitz *et al.*,[2] Palstra *et al.*,[3,4] and Maple *et al.*,[5] show behavior typical of a heavy-fermion system. All these measurements indicate some type of magnetic ordering at $T_N=17$ K and that superconductivity occurs at $T\simeq 1$ K. Even more important, transport and specific heat data can be interpreted by postulating that a "gap" opens up in the Fermi surface topology below the magnetic transition. The magnitude of this gap varies 8–12 meV, depending on the measurements being interpreted. We report here for the first time the direct observation of such a gap by a *microscopic* probe, viz., inelastic neutron scattering.

Neutron inelastic scattering experiments were performed on a 105-g polycrystalline sample of URu_2Si_2 and on the nonmagnetic reference $LaRu_2Si_2$ prepared by arc melting. X-ray patterns of these materials consisted of sharp diffraction lines corresponding to the expected $ThCr_2Si_2$ structure. The absence of additional lines excludes the possibility of minority ($>2\%$) phases. The lattice parameters derived are $a_0=4.128$ (4.217) Å and $c_0=9.574$ (9.945) Å for URu_2Si_2 ($LaRu_2Si_2$). Time-of-flight measurements have been made using thermal (incident energy $E_0=50$ meV) and hot neutrons ($E_0=250$ meV) with the high-resolution medium-energy chopper spectrometer at the Intense Pulsed Neutron Source at Argonne National Laboratory and with cold neutrons ($E_0=3.1$ meV) at the IN6 spectrometer at the Institut Laue-Langevin, Grenoble, France. In each case independent background measurements and vanadium calibration allow the measurements to be placed on an absolute scale.

We define the scattering function $S(Q\hbar\omega)$ as usual through the neutron cross section $d^2\sigma/[d\Omega\, d(\hbar\omega)]$ corrected by the incident and scattered neutron wave vectors. The magnetic part of $S(Q,\hbar\omega)$ is directly proportional to the dynamical susceptibility $\chi''(Q,\hbar\omega)$.[6] Note that since we have a polycrystalline sample we can obtain no information on the vector dependence of **Q**, and thus write Q as a scalar averaged over all crystalline directions.

The $S(Q,\hbar\omega)$ functions using thermal neutrons are shown in Figs. 1(a)–1(d). For URu_2Si_2 this function contains both phonon S_{ph} and magnetic S_{mag} contributions and these must be separated. The form factor of the magnetic electrons gives rise to a steady decrease in $S_{\text{mag}}(Q,\hbar\omega)$ with Q and no contribution can be seen at high Q ($Q\gtrsim 5$ Å^{-1}). We also know that $S_{\text{mag}}=0$ for $LaRu_2Si_2$. At high Q the phonon spectra of the two materials are similar [Figs. 1(b) and 1(d)], except that the acoustic branch in the density of states of URu_2Si_2 is shifted to lower energy, consistent with the replacement of La with the heavier U atom. At low Q a small amount of phonon scattering remains. A comparison of Figs. 1(c) and 1(d) establishes an empirical Q dependence for the phonon scattering. Applying this to the high Q spectrum of URu_2Si_2, where $S_{\text{mag}}=0$, gives the dotted line in Fig. 1(a). A subtraction then yields the dashed curve in Fig. 1(a), which now represents the magnetic scattering of URu_2Si_2 only and has a clear maximum below 10 meV.

The temperature dependence of $S(Q,\hbar\omega)$ is shown in Fig. 2. The magnetic part (dashed line) obtained by the procedure described above obviously changes drastically in the temperature region $10\leqslant T\leqslant 50$ K. At $T=50$ K it consists of just one quasielastic (QE) Lorentzian line, the half-width at half maximum (HWHM) of which is $\Gamma_{\text{QE}}=6.0$ meV. The full line is a fit to the spectrum also including phonon and elastic scattering. However, at lower temperatures there is a discrepancy between the spectrum and the calculated, adjusted QE line (dotted line in Fig. 2), which is obvious at 20 K and very substantial at 10 K. In fact, it is clear that below $T=30$ K (data not shown) the inelastic scattering *cannot* be described by a single QE-response function. In this sense the data on URu_2Si_2 differ markedly from those on UPt_3 (Ref. 7) or UBe_{13},[8] where the response can be described as a QE line down to the lowest temperatures so far examined.

We shall now examine more carefully the data at $T=10$ K. In Fig. 2(a) the solid line is a guide to the eye whereas

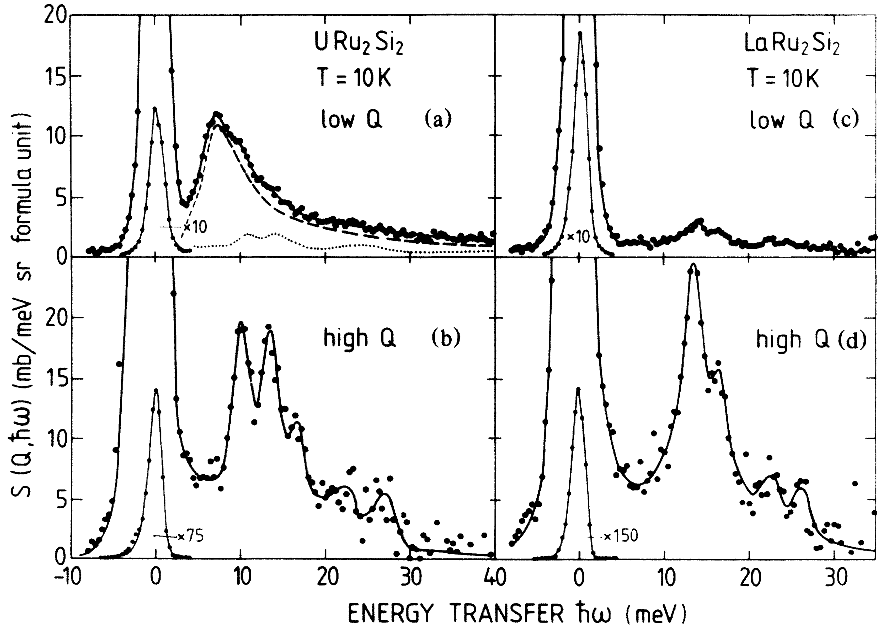

FIG. 1. The spectrum of URu_2Si_2 [(a) and (b)] at $T=10$ K compared with that of the nonmagnetic reference compound $LaRu_2Si_2$ [(c) and (d)]. The incident-neutron energy is 50 meV. For both compounds the covered Q range is $Q=0.8\text{–}1.7$ Å^{-1} (low Q) and $Q=6.2\text{–}8.4$ Å^{-1} (high Q) at $\hbar\omega=13$ meV. The dotted and dashed lines indicate the phonon and magnetic contributions, respectively, in URu_2Si_2.

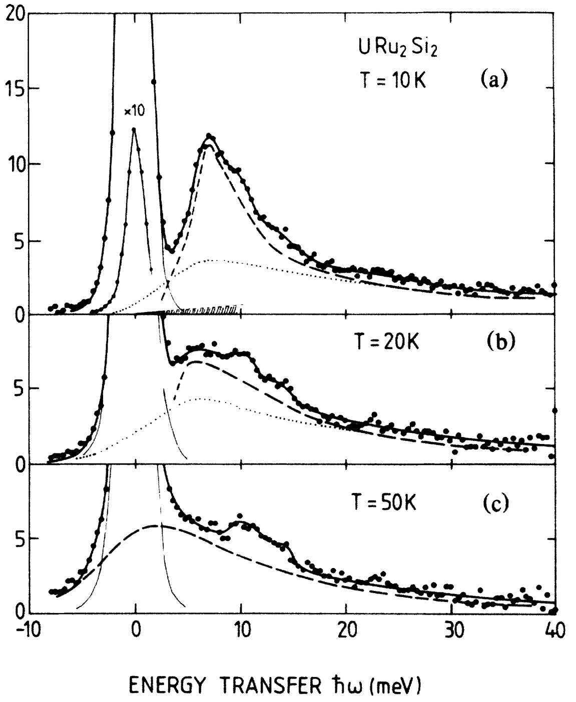

FIG. 2. The temperature dependence of the magnetic response (dashed lines) of URu_2Si_2. The dotted curve is a calculated QE line with a linewidth adjusted to the maximum of the dashed line. The shaded area in (a) gives the upper limit of the quasielastic scattering for $\hbar\omega \leq 2$ meV as deduced from the results of low-energy neutron spectroscopy. For an explanation of the dashed and solid lines, see text.

the dashed line is the magnetic contribution. The small difference between both is simply the phonon contribution as deduced from $LaRu_2Si_2$ [see Fig. 1(c)]. The resolution of the spectrometer used in this experiment is 1.5 meV. It should be kept in mind that this smears out the shape of the inelastic contributions considerably. We believe our results can best be fit with the following function: *no* scattering until 5.5 ± 0.2 meV, at which energy a sharp "step" appears. This is then followed by a long relaxation tail that has a $1/\omega$ intensity dependence. We therefore obtain the gap energy as 5.5 ± 0.2 meV. We should stress that there is, at present, no theoretical justification for this function, but it does give an excellent fit to the data except around the maximum of the excitation where this function overshoots the spectrum by roughly 20%. *Any* further model therefore must clearly include three features: a gap, a sharp rise in the intensity, followed by a slow decrease in the magnetic response function. More complete measurements of this can be done with single crystals and better instrumental resolution. The important point is that we confirm the inferences of Refs. 2–5.

The scattering at $T=20$ K is more difficult to interpret because at this temperature QE scattering is also present. However, our measurements strongly suggest that the gap is still present at this temperature but it has decreased in energy to 3.8 meV.

Our measurements can investigate three further properties of the magnetic response function. The questions we address in turn are the following:

(1) Is there any Q dependence associated with the intensity above the gap?

(2) How sure are we that there is *no* QE scattering within the gap?

(3) Is there any other magnetic scattering, e.g., crystal field states, above the energy transfer of 40 meV?

(i) We examine the Q dependence of the intensity just above the gap by integrating the differential cross section $d\sigma/[d\Omega\, d(\hbar\omega)]$, divided by the form factor $F(Q)$, between 4.1 and 8.4 meV and for each Q. This window omits the phonon contribution at 11 meV and above; however, even with an upper integration limit of 15 meV, the same qualitative results are obtained. The results are shown in Fig. 3. With time-of-flight experiments there is some "averaging" over scalar Q in this window, which amounts to ±7%. Clearly, a maximum occurs around $Q=0.8$ Å^{-1}. We attribute this to antiferromagnetic correlations in the a plane, because it occurs outside the first Brillouin zone of the c axis and just below the Brillouin zone of the basal plane. A further examination of the Q dependence must await experiments on single crystals.

(ii) To examine with good resolution what happens to the QE scattering we have used cold neutrons ($E_0=3.1$ meV) at temperatures between 3 and 120 K. In addition to low-energy transfers $\hbar\omega < E_0$ energy transfers higher than E_0 can be observed at elevated temperatures in processes where the neutrons gain energy from the sample. The resulting spectra are shown in Figs. 4(a) and 4(b). At high temperatures there is an intense and broad QE line, $\Gamma_{QE}=6.7(5.6)$ meV at $T=120(50)$ K, with some small additional phonon contributions. This agrees very well with the results presented in Fig. 2. The quasielastic intensity I_{QE} which we define as the integrated QE scattering as seen in Fig. 4 stays nearly constant down to $T=50$ K and then drops drastically. Below $T=10$ K no indication of any QE scattering within $\hbar\omega \leq 2$ meV was found, as also anticipated by Fig. 4(a), which confirms the interpretation of a gaplike magnetic excitation below $T=10$ K. We have indicated the upper limit of the possible existence of QE intensity as deduced from the spectrum at $T=3$ K (spectrum not shown) by the shaded area in Fig. 2(a). The observed decrease of I_{QE} is faster than linear with T so that since by definition $I_{QE} \propto T\chi_c$, where χ_c is the Curie susceptibility, χ_c tends to zero at low temperature. The implication of this is that the uranium ground state has zero magnetic moment. Note that the existence of the steplike inelastic scattering is anticipated by the spectrum in Fig. 4(a), even though the main scattering is outside the range of this spectrometer.

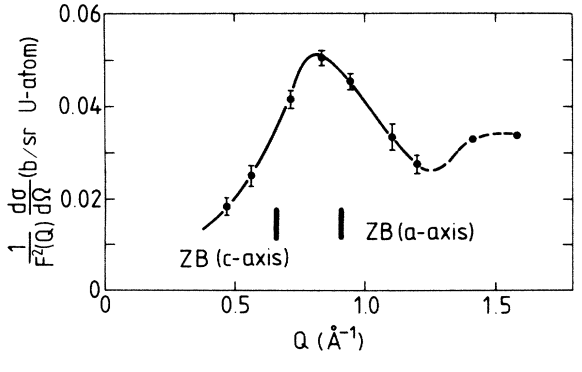

FIG. 3. The Q dependence of the magnetic scattering ($\hbar\omega=4.1$–8.4 meV) of URu_2Si_2 at $T=10$ K (see Fig. 2). The Brillouin zone boundaries along the a and c axes are indicated. The Q averaging at each point corresponds to $\Delta Q \approx 7\%$.

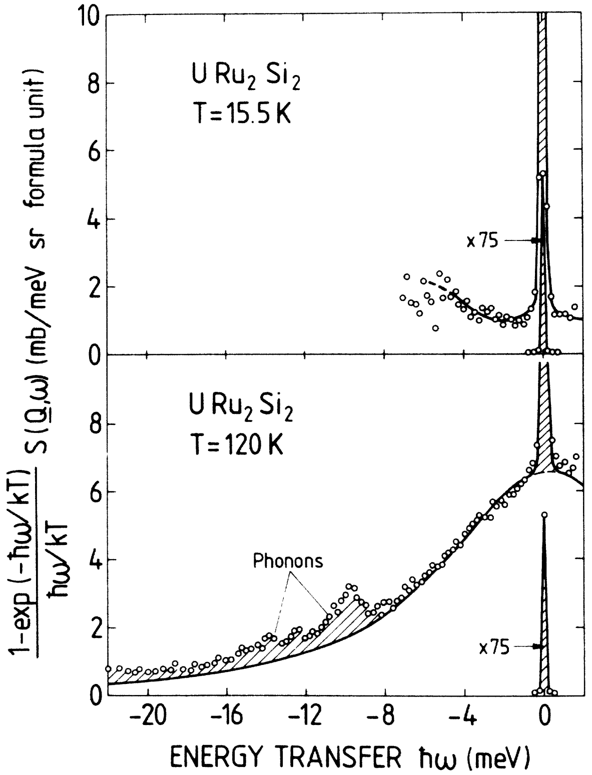

FIG. 4. The spectrum of URu_2Si_2 at $T=15.5$ K and $T=120$ K as observed by cold neutrons with incident energy of 3.1 meV. The shaded area around $\hbar\omega=0$ indicates the incoherent nuclear scattering (elastic line). The ordinate scale is proportional to $T\chi''(\omega)/\omega$. At 10-meV (20-meV) energy transfer $Q=1.5$ (2.3) Å^{-1}.

(iii) To search for magnetic scattering above 40 meV we have used a spectrometer with $E_0=250$ meV. No indication of additional magnetic scattering such as crystal field states has been found up to 150 meV.

Due to a sum rule[6] we were able to calculate both the Curie and the Van Vleck part of the static susceptibility χ_{st} separately from the thermal- and cold-neutron scattering function. In agreement with Schlabitz *et al.*[2] we observe a Curie law above $T=50$ K with $\mu_{eff}=(1.79\pm0.2)\mu_B$ at $T=120$ K (Schlabitz *et al.*: $\mu_{eff}=1.70\mu_B$) and a maximum at $T=50$ K. However, as $T\rightarrow 0$ we find only a Van Vleck term of $\chi_{VV}=(1.1\pm0.4)\times10^{-3}$ emu/mol, which is in disagreement with the results of Schlabitz *et al.*[2] but in reasonable agreement with that of Palstra *et al.*[3]

In discussing the theoretical interpretation of our work we conclude from the strong Q dependence, the steplike rise and the $1/\omega$ relaxation tail of the inelastic magnetic excitation at $T=10$ K, that its interpretation as one or several merged crystal-field excitations can clearly be excluded. Furthermore, this particular shape also seems to exclude the interpretation as a simple magnetic excitation, even with a modest amount of dispersion.

We point out that even the Anderson model, which is generally believed to apply for nonstable f systems, also seems to be inadequate to describe the low-temperature scattering function of URu_2Si_2 at $T=10$ K. This is because none of the various theories[9] based on this model predict *zero* intensity in a finite region around the elastic line, i.e., they still expect finite QE scattering. They also predict that the gap position is independent of temperature and this appears somewhat inconsistent with our results.

In conclusion, our measurements on this highly correlated $5f$ electron system are the first in such a material to show a true *inelastic* scattering, as opposed to the quasielastic (spin fluctuation) scattering found in other heavy-fermion systems[7,8] and more generally in many other uranium compounds.[1] Our measurements directly confirm the inferences drawn from macroscopic measurements[2-5] on this interesting material, that a gap of finite energy occurs in the magnetic response function. One possible explanation is that a spin-density wave occurs and opens up a gap with antiferromagnetic correlations. This would account for the drop of the static susceptibility below $T = 50$ K and for the small magnetic entropy found in the specific heat of the phase transition at T_N. Further investigations, particularly of the magnetic phase transition and the detailed **Q** dependence are necessary. The very strong temperature and unusual energy dependence of the dynamic susceptibility in this system clearly provide a rigorous test for many current theories.

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