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Dynamics of the Magnetization in the Heavy Fermion System $CeCu_6$

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We have studied $CeCu_6$ by inelastic neutron scattering. We found both quasielastic and also inelastic transitions, which we interpret as residual crystal field transitions. The quasielastic linewidth is a strongly nonlinear function of temperature, with approximately $\Gamma_{QE} = 5.0$ meV at 300 K, with a crossover $\Gamma_{QE} = kT$ at about 13 K and with a residual value of $\Gamma_{QE}(0) = 0.50$ meV for $T = 0$ K. Below 5 K the quasielastic intensity I_{QE} decreases linearly with temperature. $\chi_{st} \propto I_{QE}/T$ is in good agreement with direct measurements of the static susceptibility. The data are fully consistent with a nonordering groundstate of $CeCu_6$.

Introduction

$CeCu_6$ is a new member [1–3] of the rapidly growing class of the so called heavy fermion systems. This class of intermetallic compounds is characterized by a large effective mass of the conduction electrons near the fermi level, as detected by specific heat measurements at helium temperatures [1, 4]. The modern heavy fermion classification may be regarded as a generalization of the older classification by the term Kondo compounds or Kondo lattice systems, which has been applied to some of these systems for a long time. Some of these heavy fermion systems become superconducting ($CeCu_2Si_2$, UBe_{13}) or order magnetically (UCd_{11}, $CeAl_2$, $CePb_3$ [5], YbPd [6], YbCuAl [7]) or show both types of phase transitions (URu_2Si_2 [8]). Others remain paramagnetic ($CeAl_3$). $CeCu_6$ seems to belong to the latter subclass, according to the static suceptibility and especially the specific heat, which shows a temperature independent coefficient $\gamma = C/T = 1.53$ J/mol·K^2 [9] (=320 states per eV and unit cell) below 0.5 K; no gap of any type seems to be developing in this compound at the fermi level above 40 mK [9].

Apart from the high electronic specific heat at low temperatures, heavy fermion systems, at least those with Rare Earth components, show a very abnormal temperature dependence of the quasielastic (QE) magnetic line width $\Gamma_{QE}(T)$, as detected by inelastic neutron scattering: while at low temperatures this width is very close to the inverse linear specific heat coefficient $\Gamma_{QE}(0) \cong \gamma^{-1}(0)$*, the width $\Gamma_{QE}(T)$ grows by one order of magnitude or more when the temperature grows to a few 100 K. The Rare Earth heavy fermion systems $CeCu_2Si_2$, $CeAl_3$ [11], YbPd [12] and YbCuAl [13] show invariably also inelastic excitations in the magnetic neutron spectra at energies

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* This relationship follows immediately if one refers the linear specific heat coefficient not to the mole but to the unit cell of the compound, i.e. if one expresses γ in states/energy interval·atom. We feel that this is the most transparent description of the essential physics

of about 10 meV, which are usually interpreted as CF-excitations.

In this paper we report a study of the magnetic excitation spectrum of $CeCu_6$ between 1.5 K-120 K by inelastic neutron scattering. The measurements were done with cold neutrons ($E_0 = 3.07$ meV) at the TOF-spectrometer IN6 at the high flux reactor at the ILL/Grenoble. The choice of cold neutrons was motivated by the following facts:

- the subtraction of phonon scattering is particulary easy at low momentum (Q) transfer, which can be best achieved by low incoming energy;
- the incoming energy of $E_0 = 3.07$ meV is on the other hand large enough to ensure a nearly complete view of the QE-spectrum at low temperatures in energy loss, where it is essential to have $E_0 \gg \Gamma_{QE}(0)$, when $kT \ll \Gamma_{QE}(0)$;
- the spectrometer IN6 has a resolution, which is at least one order of magnitude higher compared to all the other high flux TOF-spectrometers used in the past to study the magnetic spectra of heavy fermions.

Experimental Results and Discussion

We have prepared a sample of 23 g of polycrystalline $CeCu_6$ by arc melting. After quenching the sample was not heat treated. X-ray diffraction revealed sharp Bragg reflexes of the orthorhombic $CeCu_6$ structure. No indications of any other phases (especially $CeCu_5$) could be detected. The lattice parameters are $a_0 = 8.108$ Å, $b_0 = 5.103$ Å and $c_0 = 10.14$ Å. These values match closely those found in literature. We also prepared a sample of $LaCu_6$ with similar weight and found $a_0 = 8.145$ Å, $b_0 = 5.150$ Å and $c_0 = 10.20$ Å.

Inelastic neutron scattering experiments were done with cold neutrons between $T = 1.5$ K and 120 K. The measured spectra were corrected for angular and energy dependent sample absorption, but not for multiple scattering, which is negligible in the case of cold neutrons. They were also corrected for background scattering with the aid of independent background measurements. This procedure yields $(k_1/k_0) S(Q, \hbar\omega)$, where k_0 and k_1 are the incoming and outgoing neutron wave numbers.

In Fig. 1 we have plotted $S(Q, \hbar\omega)$ and in Fig. 2 and Fig. 3 $S(Q, \hbar\omega)$ multiplied by $(1 - \exp(-\hbar\omega/kT)/(\hbar\omega/kT) \propto \frac{T\chi}{\omega}$ against temperature. The plots of Fig. 2 and Fig. 3 reveal directly the symmetric shape of QE- and inelastic (IN) lines.

In the spectra of $LaCu_6$ (Fig. 1) there are clear in-

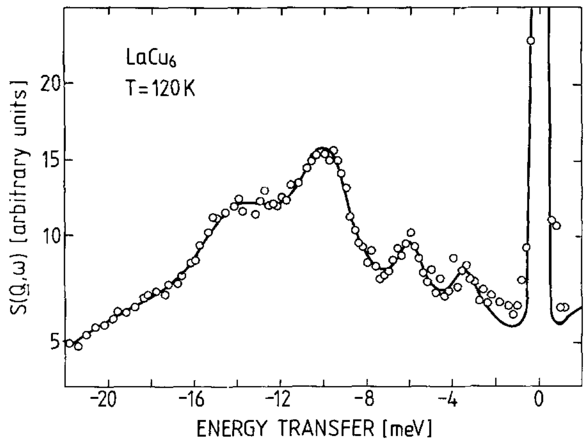

Fig. 1. Phonon spectrum of the reference sample $LaCu_6$ at high momentum transfer (high scattering angles). The full line is a fit to the spectrum

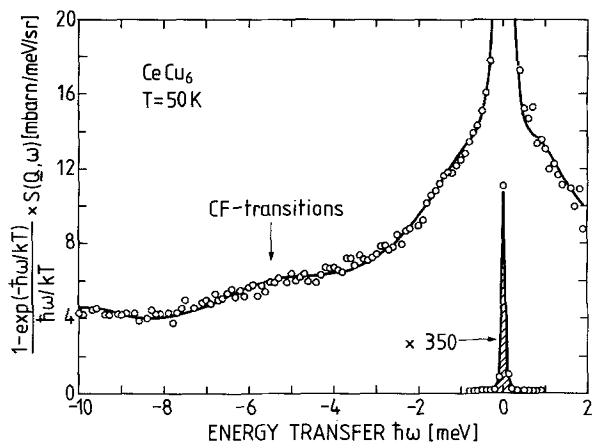

Fig. 2. Inelastic magnetic spectrum of $CeCu_6$ at $T = 50$ K and at low momentum transfer (low scattering angles). The full line is a fit to the spectrum taking 1 QE- and 1 IN-line and the phonon spectrum (Fig. 1) into account

dications of phonon excitations at

$$E_1 = 3.2 \text{ meV}, \quad E_2 = 6.0 \text{ meV}, \quad \text{and}$$
$$E_3 = 9.7 \text{ meV}, \quad E_4 = 13.0 \text{ meV}.$$

The spectra of $CeCu_6$ contain contributions from phonons and from magnetic scattering. As usual the phonon contribution can be identified by comparison with the pure phonon spectra of $LaCu_6$, assuming that the phonon spectra are nearly identical in both compounds. Since the phonons at 3.2 meV and 6.0 meV are less intense and correspond to less momentum transfer than the phonons at higher energies, they quickly diminish in intensity on the way to the forward scattering angles used in taking the

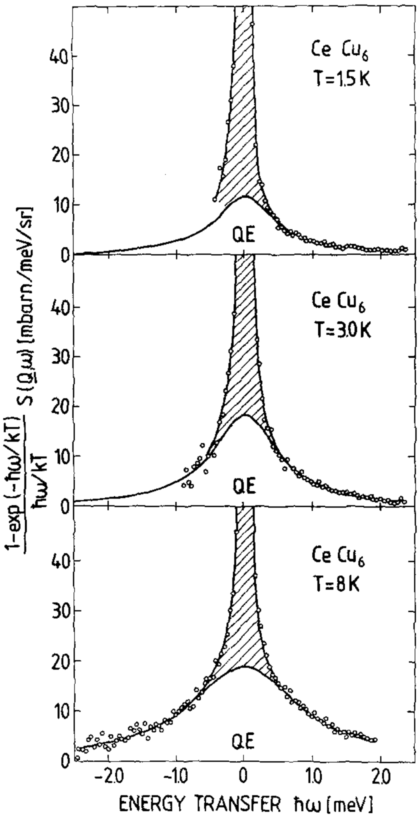

Fig. 3. Quasielastic magnetic spectrum of $CeCu_6$ below $T = 10$ K. The fit to the spectrum (full line) considers the incoherent nuclear scattering (shaded area) and one QE-line with Lorentzian shape

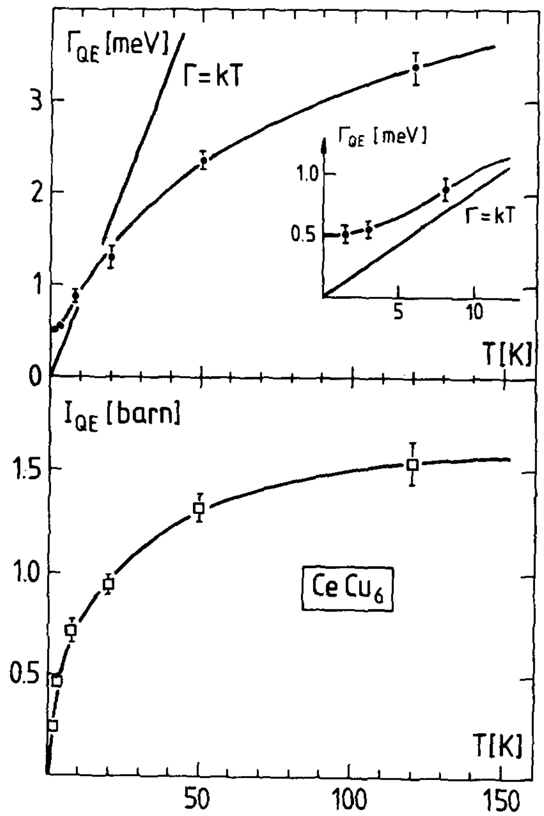

Fig. 4. Temperature dependence of the QE-line width Γ_{QE} (HWHM) and the QE-intensity I_{QE} as defined in Eq. (1) of $CeCu_6$

$CeCu_6$ spectra. Thus, in the case of $CeCu_6$, only the high energy phonons give rise to visible small phonon contributions for $\Delta E < -9$ meV at all temperatures on top of the magnetic spectrum, which dominates for $|\Delta E| < 3$ meV (Fig. 2).

The spectrum of $CeCu_6$ at $T = 50$ K shown in Fig. 2 is typical of a high temperature spectrum. From this spectrum two features can be extracted clearly: a distinct QE-line with Lorentzian shape and with a width of $\Gamma_{QE}(50\,\mathrm{K}) = 2.3$ meV, and a broad inelastic excitation at about $\Delta E = 5.5$ meV. The existence of any other QE-line with a width of less than 10 meV and with an intensity of more than 10% of the first QE-line can be excluded.

To interpret the broad inelastic magnetic excitation one may start as usual by assuming that the excitation at $\Delta E = 5.5$ meV is a crystal field (CF) transition. Support for this interpretation is given by the observation that the excitation energy remains constant at all temperatures. Moreover, specific heat measurements [1, 4] of $CeCu_6$ at $1\,\mathrm{K} < T < 80\,\mathrm{K}$ show a Schottky type anomaly due to a doublet at 5.6 meV above a doublet ground state (the degeneracy follows from the entropy in the specific heat measurements). In order to interpret the magnetic neutron spectra in terms of a CF-scheme, one must realize that in principle three IN-lines are expected, because of the monoclinic point symmetry of Ce in $CeCu_6$. The absence of the other two inelastic excitations in the spectrum may be either due to the fact that the uppermost CF-doublet is at energies of about 11 meV (about twice the first excitation energy) or very far above that value. The recent measurement of the CF-splitting in $PrCu_6$ [14], which reveals a total CF-splitting of 8.95 meV, suggests that $CeCu_6$ should show a corresponding total CF-splitting of roughly 10 meV, due to the slightly more extended 4*f*-shell of trivalent Cerium. Since this is in good agreement with the first assumption, we suggest the following CF-scheme for $CeCu_6$: 0-5.5 meV-11.0 meV.

In Fig. 3 we show the neutron spectra of $CeCu_6$ below 10 K. In this temperature range the inelastic transitions can be ignored for two reasons: On the energy gain side (left hand side from $\Delta E = 0$) the excited levels are no longer thermally occupied and on the energy loss side (right hand side) the incoming energy is less than the first CF-excitation. This

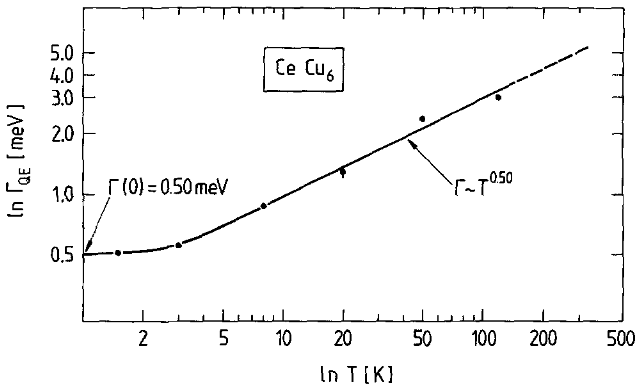

Fig. 5. The square root like temperature behavior of the QE-line width Γ_{QE} above $T=5$ K, shown in a double logarithmic plot

enables one to extract in a very clean manner the temperature dependence of the QE-linewidth Γ_{QE} (Fig. 4(a)) and of the QE-intensity I_{QE} (Fig. 4(b)) of the CF-groundstate doublet.

We point out, that there is no indication of any QE-line with Gaussian shape in our spectra. On the other hand, such additional Gaussian QE-line were found in the magnetically ordering intermediate valent systems $YbBe_{13}$ [15] and YbPd and Yb_3Pd_4 [12]. In these compounds an additional Gaussian QE-line precedes the onset of magnetic order. In all of them the Gaussian line could clearly be distinguished up to temperatures which were roughly 10 times larger than the ordering temperature. From this experience we conclude that $CeCu_6$ will not order magnetically above approximately $T=0.1$ K. This is in fact consistent with the specific heat, which was measured down to 40 mK [9] and showed no temperature dependence of the linear specific heat coefficient.

We observe a drastic decrease of the QE-line width with decreasing temperature. At $T=13.2$ K the QE-linewidth is equal to kT ($\Gamma_{QE}=1.1$ meV). There is a substantial further decrease of the QE-line width below this temperature, but finally one observes a clear saturation at $\Gamma_{QE}=0.50$ meV below 3 K (insert of Fig. 4). In Fig. 5 we have plotted the QE-line width versus temperature in a double logarithmic plot. We find $\Gamma_{QE}(T) \propto T^{1/2}$ above 5 K, which is the same as in $CeAl_3$ at comparable temperatures [11].

We define the QE-intensity I_{QE} (Fig. 4, lower part) by

$$I_{QE}=I_0 \cdot kT \cdot \sum_i \chi_{ii}^{zz}(Q)/\mu_B^2 = I_0 \cdot kT \cdot \chi_c/\mu_B^2 \quad (1)$$

$$I_0 = 1.826 \text{ barn}$$

where χ_{ii}^{zz} are the diagonal parts of the Q-dependent susceptibility [16]. I_{QE} is constant above 150 K. This corresponds to a Curie like behavior of χ_c at these temperatures. Below 150 K I_{QE} first decreases slowly with temperature, perhaps due to a decrease of the effective magnetic moment. At helium temperatures the decrease is linear with temperature. The constant QE-line width below 3 K as well as the linearly decreasing QE-intensity can be observed clearly in Fig. 4 (By definition of (1), the QE-intensity is just the integrated area below the lorentzian (QE) lines in Fig. 3). The linear behavior of I_{QE} indicates that χ_c has become constant below 3 K.

In order to show the correspondence of the measured magnetic scattering intensity to the measured static susceptibility χ_{st}, we have calculated χ_c and χ_{st} from the QE-intensity I_{QE} alone and the total magnetic scattering, respectively (Fig. 6). The full dots give χ_c. This corresponds to the pure Curie terms. The open circles give the susceptibility calculated from the QE- and from the inelastic intensity together; this corresponds to Curie and Van Vleck terms together. Also shown is the susceptibility as measured by Stewart et al. [1]. It is obvious from Fig. 6, that the susceptibility calculated from the measured neutron data does not give the full static susceptibility. For instance a slope of χ_{st}^{-1} vs. T, taken from the open symbols gives an effective moment of $\mu_{eff} = 2.21\,\mu_B$, which is 11% below the effective moment of the measured susceptibility. The deficiency may be due either to a broad magnetic inelastic scattering, or to a very broad QE-line ($\Gamma_{QE} > 6$ meV), which both could be hidden by an overestimation of the phonon contribution. However, it may also be simply due to the uncertainty in the normalization of the spectra by independent vanadium measurements. Unfortunately we cannot decide between these possibilities on the basis of the experimental data taken with cold neutrons.

Besides the QE-intensity I_{QE} there is also the zero temperature QE-line width $\Gamma_{QE}(0)$, which should be related directly to the static susceptibility at lowest temperatures. A more general equation for this dependence is given by the Korringa relation [17, 18], which reads in our case

$$\chi_{st}(0) = \frac{1}{3\pi} \frac{\mu_{eff}^2 \cdot N}{\Gamma_{QE}(0)}. \quad (2)$$

This expression for χ_{st} is valid for an N-fold degenerate multiplet at temperatures very small compared to $\Gamma_{QE}(0)$ assuming a Lorentzian distribution of the amplitude of each of the N states over energy. $\Gamma_{QE}(0)$ is the HWHM of the Lorentzian at $T=0$.

As an aside we mention that another phenomenological formula is often used to describe the suscepti-

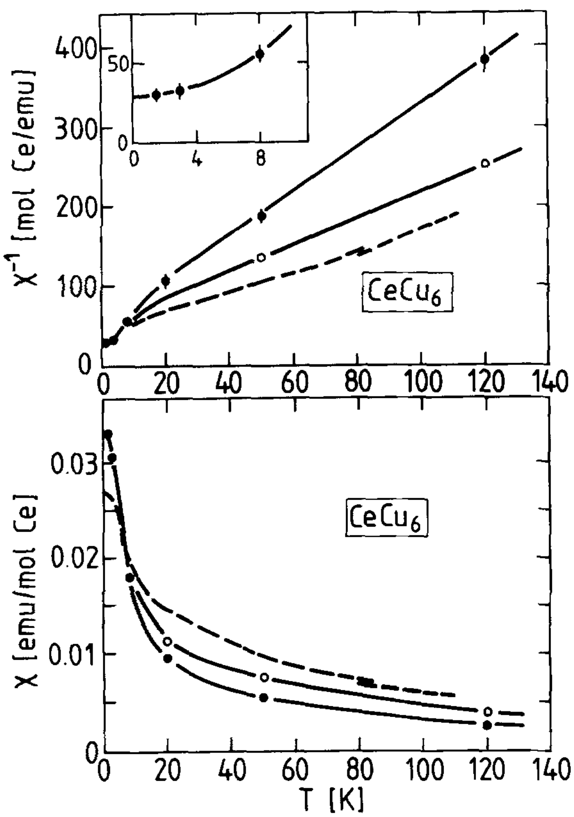

Fig. 6. The temperature dependence of the static susceptibility of $CeCu_6$ as measured (broken line [1]), compared to the values calculated from the neutron spectra (full lines). The full circles denote the susceptibility calculated from the pure QE-scattering and the open circles that from the total magnetic scattering

bility in terms of an effective temperature, namely

$$\chi_{st}(0) = \frac{1}{3} \cdot \frac{\mu_{eff}^2}{k T_f}.$$

Obviously the connection between T_f and $\Gamma_{QE}(0)$ is

$$k T_f = \frac{\pi}{2} \Gamma_{QE}(0)/N$$

which gives $T_f = 4.6$ K in $CeCu_6$ from $\Gamma_{QE}(0) = 0.50$ meV and $N = 2$.

We shall now show that the neutron data given above enable an unequivocal determination of μ_{eff} from (2) for $T < 3$ K. Note first of all, that $\Gamma_{QE}(0)$ can be taken directly from the spectra. Secondly, our above discussion shows, that we are dealing with a CF-doublet groundstate; therefore $N = 2$. Thirdly, since the measured integrated intensity I_{QE} gives χ_c (Eq. (1)), which agrees with χ_{st} below 3 K ($\chi_{st} = 0.035$ emu/mol), we can determine μ_{eff} with confidence. We find $\mu_{eff} = 1.60 \pm 0.07\,\mu_B$. Contributions to χ_{st} from VanVleck terms are small compared to this pure Curie term of the CF-ground state, to order 1/13. We see that this moment is reduced considerably with respect to the Hund's rule ground state $\mu_{eff} = 2.54\,\mu_B$. If one uses (2) with $N = 2J + 1 = 6$ (no CF-splitting), one obtains $\mu_{eff} = 0.92\,\mu_B$, which is totally inconsistent with the Hund's rule ground state (of course the assumption $N = 6$ is also inconsistent with the entropy found in the specific heat measurement [4].).

One of the problems in the physics of heavy fermion systems is the experimental determination of the so called Wilson ratio

$$R = \pi^2 k_B^2 \frac{\chi_{st}(0)}{\mu_{eff}^2 \cdot \gamma}. \tag{3}$$

For $CeCu_6$ we find 1.95 for the r.h.s. of Eq. (3) with $\gamma = 1.53$ J/mol/K^2 and $\chi_{st}(0) = 0.035$ emu/mol and $\mu_{eff} = 1.60\,\mu_B$. With the assumption that Kondo theory of dilute alloys holds also for Kondo compounds, a more elaborate theory of the Kondo effect [19] implies that the r.h.s. of Eq. (3) has to be lowered by a factor of $N - 1/N$ leading to 0.98 for the Wilson ratio in $CeCu_6$.

Conclusion

Above $T = 3$ K $CeCu_6$ exhibits the same square root like behavior of the QE-linewidth as other heavy fermion systems ($CeCu_2Si_2$, $CeAl_3$) or Kondo lattice systems (CeB_6, $CeAl_2$). Below 3 K neutron spectra exhibit only a single QE-lorentzian of constant line width ($\Gamma_{QE}(0) = 0.50$ meV). No additional gaussian QE-line could be observed, which indicates that no magnetic ordering can occur above $T = 0.1$ K. This prediction is in agreement also with the measured QE-intensity, which vanishes linearly with temperature below 3 K. This corresponds to a finite value of the measured static susceptibility. The neutron scattering cross section is evaluated to extract the effective moment and the QE-line width near $T = 0$. We find $\mu_{eff} = 1.60\,\mu_B$ from our measurements. Using this effective moment and the measured linear specific heat coefficient, we find a Wilson ratio of 1.95. At high temperatures the neutron spectra show only one inelastic transition at 5.5 meV, from which we indirectly derive the CF-scheme: 0–5.5 meV–11.0 meV.

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