Field dependence of the magnon dispersion in the Kondo lattice $CeCu_2$ up to 12 T

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$CeCu_2$ can be classified as a Kondo lattice which shows antiferromagnetic (AF) order below T_N=3.5 K [R. Trump *et al.*, J. Appl. Phys. **69**, 4699 (1991)]. The orthorhombic crystal and the simple AF magnetic structure with two magnetic moments in the primitive unit cell requires two magnon modes which are observed in zero and low magnetic fields and well described by spin wave theory. However, at higher fields, at and above 3 T, an unexpected, additional magnetic excitation is observed. In contrast to the two low-energy magnon modes, it exhibits a steeper (factor 2) field dependence and a flat dispersion. Its origin is unclear. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1850369]

INTRODUCTION

Anomalous rare earth and actinide compounds, including the so-called Kondo lattices and heavy fermion systems have been studied extensively in the past. These systems are investigated because of the fluctuations in the *f* shell which strongly modify the low-temperature properties. $CeCu_2$ is such a Kondo lattice (T_K=6 K) which also shows antiferromagnetic (AF) ordering at T_N=3.5 K. It crystallize in the orthorhombic $CeCu_2$ structure (Imma No. 74, a=0.442 nm, b=0.704 nm, c=0.745 nm). The primitive unit cell contains two Ce atoms and the J=2/5 Hund's rule ground state of the Ce^{3+}-ions is split into three doublets due to the crystalline electric field (CEF). That leads to two magnon modes and a CEF level scheme with energies of 9 and 23 meV. In the magnetically ordered state the moments are aligned along the c axis even though the CEF prefers the a direction as the easy axis of magnetisation.[1] The reason for this behavior might be that the magnetic exchange is strongly anisotropic in the ac plane and the exchange interaction cancels out the CEF anisotropy.

EXPERIMENTAL RESULTS

With inelastic neutron scattering experiments in magnetic fields up to 12 T the dispersion of the magnetic excitations was investigated (Fig. 1). The experiments have been performed at the triple axis spectrometer IN12 at the Institut Laue-Langeuin in Grenoble, France, in different Brillouin zones, at different magnetic fields, and at slightly different temperatures (1.6, 1.8, and 2.1 K) well below the magnetic ordering temperature. The orthorhombic crystal and the simple AF magnetic structure with two moments in the primitive unit cell requires two magnon modes.

Preliminary[1,2] neutron measurements at zero magnetic field showed the predicted two magnon modes. Additional experiments were performed at 5 T to determine the field dependence of these magnon modes,[2] i.e., for the mode with stronger structure factor along the [0K0] direction. Modeling

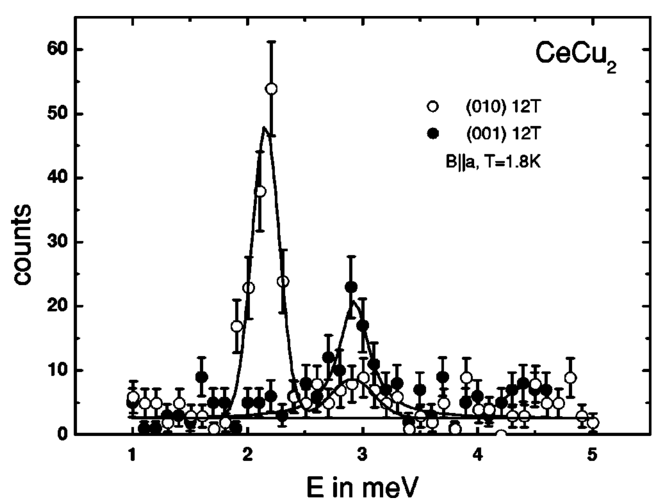

FIG. 1. Inelastic neutron spectra of $CeCu_2$ measured at IN12 with a magnetic field of 12 T along the a direction. At two orthogonal X points in the bc-scattering plane we obtain different intensities because of different structure factors.

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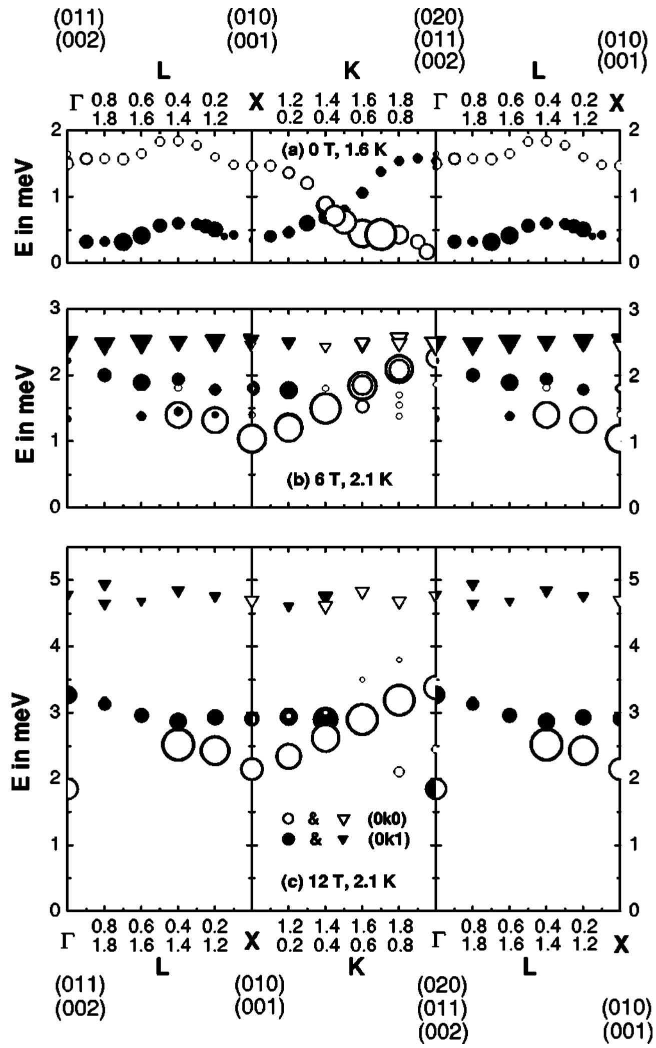

FIG. 2. Magnon dispersion curves for $CeCu_2$ measured at IN12 in different Brillouin zones and at different magnetic fields: the upper diagram at zero field, the middle at 6 T, and the lower at 12 T. The size of the symbols correlates with the measured intensity of the excitations. In the plots at higher fields a third, unexpected excitation occurs (triangles), which is almost flat.

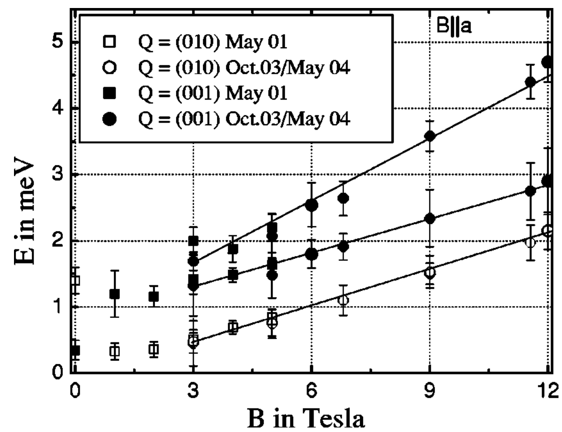

FIG. 3. Field dependence (B$\|$$a$) of the low-energy magnetic excitations of $CeCu_2$ measured at the X points (001) and (010). Data are from three different experiments using two different crystals and cryomagnets, at slightly different temperatures (1.6 and 1.8 K). The bars indicate the full width at half maximum of the excitations. Below 3 T we observe the two expected magnon modes. Above 3 T a third, unexpected magnetic excitation occurs with considerably steeper slope than that of the two lower magnon modes.

of the observed drastic change of the dispersion of this mode (positive slope for zero field, negative slope for 5 T for the Γ-X direction) required a spin wave calculation with anisotropic exchange interactions.[2] An additional finding from these experiments was the observation of an apparent splitting of the magnon mode with the higher energy as deduced from different, field dependent scans at selected reciprocal lattice points (mainly the X point). The splitting was only observed in fields at and above 3 T.

To elucidate and verify this finding we performed an inelastic neutron scattering experiment on another $CeCu_2$ single crystal in fields up to 12 T. The crystal was mounted in the *bc* scattering plane with the vertical field in the *a* direction. Magnetization data for fields along the *a* direction[1,3] show that for temperatures below 2 K, the system switches from the AF state into the ferromagnetically induced state for field values between 1 and 2 T.

In Fig. 2 we present the measured dispersion curves along the Γ-X direction for three different magnetic fields: zero field, 6 T, and 12 T. Comparison with previous measurements yields agreement within the accuracy of the corresponding data. We can deduce from the data that the apparent splitting of the magnon mode with the higher energy is in fact an additional excitation with an flat dispersion as clearly seen in the dispersion curves at 6 and 12 T.

The field dependence of the energy of the magnetic excitations at the X point in the ordered state is presented in Fig. 3, including recent and previous data. Note the different structure factors for data taken at (001) and (010) in the AF and the ferromagnetically induced state.

DISCUSSION

A preliminary model calculation[5] was based on an anisotropic exchange interaction treated in mean-field theory and using the random phase approximation. The results of this calculation agree well with the measured magnon dispersion (two modes) at low fields. However, at higher fields (above 3 T) the additional, almost nondispersive excitation cannot be explained within this model.

In a first approximation, the field dependence of the magnon modes in the field-induced ferromagnetic state can be described by the Zeeman splitting of the ground state doublet ($I+>$, $I->$)

$$\text{Zeeman splitting:}\quad \Delta = 2g_j m\mu_B H, \tag{1}$$

$$\text{magnetic saturation moment:}\quad M_{sat} = g_j\langle J\rangle\mu_B, \tag{2}$$

$$\text{if CEF splitting} \gg mH \gg \text{kT:}\quad M_{sat} = g_j m\mu_B, \tag{3}$$

$$\text{therefore}\quad \Delta = 2M_{sat}H. \tag{4}$$

($m=\langle +|J_a|+\rangle$, g_j=Landé factor, μ_B=0.057 88 meV/T; the crystal electric field (CEF) splitting of $CeCu_2$ was deduced to be 9 and 23 meV.

From the slopes of the lines in Fig. 3 the magnetic moments can be determined according to Eq. (4). The values for the slopes are 0.18 meV/T (lower), 0.17 meV/T (middle), and 0.31 meV/T (upper). Within the error, the slopes of the two lower lines agree. That leads to saturation moments of $M_{sat}=1.5\ \mu_B$ (for the two lower excitations) and $M_{sat}=2.7\ \mu_B$ (for the upper excitation).

In the literature, the value for the saturation moment in the observed direction ($H\|a$) is reported to be 1.6 μ_B.[1,3] It can be seen that the value for the two lower excitations is in good agreement to that. The moment, which leads to the third excitation must be much higher (factor 2) than the known saturation moment.

A possible explanation could arise from the strong crystal-field phonon interaction which was found in $CeCu_2$.[6] In previous measurements it was found that phonons with a suitable symmetry interact with the crystal field transitions and becomes shifted in energy more than expected from the normal temperature behavior. At very high magnetic fields, a structural conversion was reported.[7] A possible scenario for this unusual excitation might be the 7 meV optical phonon, which at 3 T maybe becomes very soft and then exhibit the observed behavior.

A second explanation could be the large quadrupolar interaction, which have been reported in RCu_2 (R=rare earth) compounds.[4] A dynamical treatment of these interactions leads to a splitting of the CEF excitations. An explanation of the additional mode could be based on such a model, in which the 9 meV CEF transition is split and the lower branch occurs at 1 meV and then shows a large field dependence.

To verify the assumptions given above experiments with polarized neutrons are necessary because they can distinguish between magnetic and structural excitations.

CONCLUSION

By inelastic neutron scattering experiments at high magnetic fields using different samples of the Kondo lattice compound $CeCu_2$, an unexpected magnetic excitation has been observed in the magnetically ordered state in addition to the two expected magnon modes. Its flat dispersion relation and its field dependence differ drastically from the behavior of the two magnon modes. The origin of this additional magnetic excitation is still unclear.

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