Spin-resolved neutron spectroscopy from the heavy Fermion compound CeCu_6

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Abstract

Neutron time-of-flight spectroscopy with neutron polarisation analysis permits the in situ separation of magnetic and lattice vibrational energy spectra. Preliminary experiments on the heavy Fermion compound, CeCu_6 , in which the Ce magnetic moment is suppressed by the Kondo effect, allow an indicative separation of a broadened crystal field transition and features due to lattice vibrations. An inelastic spin-flip feature at -12 meV is due to the crystal field while an inelastic non-spin-flip feature at -6 meV is predominantly due to phonon scattering.

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The heavy Fermion behaviour in CeCu_6 is a consequence of the competition between the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction and the Kondo effect due to the admixture of the 4f and conduction electrons. The interaction between the heavy Fermion and crystal field (CF) excitations leads to broad transitions which are further complicated by the presence of phonon scattering over the region of interest. A number of investigations of the CF level scheme in the heavy fermion compound

CeCu_6 have been made. These include inelastic neutron scattering (INS) measurements [1–3]. These measurements proposed different CF level transitions. Walter et al. proposed a transition at 5.5 meV from a 50 K measurement [1]. Goremychkin and Osborn found possible transitions at 0, 7.0 and 13.8 meV from a 20 K measurement [2]. Stroka et al. reported possible transitions at 7–8 and 12–14 meV from a 3 K measurement.

Conventional INS spectra without polarisation analysis (PA) require an additional spectrum of an equivalent system with a non-magnetic rare earth to distinguish between magnetic and lattice spectral features. Neutron time of flight spectroscopy with PA analysis permits the in situ separation of magnetic and lattice vibrational energy spectra [4]. In this work we report on preliminary measurements of this type performed on CeCu_6 .

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Above 230 K, CeCu_6 has an orthorhombic Pnma structure in which the Ce atoms are located at 4c sites having monoclinic m (C_s) local symmetry [5]. Below 230 K a slight monoclinic distortion takes effect and CeCu_6 has the monoclinic $\text{P2}_1/\text{c}$ structure with the Ce atoms located at 4e sites having triclinic 1 (C_1) local symmetry [6]. The Ce^{3+} ion has a $^2F_{5/2}$ ground state which is split into three Kramer's doublets by the crystal field due to the surrounding ions and hence there are three transitions between the CF split levels.

A polycrystalline CeCu_6 sample was prepared by argon-arc melting of stoichiometric amounts of high purity Ce and Cu. The ingot was annealed for 120 h at 900°C in flowing argon. The annealed sample was analysed by neutron diffraction at ANSTO. A Rietveld refinement showed the composition to be very close to pure, with at most a few atomic percent of Cu metal as impurity. As Cu will not contribute to the CF spectrum, this was deemed quite acceptable.

Neutron time-of-flight spectra were taken on LONGPOL, at 26 K with an incident wavelength of $3.6 \text{ \AA}$ (6.3 meV), using the polycrystalline sample. The neutron polarisation direction was along the elastic scattering vector of the middle of eight detectors which spanned an angle of 50° ($0.3\text{--}1.75 \text{ \AA}^{-1}$). The polarisation of the beam was statistically chopped and the spectrum reconstructed by cross-correlation of the flipping sequence with the time sequence of the intensity in the detectors [7]. The flipping sequence was a shift register sequence of 31 elements and the ratio between flipper on and flipper off was 15/16. Shift register sequences have an autocorrelation function which is a single triangle at zero shift. This is illustrated schematically in Fig. 1.

In the configuration with neutron polarisation along the scattering vector the magnetic excitations scatter with neutron spin-flip (SF). Insofar as all the detectors are not at the inelastic scattering vector and that the inelastic scattering vector deviates from the elastic scattering vector most, but not all, of the magnetic scattering is with neutron SF. The lattice vibrations, however scatter without neutron SF. The reconstructed spectrum is proportional to the non-spin-flip (NSF) minus SF intensity [4] as is illustrated in Fig. 1. As a

Fig. 1. Schematic explanation of the random or pseudo-random time of flight system: (a) part of the pulse sequence sent to the spin flipper; (b), (c) the resulting variations in the non-spin-flip (NSF) and spin-flip (SF) scattering intensities at the detector. (d) Also shown are simple time-of-flight spectra resulting from the cross-correlation of impressed and resulting time sequences.

result the magnetic scattering is largely discriminated from nuclear scattering for which there is no change in polarisation. This is important as the nuclear inelastic scattering dominates the spectrum of CeCu_6 [2].

The spectrum obtained at 26 K summed over all detectors except one is shown in Fig. 2. One detector is excluded as it contained a large Bragg peak. With an incident energy of 6.3 meV, the usable data was mostly with neutron energy gain. The data shows a large nuclear (NSF) elastic peak, a broad nuclear (NSF) feature around -6 meV and two magnetic (SF) features at -2 and -12 meV .

The magnetic feature near -2 meV is the wing of the quasielastic peak first seen by Walter et al. This is supported by the observation of a similar

Fig. 2. Neutron time-of-flight energy spectrum obtained from CeCu_6 at 26 K as measured on LONGPOL. The crystal field (magnetic) scattering appears as dips or negative going peaks, lattice scattering appears as positive peaks.

feature of similar energy on the neutron energy loss (right-hand side) of the elastic position, suggesting that the nuclear elastic scattering is situated within a broader magnetic quasielastic dip. The magnetic feature at -12 meV is consistent with a CF transition identified by both Goremychkin and Osborn and by Stroka et al., although the full extent of this very broad transition has not been mapped, nor have higher energy transitions been ruled out. An inelastic scattering event at -6 meV has been identified as a CF transition by all previous studies. However, Fig. 2 shows a positive cross-correlation at this energy, indicating predominantly NSF scattering which cannot be due to a CF transition. This demonstrates the key strength of the PA experiment.

The spectrum in Fig. 2 is proportional to the difference between NSF and SF scattering. However the spectral information is superimposed on a relatively large uncorrelated background related to all the scattering. Definitive separation of NSF and SF events can be achieved by switching the neutron polarisation direction be-

tween the present geometry and one with polarisation perpendicular to the scattering plane. In the latter geometry the spectral information contains only NSF scattering and a subtraction of the two sets of data not only removes the uncorrelated background but isolates the magnetic scattering. The present spectra can only reveal the predominant mode of scattering at each energy transfer. Definitive separation must await an experiment in which the polarisation direction is switched. This is being pursued.

Further data will be taken at higher temperatures so that higher levels can be populated and further possible crystal field transitions seen. The transitions are broadened at higher temperatures and this might limit their visibility. We also plan to examine samples in which one Cu site is substituted with Au. This weakens the Kondo interaction and sharpens the CF levels [3].

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