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# High luminosity time of flight with polarisation analysis: CeCu<sub>6</sub>

S.J. Harker<sup>a,1</sup>, T.J. Hicks<sup>a,\*</sup>, S.J. Kennedy<sup>b</sup>, Dehong Yu<sup>b</sup>

<sup>a</sup>School of Physics, Box 27, Monash University, Vic. 3800, Australia <sup>b</sup>Bragg Institute, Ansto, Private Mailbag, Menai, NSW 2234, Australia

#### Abstract

The magnetic excitation spectrum of the heavy fermion system  $CeCu_6$  has been obtained by pseudo-randomly chopping the polarisation of the beam on LONGPOL and measuring the time of flight with polarisation analysis before the detectors. Separation of the magnetic part of the spectrum was made by changing the polarisation direction at the specimen. This method achieves a greater luminosity at the specimen compared with conventional methods at the expense of a background proportional to the total scattering from the specimen.

The form of the spectrum is as expected with two transitions and considerable inelastic intensity centred on zero energy transfer. The crystal field spectrum of the  $Ce^{3+}$  ion is considerably broadened by the Kondo nature of the system.

An analysis of the experimental accuracy was made based on the statistical variation between the runs which make up the experiment. LONGPOL is a very slow instrument, based as it is on a thermal source with an incident wavelength of 3.6 Å. The demonstrated possibility to obtain a spectrum incorporating polarisation analysis provides a basis to evaluate the usefulness of future implementation.  $\bigcirc$  2007 Published by Elsevier B.V.

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### 1. Introduction

To be able to definitively isolate the magnetic part of a crystal field spectrum would be a powerful aid in its analysis and interpretation. Until now separation has been achieved with the aid of a second non-magnetic specimen in which the phonon spectrum closely resembles that of the magnetic specimen.

The  $Ce^{3+}$  ion in  $CeCu_6$  has low lying crystal field levels which are seen by neutron time of flight spectroscopy [1]. The local symmetry is triclinic so that the crystal field splits the ground state into three Kramer's doublets. Neutron spectroscopy sees two transitions of approximately 7 and 14 meV [1,2] which are interpreted as being between the ground state and the first and second excited states. The transitions are broad because of the mixing between atomic and conduction electrons described by the Kondo effect. The atomic moment is stabilised by the addition of a small amount of gold and the transitions become sharper [2].

The amplitude of the matrix elements connecting these states is by no means established and a recent single crystal study with polarisation analysis requires unexpected amplitudes to explain the results [3].

#### 2. The pseudo-random method

The pseudo-random method has been thoroughly analysed by von Jan and Scherm [4] and shown to be superior in many circumstances. With polarised neutrons, the pseudo-random method consists of providing the spin flipper with a sequence of square wave currents which switch the flipper between no neutron spin flip and full spin flip. For elastic scattering, there is one time of flight from flipper to detector so that correlation between the sequence impressed on the spin flipper and the intensity modulation at the detectors has one peak at the time of flight. This is

<sup>\*</sup>Corresponding author. Tel.: +61 3 9885 7690; fax: +61 3 9005 3607. *E-mail address:* trevor.hicks@sci.monash.edu.au (T.J. Hicks).

<sup>&</sup>lt;sup>1</sup>Now at Physical, Environmental and Mathematical Sciences, UN-SW@ADFA, Canberra, ACT 2600, Australia.

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Fig. 1. Schematic explanation of pseudo-random time of flight system: (a) part of the pulse sequence sent to the spin flipper; (b) and (c) the resulting variation for NSF and SF elastic scattering intensities; (d) the resulting cross-correlated spectra for both cases.

illustrated in Fig. 1, for a sequence which is equally on and off, where schematic (a) shows the modulation at the flipper with time, (b) and (c) show the resulting modulation at the detectors at an elastic time of flight  $\tau_0$  later for NSF and SF scattering, and (d) the resulting cross-correlated spectra from the two processes. This can also be demonstrated for each inelastic feature.

For special sequences, like shift register sequences, it can be seen that both spectra lie on top of a background which is the average of the upper and lower levels of the modulated intensity. The analysis which follows is for a sequence with equal flipper on and off times.

#### 3. Separation of the magnetic cross-section

The magnetic cross-section can be isolated by changing the neutron polarisation direction at the specimen. Specifically what is separated is the magnetic spectrum *without* its associated background of ignorance.

In this experiment, two polarisation directions were chosen, one perpendicular to the scattering plane and the other approximately perpendicular to the beam direction in the scattering plane. The latter situation is illustrated in Fig. 2.

For a many-element sequence, with the polarisation P perpendicular to the scattering vector Q the background level is proportional to the sum of all cross-sections at all energies

$$B \propto \bar{\sigma}_{\rm NSF} + \bar{\sigma}_{\rm SF} = \bar{\sigma}_{\rm nuc} + \bar{\sigma}_{\rm si} + \bar{\sigma}_{\rm m}.$$
 (1)



Fig. 2. The geometry of the scattering showing the in-plane polarisation direction.

This is the same for the polarisation in any direction although the proportion of the cross-section in NSF and SF processes in  $\bar{\sigma}_m$  will change with polarisation direction.  $\bar{\sigma}_{si}$  is the nuclear spin incoherent cross-section. The bars average over time of flight.

However, the *structure* of the spectrum is proportional to the difference in the NSF and SF processes and this will be different for the two polarisation directions. For P perpendicular to Q the difference is

$$D_1 \propto \sigma_{\rm nuc} - \frac{1}{3}\sigma_{\rm si}.\tag{2}$$

In this geometry, for the isotropic distribution of magnetic vector in a paramagnet, the magnetic cross-section subtracts out. For P in the scattering plane and not perpendicular to Q the magnetic cross-section does not completely subtract out

$$D_2 \propto \sigma_{\rm nuc} - \frac{1}{3}\sigma_{\rm si} - \sigma_{\rm m} \tag{3}$$

and the difference is

$$D_1 - D_2 \propto \sigma_{\rm m},\tag{4}$$

which is the part of magnetic cross-section which depends on the angle between P and Q in the scattering plane. The corrected magnetic cross-section is

$$\sigma_{\rm mc} = \frac{\sigma_{\rm m}}{\cos^2 \alpha},\tag{5}$$

where  $\alpha$  is the angle between **P** and **Q** which will change with energy transfer but is always calculable.

## 4. The experiment

LONGPOL is a slow instrument with an incident beam intensity of about  $5 \times 10^4 \text{ n/cm}^2/\text{s}$  at 3.6 Å (6.3 meV) [5] and even with a sample volume of 4 cm<sup>3</sup>, it was clear that long counting times would be necessary. After the supermirror polariser, the beam traverses a Mezei flipper capable of rapid pulsing and scatters off the specimen into supermirror analysers placed before eight detectors. It was

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