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## HIGH ENERGY NEUTRON SPECTROSCOPY IN CRYSTAL FIELD SPLIT 5f SYSTEMS

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**Abstract:** The new generation of spallation neutron sources, characterized by an intense epithermal flux, allows the observation of magnetic excitations up to several hundreds of *meV*. It is then possible to determine the crystal field transitions in a number of actinide systems, as it is illustrated in the case of *UO<sub>2</sub>*, *UOS* and *NpO<sub>2</sub>*.

### 1. Introduction

The knowledge of the crystal field (CF) is often essential to describe the magnetic and thermodynamic properties of actinide systems and also to answer fundamental questions about the behaviour of the 5f electrons. As known, neutron inelastic scattering provides the most effective method to observe CF transitions in opaque samples. In the case of actinide systems, the kinematic constraint of the scattering process becomes particularly severe at high energy transfer, because the form factor  $f(Q)$  falls very rapidly with  $Q$ . Neutrons with high incident energy must then be used and, for this reason, spallation neutron sources with a large slowing down component in the epithermal region of the neutron spectrum are better suited than steady-state reactors. We present here a short review of some experiments recently performed on the HET spectrometer at the UK spallation source ISIS of the Rutherford Appleton Laboratory.

### 2. Results and Discussion

The first example is the *CaF<sub>2</sub>* structure compound *UO<sub>2</sub>* for which the ninefold degeneracy of the ground multiplet is resolved into two triplets, one doublet and one singlet by the cubic CF above the ordering temperature  $T_N = 30.8K$ . Below  $T_N$ , an internal distortion of the oxygen sublattice reduces the symmetry, giving rise to a further splitting of the levels. The inelastic neutron scattering cross section at  $T = 50K$  shows two peaks, at 150 and 166 *meV*, which can be attributed to the  $\Gamma_5 \rightarrow \Gamma_3$  and  $\Gamma_5 \rightarrow \Gamma_4$  transitions respectively. No more excitations have been observed up to  $\sim 700$  *meV*. This allowed the determination of the two coefficients in the CF hamiltonian, namely  $V_4 = -123$  *meV* and  $V_6 = 26.5$  *meV* [1,2]. It has been shown also that *J*-mixing effects are far less important than previously assumed. Below the Néel temperature, more structure appears in the

neutron spectra. Detailed CF calculations have been performed [2] assuming either a 2-k magnetic structure and monoclinic lattice distortion or a 3-k magnetic structure and lattice distortion. The comparison with the experimental splittings is in favour of the second case.

The second compound we have examined is *UOS*, which belongs to the tetragonal *PbFCl* crystal structure and orders antiferromagnetically below  $T_N = 55K$ . Two main excitation peaks have been found at 74 and 82 meV for  $T > T_N$ . A redistribution of the intensities together with a slight shift of the peak positions is observed below  $T_N$ . No other CF excitations appear up to at least 200 meV [3]. The spectra have been interpreted [3,4] in the frame of a CF model accounting for the Schottky contribution to the specific heat, from which the splitting of the  $J = 4$  manifold into two groups of states with a separation not greater than 100 meV was expected. The peaks in the neutron spectra have been attributed to the transitions from the ground  $\Gamma_{t5}^{(1)}$  doublet to the second group of states. The intensity variation and the level shifts have also been interpreted as an effect of the molecular field.

Finally, a neutron spectroscopy study has been performed on *NpO<sub>2</sub>*, which is isostructural to *UO<sub>2</sub>* and contains *Np<sup>4+</sup>* ions with  $5f^3$  electronic configuration. The tenfold degeneracy of the  $J = 9/2$  ground multiplet is split by the cubic CF into two quartets and one doublet. This compound presents a phase transition at  $T_c = 25K$  whose origin is as yet unexplained. We have examined a sample of 32g of powder, together with a similar *ThO<sub>2</sub>* sample, allowing us to perform a reliable phonon density of state subtraction. Although no sharp peaks have been found up to 350 meV, magnetic scattering is clearly visible from our spectra around  $\sim 60$  meV. The difference of *NpO<sub>2</sub>* and *ThO<sub>2</sub>* spectra shows in fact a broad peak centered at this energy value, having  $\sim 20$  meV FWHM. No significant changes have been observed in passing through the transition temperature. CF calculations starting from the CF parameters determined for *UO<sub>2</sub>* predict for *NpO<sub>2</sub>* a strong  $\Gamma_8^{(2)} \rightarrow \Gamma_8^{(1)}$  transition at  $\sim 55$  meV. We believe to have observed this CF excitation, the broadening of the peak being probably due to interaction with optical phonons at the same energy.

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