

**Experiment title:**Dipolar v. Quadropolar order in $U_{0.75}Np_{0.25}O_2$ **Experiment number:**

HE-1386

Beamline:

ID20

Date of experiment:

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Date of report:

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Shifts:

18

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We have successfully used element specific, resonant X-ray scattering to observe both dipole and quadrupole ordering in a single crystal sample of $U_{0.75}Np_{0.25}O_2$. The parent compound UO_2 orders antiferromagnetically at 30 K with the type-I structure. As a result of RXS measurements at ID20, pure NpO_2 is known to exhibit quadrupole ordering at 25 K with the same wavevector as the dipole ordering in UO_2 . Early experiments (Fig. 1 taken in 2000) on single crystals of the mixed system $U_{0.75}Np_{0.25}O_2$ showed signals at the M edges of both elements, but the lack of polarisation analysis, which was then not available, prevented any quantitative analysis.

By using such a method it is possible to distinguish between dipole (magnetic) and quadrupole (electric) scattering due to their different polarisation states.

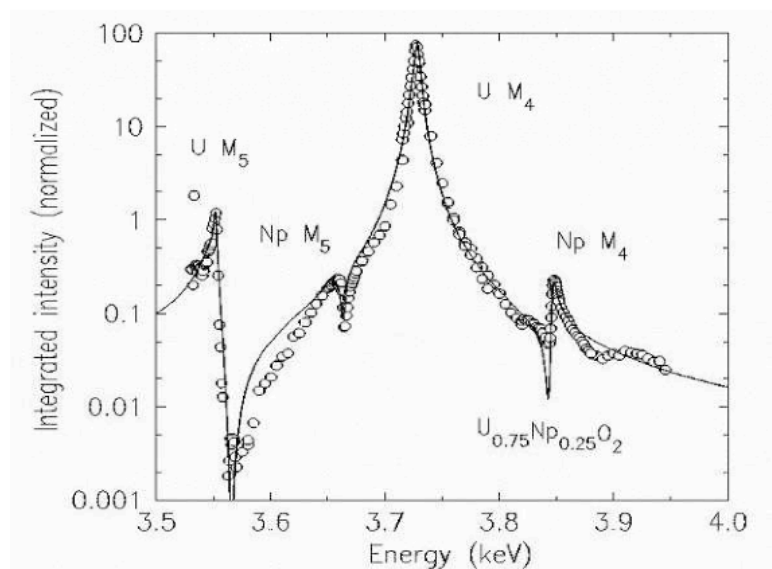


Figure 1 - Normalized integrated intensity as a function of energy for the (112) AF reflection of $U_{0.75}Np_{0.25}O_2$

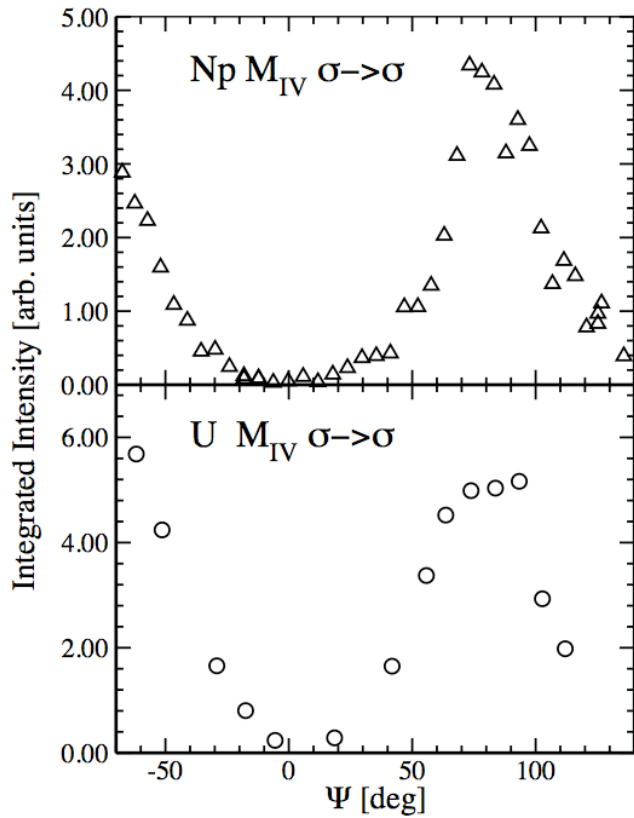


Figure 2 - Azimuthal dependence of the scattered beam in the unrotated sigma - sigma channel at both the U and Np M_{IV} edges.

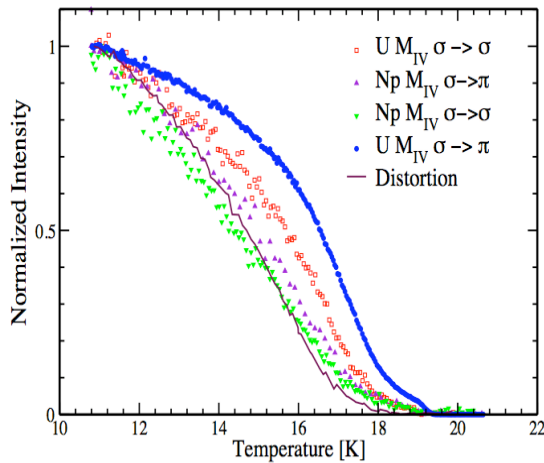


Figure 3 - Temperature dependence of the (112) AF reflection in both the rotated and unrotated polarization channels as a function of temperature. Also shown is the (421) internal lattice distortion measured at incident energy of 7.5 keV.

In the experiment presented here we have studied the azimuthal, energy, and temperature dependence of the scattering from the (112) superlattice reflection in the mixed oxide $U_{0.75}Np_{0.25}O_2$. We find that at a temperature of 10 K there exists quadrupole and dipole order on *both* the U and Np ions, displayed by strong scattering in the unrotated channel ($\square - \square$). The results of azimuthal scans are shown in Figure 2. Here we can see that the quadrupolar order on both the uranium and neptunium ions exists with the symmetry-breaking vector along the same direction. By analysis of the relative intensities of the unrotated ($\square - \square$) and rotated ($\square - \square$) channels at the neptunium and uranium edges it appears that on the uranium ions the dipole order dominates whereas on the neptunium the ratio of the dipole to quadrupole long-range order is much smaller. It should be noted that *no* quadrupole ordering has been found in pure UO_2 .

In addition, the temperature dependence of the scattered intensity was measured at both the uranium and neptunium edges in both polarization channels, along with the dependence of the (421) internal lattice distortion (Fig. 3). Initially, on cooling, the U dipole moment orders first. This is then followed at about 1.5 K lower temperature by the ordering of the U quadrupoles and both signals at the Np sites. At the same time the internal lattice distortion occurs.

From these data it appears that the U magnetic order dominates and as the temperature is lowered a long-range ordered Jahn-Teller distortion develops along with electric-quadrupolar order on both the uranium and neptunium. In addition the neptunium becomes magnetic. Such results differ from both parent compounds as NpO_2 does not display a dipole moment, and no quadrupolar order exists in UO_2 .