

ESRF

**Experiment title:**

Resonant x-ray magnetic scattering from Np and U mixed oxide compounds (LTP)

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HE-818

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**Local contact(s):**

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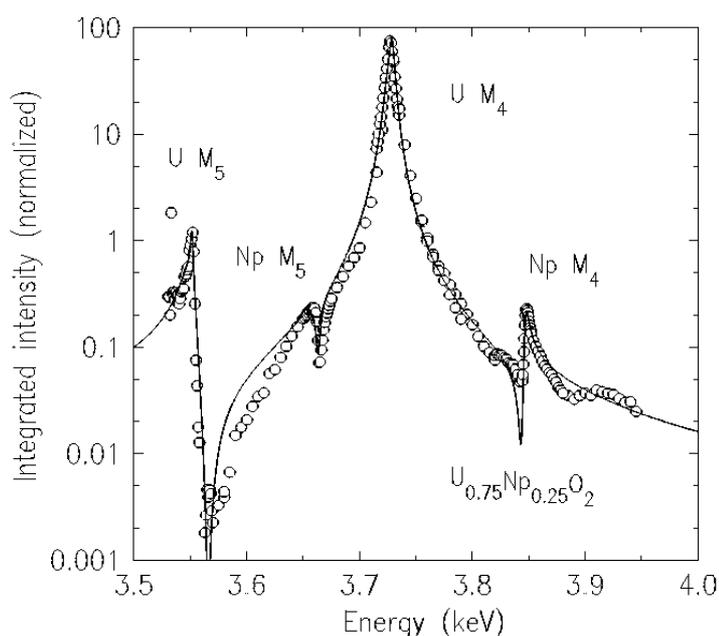
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We have used element specific resonant magnetic scattering to investigate the M edge resonances of U and Np in single crystals of  $(U_xNp_{1-x})O_2$ . Pure  $UO_2$  is a type-I antiferromagnet.  $NpO_2$  has a very small magnetic moment and does not exhibit hyperfine splitting of the ground state. However, strong resonantly enhanced scattering has previously been observed at the Np  $M_4$  edge of  $NpO_2$ , which exhibited a much sharper than expected resonant line-shape that could not be modeled to a standard dipole oscillator [Mannix et al. RRB 60 (1999) 15187]. One of the aims of this present experiment was therefore to examine the line shapes of the resonant scattering from the  $(U_xNp_{1-x})O_2$  mixed oxides. The data in **figure 1** show the response of the resonant magnetic scattering as the photon energy is increased across the U and Np  $M_5$  and  $M_4$  absorption edges of  $(U_{0.75}Np_{0.25})O_2$ . We have modeled these resonances to 4 dipole oscillators that interfere with each other. The fit to these data includes a preliminary sample absorption correction of  $1/\mu$ , which takes into account the white-lines estimated from absorption measurements of  $UO_2$ .

**Figure 1**

Normalized integrated intensity as a function of energy for the (1 1 2) AF reflection of  $(Np_{0.75}U_{0.25})O_2$ . At a temperature of 10K. The solid line is a simulation of 4 dipole oscillators (Lorentzian functions) corrected for sample self absorption and the energy dependence of the beam attenuation.



Another aspect of this experiment is that in using this element specific technique we can relate the respective amplitudes of the M edge resonances to obtain a ratio of the uranium to neptunium magnetic moments. By comparing the relative amplitudes of the signals after normalizing them by the atom concentrations our results suggest a Np/U moment ratio smaller than found by a combination of neutron and Mössbauer studies. Following theoretical crystal-field calculations we will compare the relative strengths of these mixed oxide M edge resonances with those of the previously studied  $(U_{1-x}Np_x)Ru_2Si_2$  alloys [Lidstrom et al. PRB 61 (2000) 1375].

The temperature dependencies of the normalized integrated intensities of the (1 1 2) magnetic peak measured at the U and Np  $M_4$  resonances are shown in **figure 2**. On lowering the temperature the Np moments order at a slower rate than the U atoms. The ordering parameters for U and Np are 0.32 and 0.42, respectively, obtained from fits to the  $I=I_0(1-T/T_N)^{2\beta}$  power law near to  $T_N$ . This appears to be consistent with a picture of the Np moments being "pulled-up" by the molecular field of the U atoms. However, unexpectedly the U and Np sublattices appear to have different ordering temperatures ( $T_N=17.5$  K for U and 16.8K for the Np sublattice) and so it is difficult to model this system using a simple molecular field model [Huppeld et al. EPL 49 (2000) 92].

The temperature dependence of the (4 2 1) forbidden reflection is also shown in Fig. 2. This charge scattering arises due an internal distortion of the oxygen sublattice induced by the magnetic ordering, as previously observed for pure  $UO_2$ . Interestingly, it follows rather closely the temperature dependence of the Np magnetic scattering.

### Figure 2

The normalized ( $T=10K$ ) integrated intensity as a function of temperature for the (1 1 2) magnetic reflection measured at the U and Np  $M_4$  edges (open and closed symbols, respectively) and the (4 2 1) internal lattice modulation (+ symbols), which was measured close to 7.5 keV. The solid lines are fits to a power law as described in the text.

