ESRF	**Experiment title:** **Quadrupolar ordering in NpO_2**	**Experiment number**: HE-818
Beamline: ID20	**Date of experiment**: from: 6 Feb 2002 to: 14 Feb 2002	**Date of report**: 19 Feb 2002
Shifts: 18	**Local contact(s)**: Matt Longfield	*Received at ESRF:*

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Report:

A previous x-ray experiment on NpO_2 at ID20 showed that extra resonant diffraction peaks at the $M_{4,5}$ edges appeared in NpO_2 when the crystal was cooled below the "transition" of 25K [1]. This transition, first discovered in specific heat experiments in 1953, has remained a mystery since that time, despite extensive experiments with many techniques, including neutron diffraction. Recently, a theory paper [2] has suggested that the ordering involves octupole moments.

This experiment at ID20 involved a new mounting of a small (<1 mg) single crystal of NpO_2 so that azimuthal scans could be taken as a function of the rotation about the scattering vector. Polarization analysis was also performed. The initial polarization was σ, with the rotated channel being π. We confirmed the presence of these extra peaks and showed that they have a contribution in both the σ-σ and σ-π channels, where the incident polarization is σ and the final polarization may be σ or π. These polarization states exclude the first order term of purely magnetic dipole scattering that gives scattering only in the σ-π channel. The azimuthal dependence of the scattering may be conveniently shown as an asymmetry ratio (Fig. 1), and the solid line in the figure is the expected dependence for an ordered quadrupole moment. This shows that the ordering at 25K in NpO_2 could be of a quadrupole type and that there appears to be no measurable magnetic dipole contribution. Since neutron diffraction is insensitive to electric multipole order, this explains why all neutron experiments on this material have failed to find a magnetic signal.

In the previous x-ray experiment this resonance was observed to have a narrow energy width with a Lorentzian squared profile. We have confirmed that this exists for both polarization channels; the energy dependence of the σ-π scattering is shown in Fig.2. (Both channels also have the same temperature dependence - as expected.) If, as we believe, the origin of the signal in NpO_2 is quadrupole type ordering then it involves the $F^{(2)}$ resonant term. Consistent with this interpretation, experiments on uranium materials have seen similar Lorentzian squared energy profiles when probing the matrix elements involving the $F^{(2)}$ term [3].

[1] D. Mannix et al. Phys. Rev. B 60, 15187 (1999).

[2] P. Santini and G. Amoretti, Phys. Rev. Lett. 85, 2188 (2000).

[3] M. J. Longfield et al., sub. to Phys. Rev. B (Feb 2002).

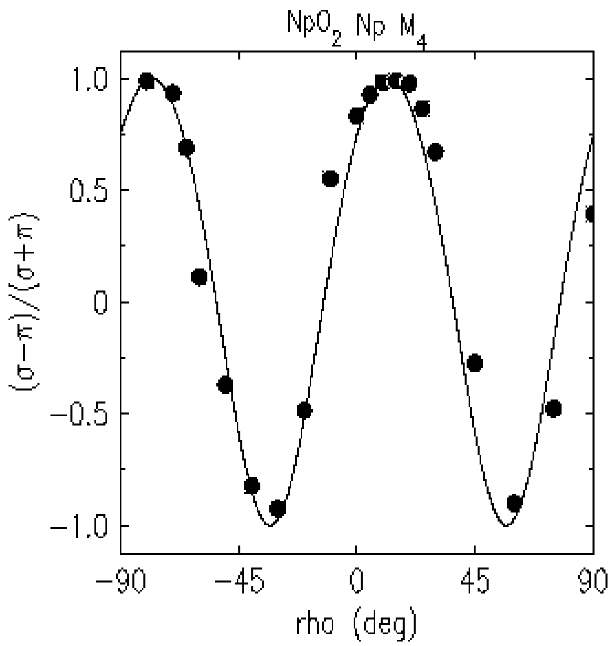

Figure 1: Azimuthal scattering from the (0 0 3) reflection in NpO_2 at the Np M_4 edge.

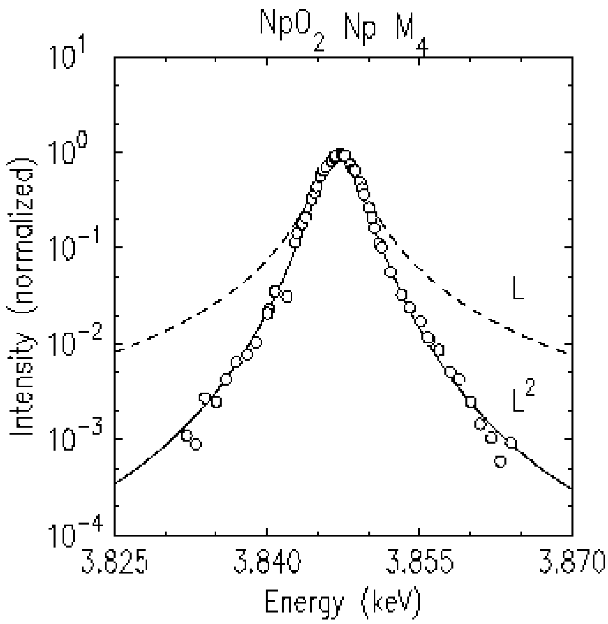

Figure 2: The energy dependence of the resonant scattering from the (0 0 3) reflection at the Np M_4 edge. Dashed line is Lorentzian and solid line is Lorentzian squared.