ESRF	**Experiment title:** Search for antiferromagnetism in NpO_2	Experiment number: *HE-302*
Beamline: ID20	**Date of experiment:** from: 4/6/98 to: 9/6/98	**Date of report:** 25/8/98
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Report:

The nature of the magnetic ordering in NpO_2 has remained an enigma since the specific heat and Mössbauer measurements were performed on pure material over 30 years ago. The specific heat shows a peak at 25 K, and the Mössbauer spectra show a small line broadening at this temperature, which was initially associated with a hyperfine field arising from a magnetic ordering of moments at the level of ~0.1μ_B. Later Mössbauer experiments were interpreted as arising from relaxation phenomena [1]. Many neutron experiments have tried unsuccessfully to determine the nature of the 25 K phase transition, but this technique has been hampered by the fact that crystals larger than ~ 1 mm^3 do not exist. From polycrystalline samples any antiferromagnetic (AF) moment is known to be < 0.25 μ_B. Using polycrystalline samples, such experiments have seen a change in the inelastic response function at 25 K [2]. However, no microscopic evidence for a magnetic phase transition was available until 1996 when a muon experiment at PSI showed the presence of a back-forward asymmetry that can only be associated with AF ordering [3].

Within the framework of our programme on small (≤ 1 mg) transuranium samples we examined a single crystal of NpO_2 at ID20 by searching reciprocal space with photons tuned to the M_4 resonance of Np. The crystal mass was 0.7 mg, and we used a photon beam of 0.2 x 0.2 mm^2. This resulted in finding a very strong (38,000 cts/s) peak at the (001) position, indicating that the magnetic order has the same wavevector as UO_2. By measuring the (003) intensity as well, we were able to show that the moment directions are ∥ **q**, unlike in UO_2 where they are ⊥**q**. Fig. 1 shows the T-dependence of the magnetic reflections giving a T_N ~ 25 K, as expected. Lattice effects were also examined and a gradual contraction of the unit cell is observed below ~ 30 K, which corresponds to a change in the lattice parameter of $|\Delta a/a| \sim 10^{-4}$. A brief search for an internal distortion, as found in UO_2 [x], was not successful, but using an analyser may be necessary at higher energies to reduce the background from fluorescence.

Figure 1

T-dependence of the (001) and (003) magnetic reflections in NpO_2.

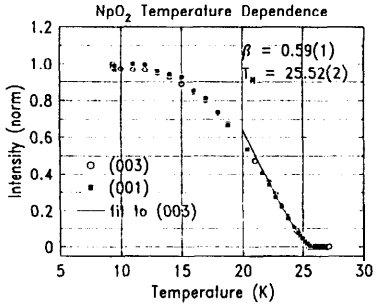
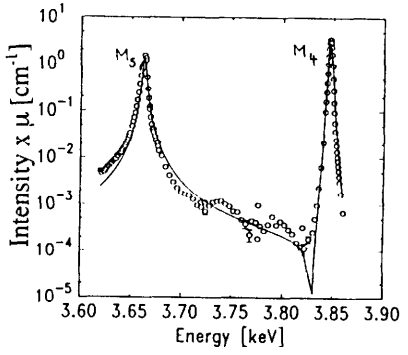


Figure 2

The intensity as a function of energy in NpO_2. The M_4 and M_5 resonances are marked. The solid line is a fit with a Lorentzian for the M_5 and a Lorentzian-squared to represent the M_4.



Although the discovery of the AF order in NpO_2 is a major achievement (unfortunately we cannot, yet, relate the intensity of the resonant scattering to the value of the magnetic moment), there is an even more interesting observation from the NpO_2 experiment, and this is shown in Fig. 2. Normally these resonant signals fit reasonably well with interfering **Lorentzians** (as expected from the atomic physics of the resonant process). However, *for the first time in an actinide material*, we have observed completely different forms for the two resonances. The extraordinary good fit of the M_4 to a Lorentzian-squared function suggests some fundamental physics might be associated with this, although we are presently unable to say what! Even more unusual is that the forms for the M_4 and M_5 are different.

This emphasises that the much improved energy resolution of ID20 is giving new information in energy space. The fact that we don't know (at the moment) what it means is unfortunate, but exciting!

References:

[1] J. M. Friedt et al., Phys. Rev. B 32, 257 (1985) - all earlier references are quoted in this paper
[2] G. Amoretti et al., J. Phys. Cond. Matter, 4, 3459 (1992); also references therein
[3] W. Kopmann et al., J. Alloys and Compounds 271–273, 463 (1998)
[4] J. Faber and G. H. Lander, Phys. Rev. B 14, 1151 (1976)