ESRF	**Experiment title:** Phonon dispersion, magneto-vibrational coupling and multipolar excitations in NpO_2.	**Experiment number**: HC-1929
Beamline: ID28	**Date of experiment**: from: 22 apr. 2015 to:28 apr. 2015	**Date of report**: 07/09/2015
Shifts: 18	**Local contact(s)**: L. Paolasini	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

R. Caciuffo & N. Magnani, European Commission, JRC-ITU Karlsruhe, Germany

G.H. Lander, ILL, France

T. Forrest, ESRF, France

Report:

The experiment was carried out on a high-quality single crystal of NpO_2 using ID28 with an incident energy E=17.794 keV, corresponding to the Si(999) monochromator and multilayer setup as in the previous experiment HC1122. The sample of dimension of 0.7×0.6×0.1 mm^3 was oriented with the specular direction along the (100) crystal axis and the (011) axis in the scattering plane, and then incapsulated between two diamond slabs of 0.5×5×5 mm^3 at the Institut of Transuranium Elements in Karlsruhe.

During the first part of the experiment we have completed the dispersion curves at room temperature, checking for the missed optic branches (see Fig. 1).

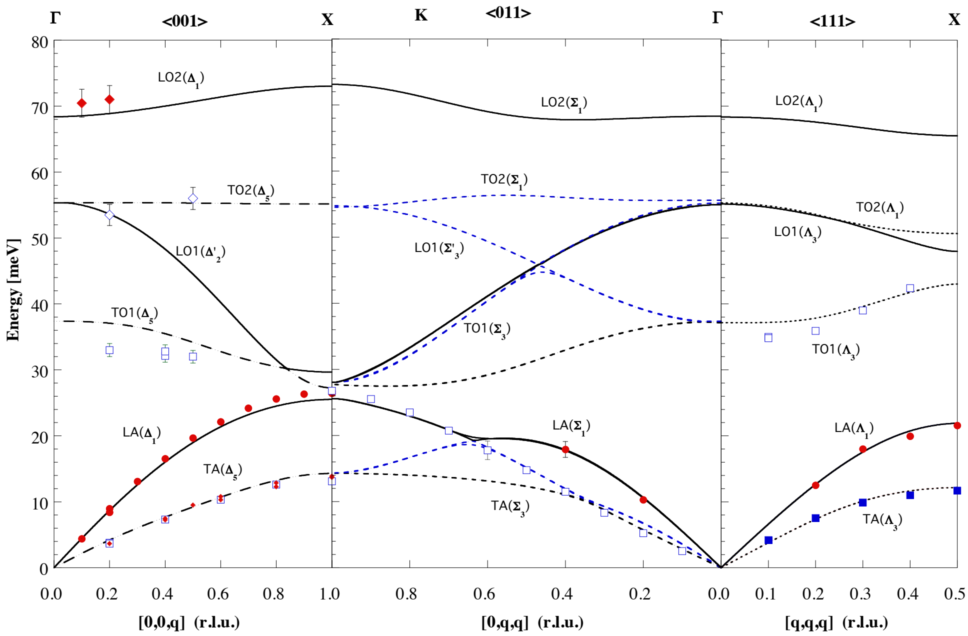

Fig.1 : Dispersion curves of NpO_2 measured at room temperature on ID28.

The second part of the experiment was performed at low temperature (10 K) searching for phonon anomalies in the acoustic region below 15 meV. Indeed, according to the theory (see the main text of the proposal) in

NpO_2 the magnetic dipoles are quenched in the ordered phase, and the primary order parameter is the rank-5 (triakontadipole) magnetic multipole. This exotic magnetic order is accompanied by an order of electric quadrupoles that are arranged in a type-I, 3-k longitudinal structure requiring neither internal nor external lattice distortions (the symmetry remains cubic and the positions of the oxygen atoms do not change). The ground-state quartet of the Np ions is split in the ordered phase, with a singlet-doublet-singlet sequence. The singlet-doublet and singlet-singlet single-ion transitions give rise to dispersive multipolar excitation branches centred around 6 and 13 meV. Similarly to what observed by inelastic neutron scattering in UO_2, it is expected that a strong interaction between magnetic and vibrational modes occurs through the mediation of electric quadrupole fluctuations. This will affect the phonon acoustic modes centered around 13 meV and at 6 meV, as shown in Fig.2 of the proposal HC-1929.
We have measured the LA (6-H,00), TA (6,K,0) and TA (5,-1,L) phonon branches at 10K, as shown in Fig.2. Whereas the LA phonon intensities decreases smoothly from the zone center to the zone boundary, as expected from the first-principle phonon calculations (that provide an excellent match to the measured dispersion curves), the two measured TA branches present an anomaly at the reduced wavevector q=0.8, reflected in a drastic loss of intensity of the corresponding phonon group, as shown in the lower panel of Fig. 2 for the TA(6,K,0) branch.
This wavevector corresponds to an energy of about 13 meV, exactely where the theoretical prediction assumes the existence of dispersive mean-field magnetic modes.
Due to the lack of time, we were unable to measure the temperature dependence of this anomaly. We plan to submit a new proposal to investigate this interesting new behaviour.

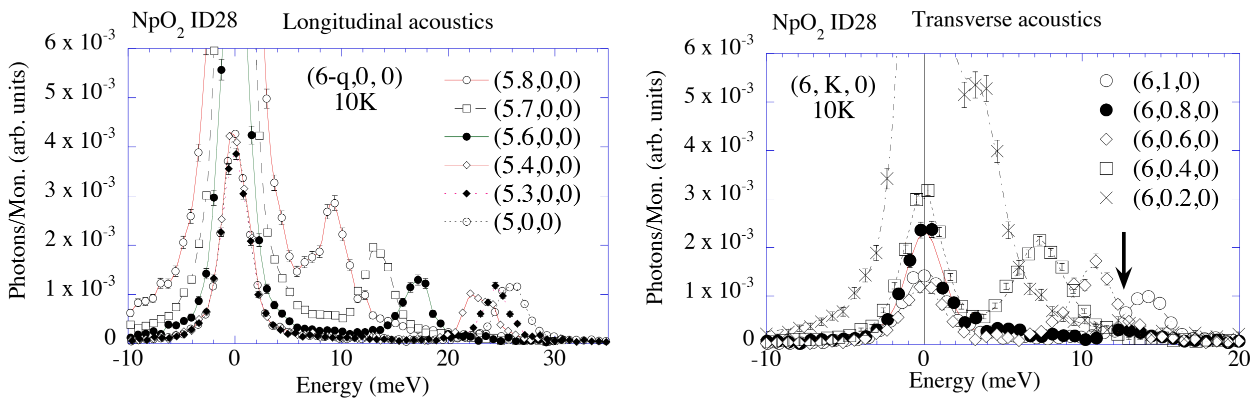

Fig.2 : Upper panel: LA (6-H,0,0) phonon dispersions of NpO_2 measured at 10 K. Lower panel: TA (6,K,0) branch showing a drastic decrease of the intensity of the phonon group at q=0.8 rlu (black dots). The arrow indicates the position of the TA phonon anomaly. Notice also that the (610) is a Brillouin zone boundary.