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| ESRF | **Experiment title:**<br>Phonon dispersion, magneto-vibrational coupling and multipolar excitations in NpO2 | **Experiment number**:<br>HC-1122 |
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| **Shifts:** | **Local contact(s)**:<br>T. Forrest | *Received at ESRF:* |


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## 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) + multilayer setups. This energy is just above the Np L3 edge energy $E_{L3}$=17.61, but below both the L1 and L2 edges ($E_{L1}$=22.427, $E_{L2}$=21.601). As a result, a severe sample photo-absorption is present, and the use of transmission scattering geometry must be avoided. This energy represent also a good compromise between the energy resolution and the optimization of the photon flux. A focusing geometry was choosen in order to reduce the beam path on the sample surface, by using a multilayer focusing configuration with a beam spot size of 30x80 $\mu m^2$ on the sample surface.

The sample of dimension of 0.7x0.6x0.1 $mm^3$ 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 0.5x5x5 $mm^3$ at the Institut of Transuranium Elements in Karlsruhe. This experiment was conducted at room temperature. The diamond slabs where oriented with the (110) axis closely parallel to the (100) crystal axis, and the diamond phonon contamination was detected mainly around the (400) and (300) $NpO_2$ Brillouin zones (BZs). These diamond phonon groups (mainly acoustic) could be easily distinguished from the $NpO_2$ because the dispersions are very steep. However, due to the weakness of the optic phonons of $NpO_2$, these contaminated Brillouin zones were avoided.

The acoustic phonons are easily detected in all the main crystallographic directions, and a complete collection of transverse and longitudinal acoustic phonon braches were determined in all the main crystallographic directions (see Fig.1).

We have investigated different BZs in order to optimize the inelastic structure factor for the different optic branches. Optic phonons arise mainly from oxigen vibration modes and are very weak. Fig. 2 show a transverse and a longitudinal scan around the (600) BZ associated to the reduced wavevectors (0,0.2,0) and (0.2,0,0), respectively. The peak intensity of the these optic phonon branches TO1 and LO2 is about two order of magnitude weaker with respect the acoustic branches, as shown in Fig.2.

Other optic branch were detected in other BZs, as shown in Fig.1, and, despite the lack of some other relevant optic branches (in particular the branch LO1-(100)), a complete picture of the lattice dynamics of $NpO_2$ was obtained.

A first-principle model calculation based on the projector augmented wave pseudopotential approach shows a remarquable agreement with the determined dispersions, and also some distinct differences with the previously determined phonon dispersion of $UO_2$ (Fig.3).
With this model we are now able to calculate the best BZ for any given optic branch and then to design a new experiment in order to determine at low temperature the anomalous behavior of the M5 optical phonon branch, as described in the proposal HC1122.

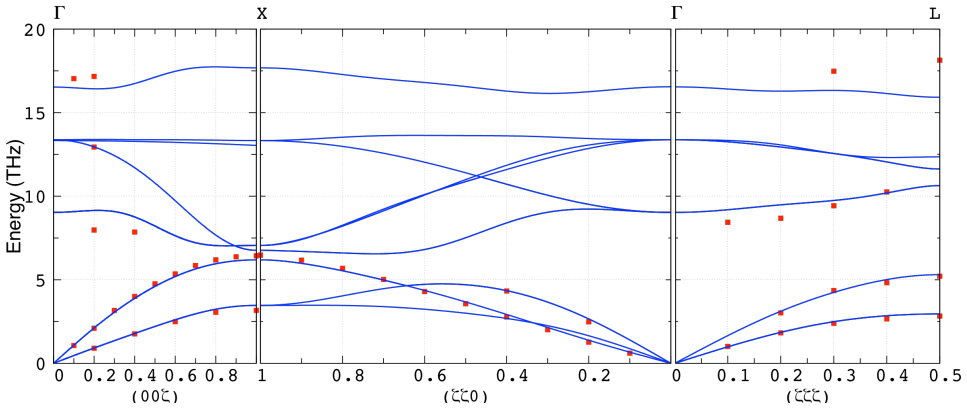


Fig.1 : Phonon dispersion curves of $NpO_2$. The red dots represent the experimental data and the continuous lines are the first-principle model calculation.

A) B)

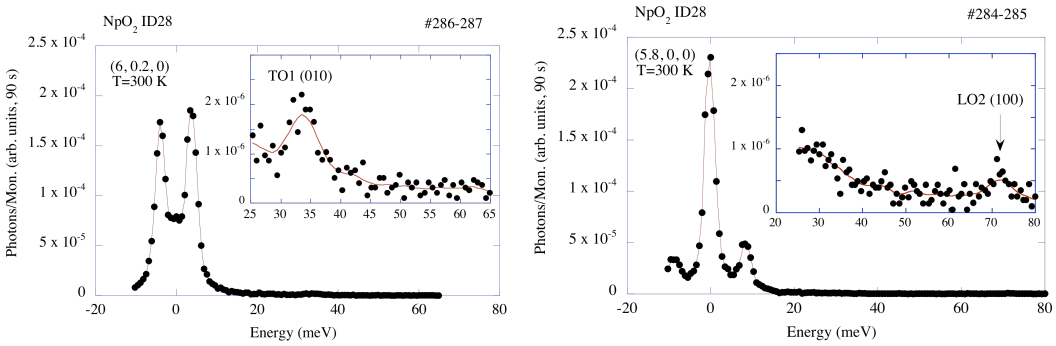


Fig.2 : Room temperature inelastic energy scans. A) Q=(6,0.2,0). The inset show the TO1 branch. B) Q=(5.8,0,0). The inset shows the LO2 branch. Data are normalised for the monitor.

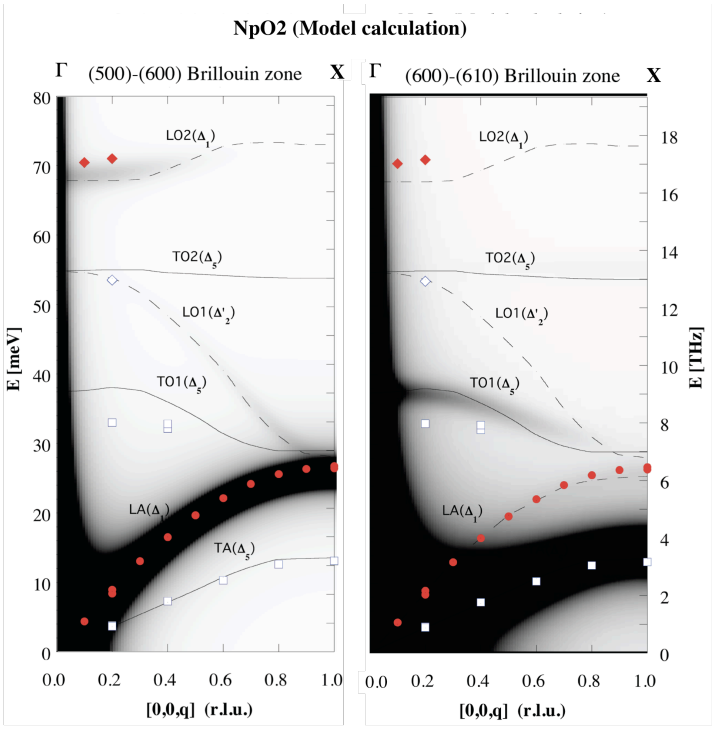


Fig.3: First principle model calculation of phonon dispersion curves (thin lines) and inelastic structure factors calculations (shaded area) for two wavevector directions around the BZ (006).