Status of the TSL activities in the framework of the NAUSICAA collaboration

DE LA RECHERCHE À L'INDUSTRIE

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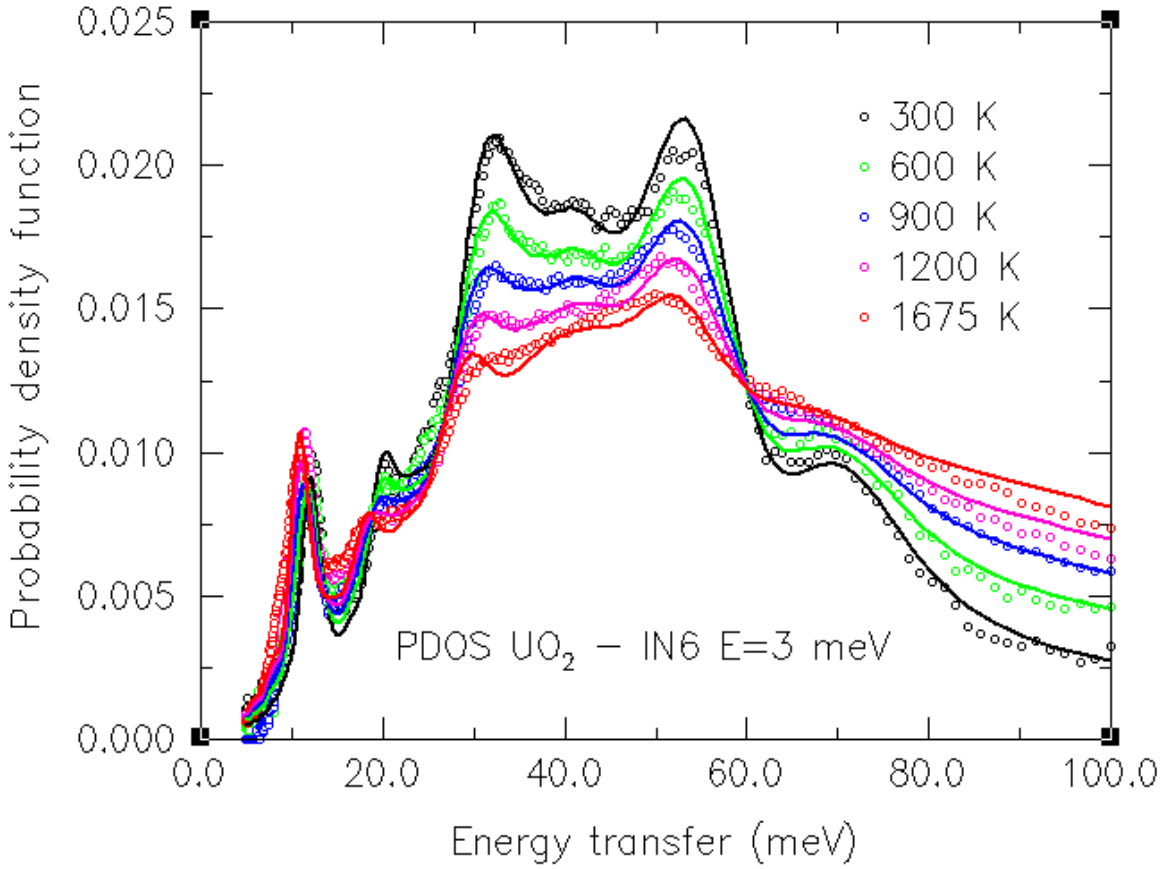
Outlines

- **Monte-Carlo analysis of neutron scattering measurements performed at ILL**
- **Inelastic neutron scattering experiments - ThO2 (IN6-SHARP)**
- **Diffraction experiments - ThO2 (D1B)**

Monte-Carlo analysis of neutron scattering measurements performed at ILL

- The **Monte-Carlo code TRIPOLI4** in association with the **CINEL code** (see Shuqi Xu presentation) is used to validate phonon density of states and diffusion model parameters involved in the calculations of the double-differential neutron scattering cross sections as a function of T.
- The adjustment of the model parameters and the propagation of the uncertainties are conducted by the **CONRAD code**



PDOS of UO2 extracted from INS data measured with the IN6-SHARP time-of-flight spectrometer of ILL

Monte-Carlo analysis of neutron scattering measurements performed at ILL

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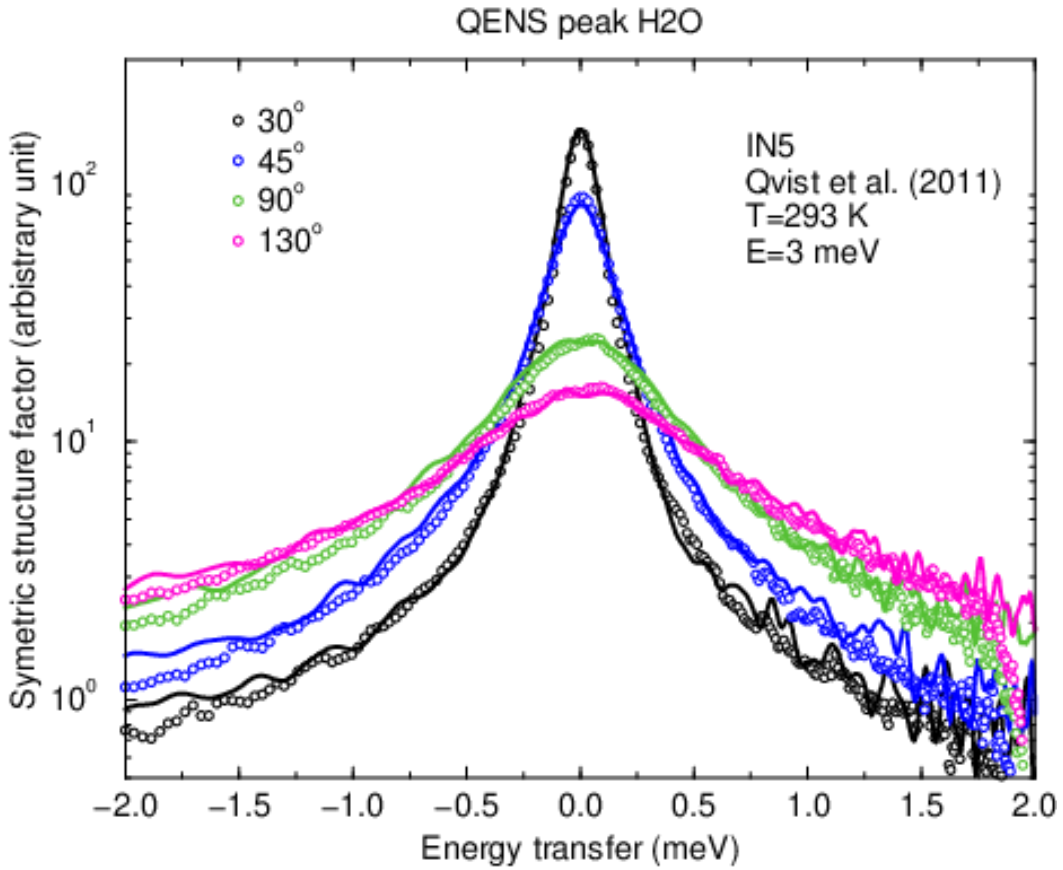
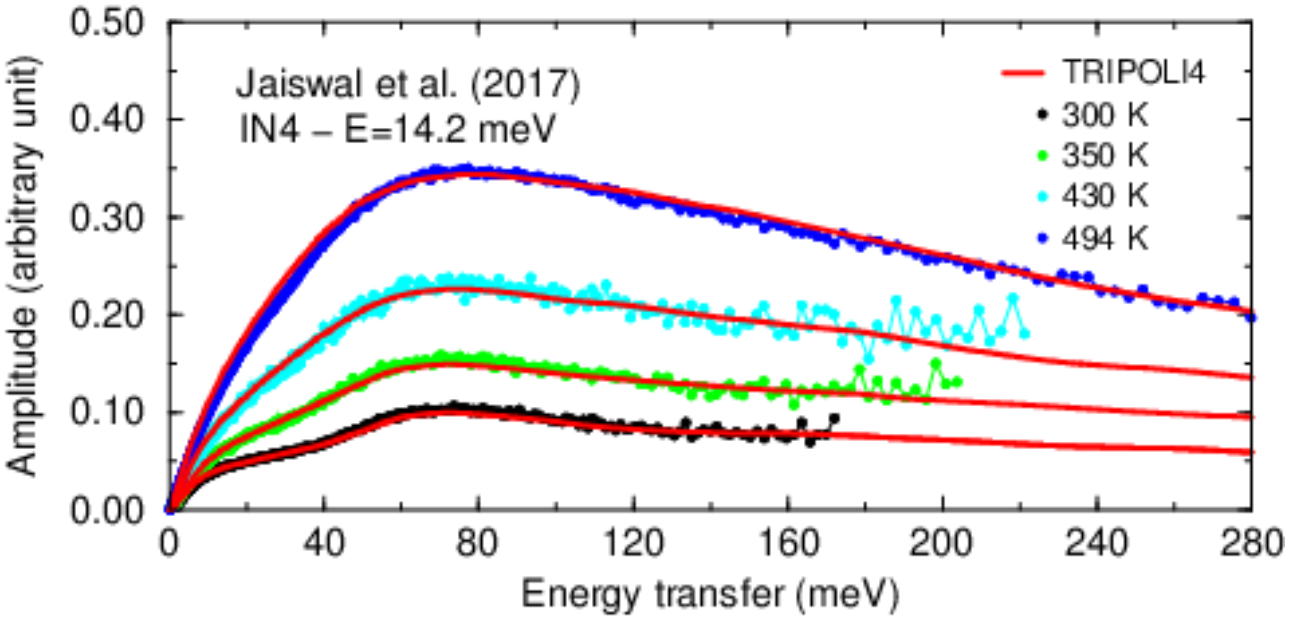
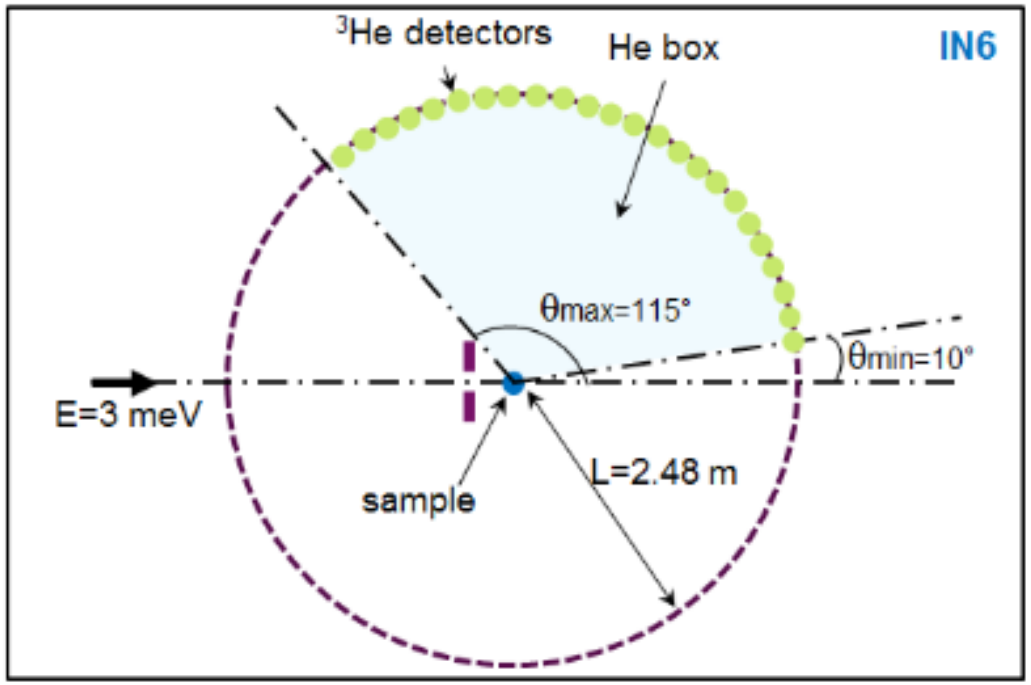


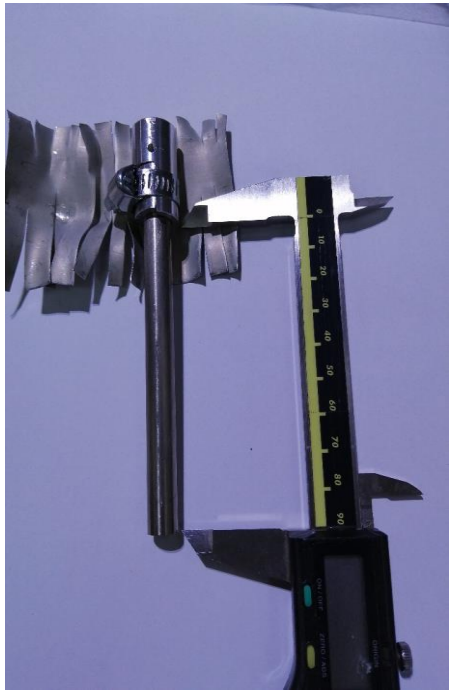
QENS and libration band of H2O measured with the IN5 and IN4 time-of-flight spectrometers of ILL

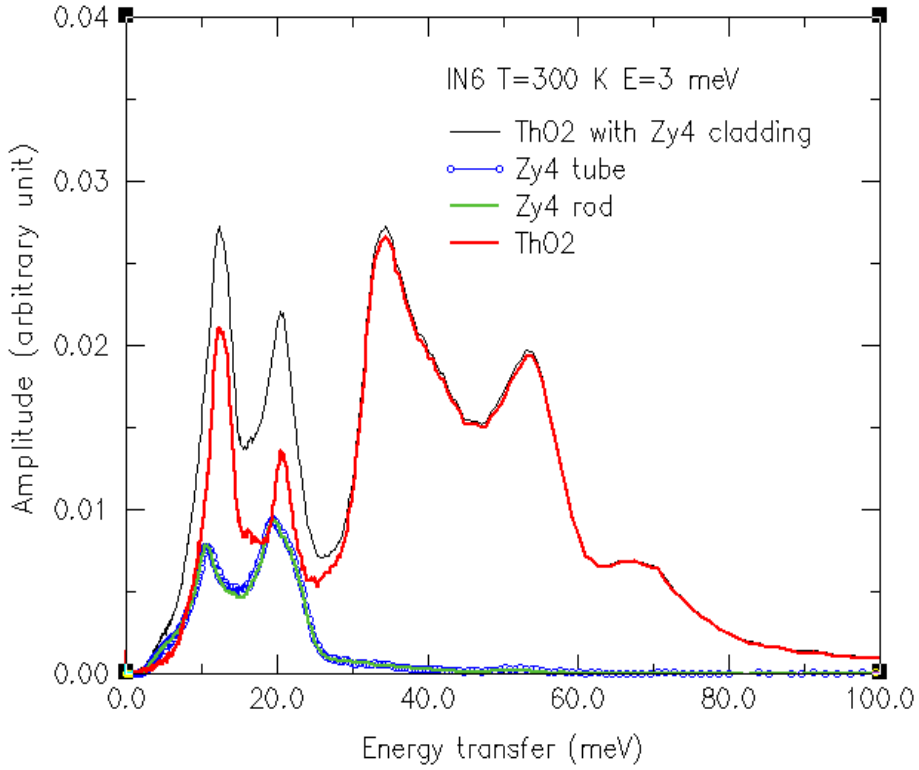


Inelastic neutron scattering experiments – THO2

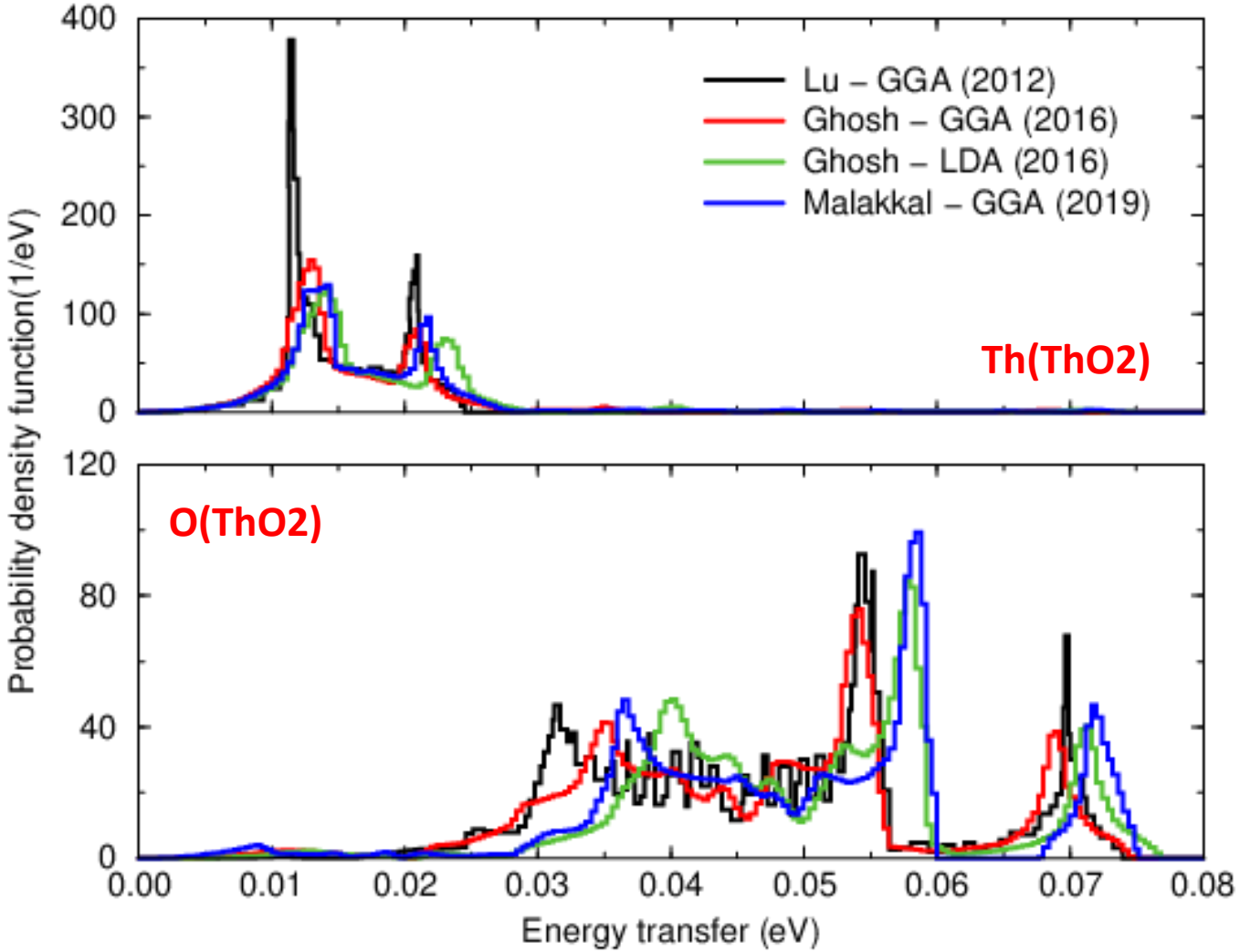
The experiments were performed on **IN6-SHARP** (λ=5.12 Å, E=3 meV) at 300 K. The ThO2 sample used for this experiment was composed of a stack of 12 ThO2 pellets in a double sealed ZrY4 container. The mass of the ThO2 sample was 45.23 g with a height and diameter (without ZrY4 container) equal to 9.31 cm and 8.16 mm, respectively. The thicknesses of the inner and outer ZrY4 containers were equal to 1.25 mm and 0.42 mm, respectively. A ZrY4 rod (Diameter of 1 cm) and a ZrY4 tube (outer diameter of 9.56 mm; thickness of 1.21 mm) were used for the container subtraction.







Inelastic neutron scattering experiments – THO2

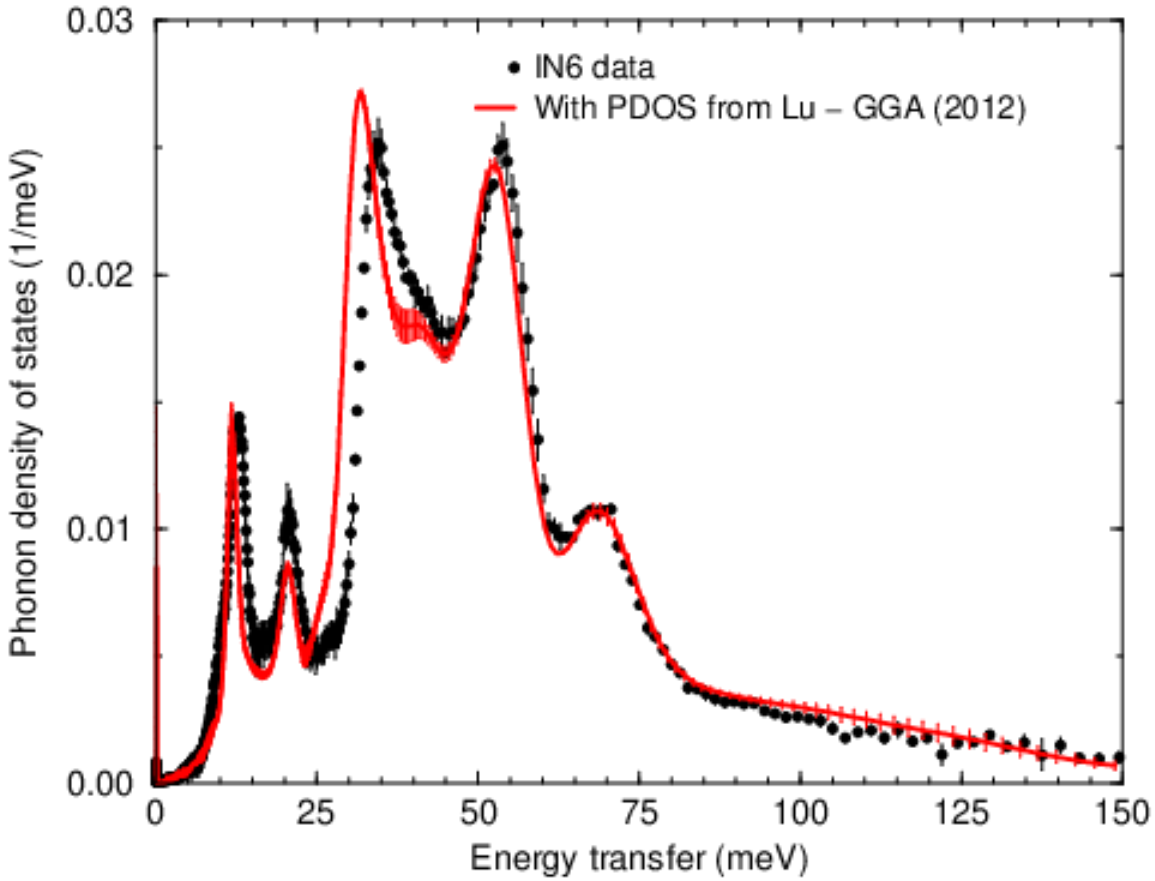


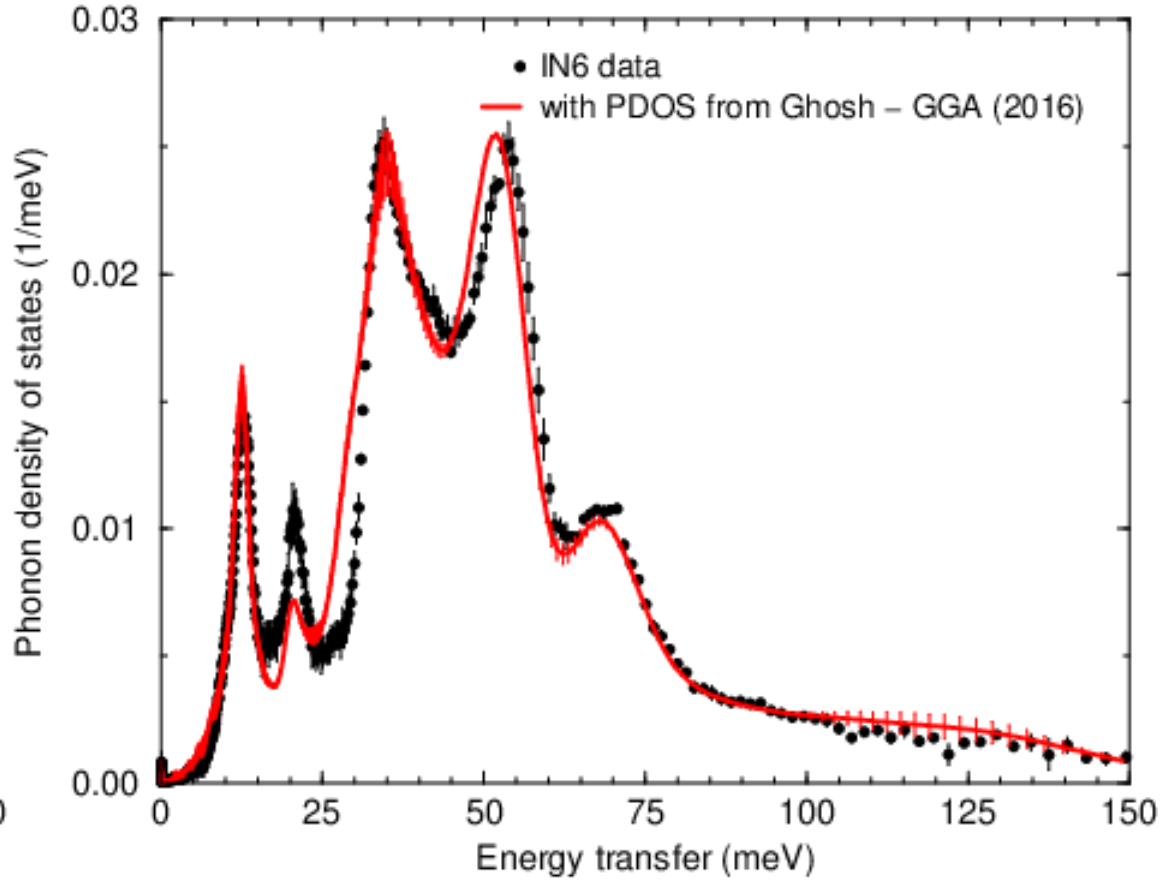
For the TRIPOLI simulations with have used PDOS reported in the literature

Large differences between the partial PDOS of O and Th in ThO2, depending of the codes (VASP, Quantum Expresso), computational options (ex. Generalized Gradient Approximation vs. Local Density Approximation) …

Inelastic neutron scattering experiments – THO2

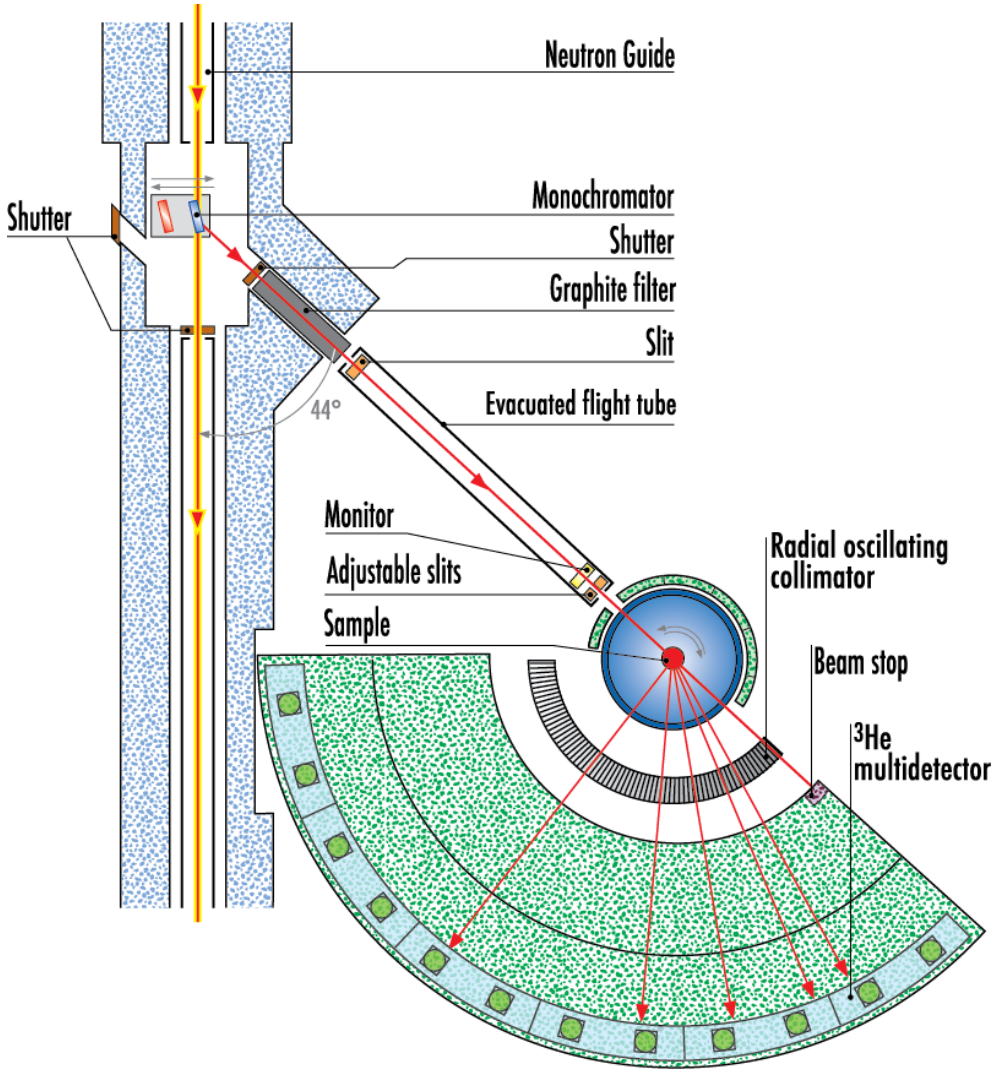
Encouraging results obtained with PDOS calculated with the GGA option → improvements of the PDOS with the CONRAD code in progress …

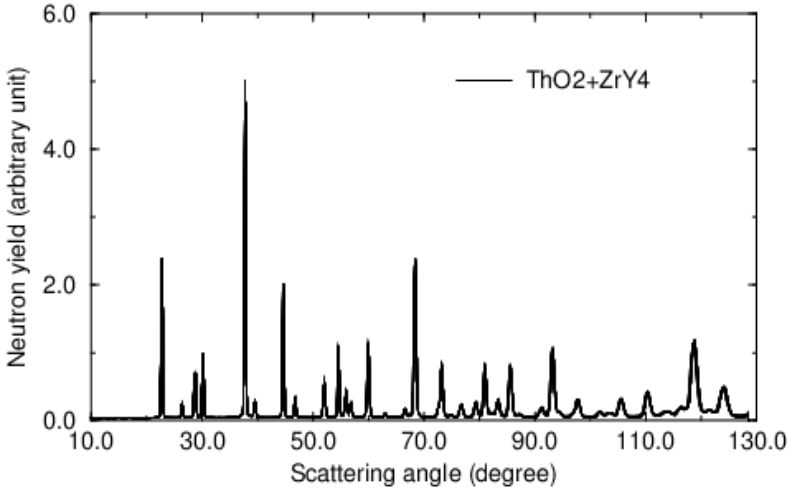


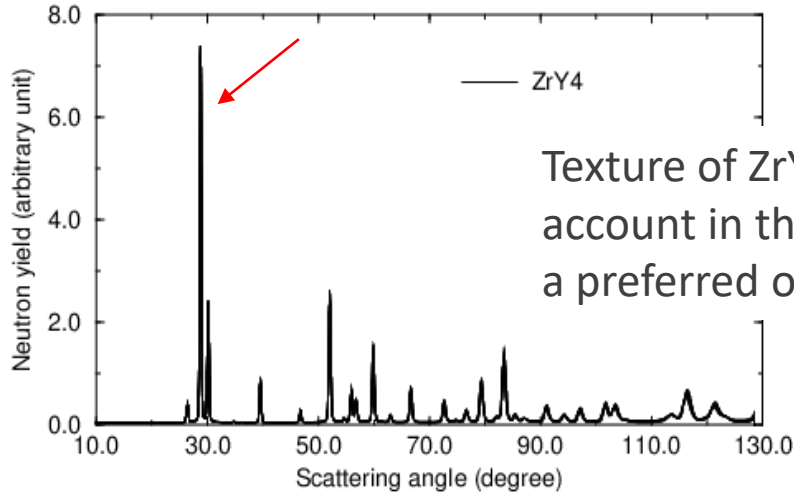


Diffraction experiments – THO2

Diffraction experiments performed on **D1B** (λ=1.28 Å, E=49.93 meV) from 2 K to 300 K with the same ThO2 sample



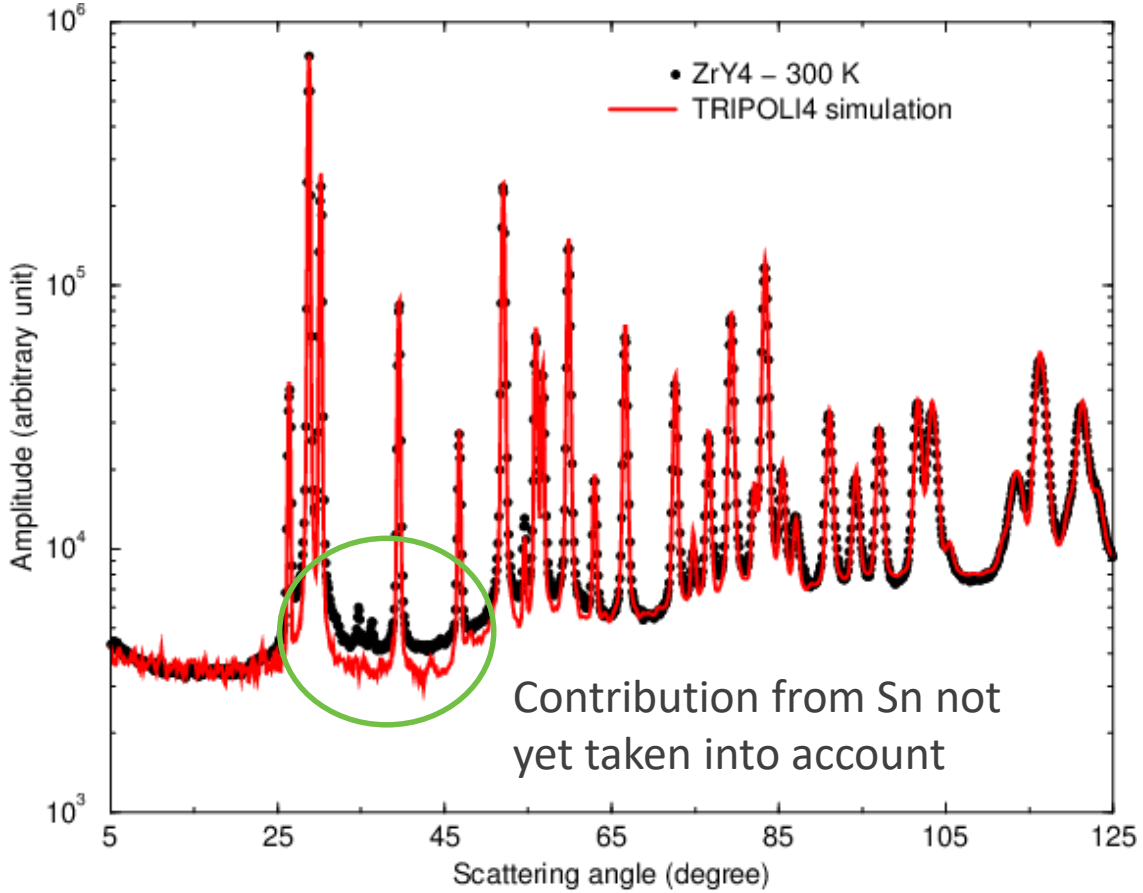


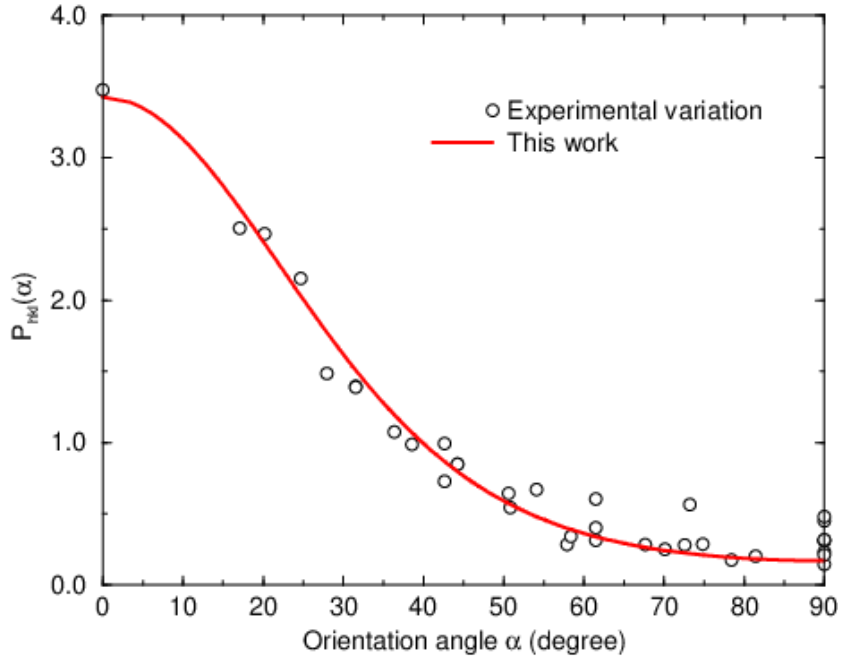


Texture of ZrY4 taken into account in the simulations with a preferred orientation (0,0,2)

Diffraction experiments – THO2

Comparison of the experimental and theoretical diffraction pattern (300 K) confirms the lattice parameters for Zr (P63mmc, group space 194) reported in the Crystallography Open Database (a=b=3.232 Å, c=5.147 Å)

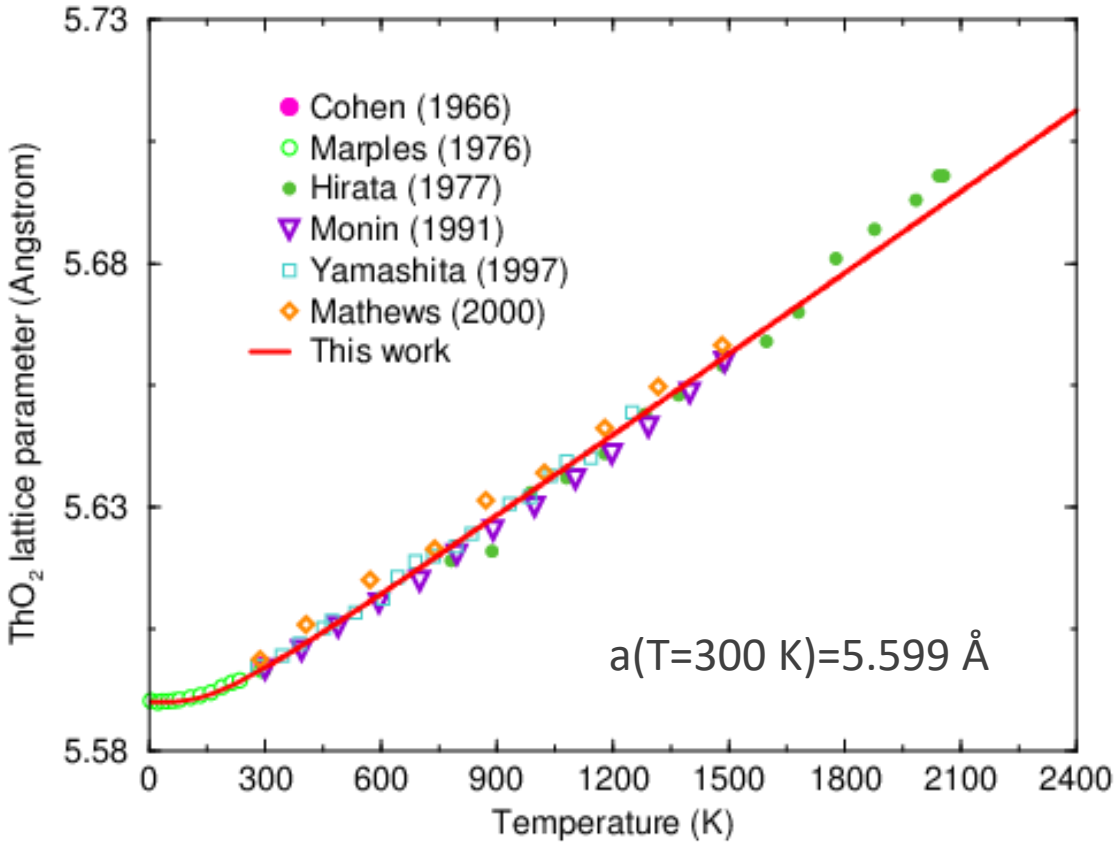




Texture correctly reproduced by the Cylindrical-symmetric pole density distribution function proposed by Altomare et al. (1994) that depend on a single free model parameter (orientation angle)

Diffraction experiments – THO2

Temperature dependence of the ThO2 lattice parameter (fm3m, group space 225) is deduced from data reported in the literature using a Debye model proposed by H. Kroncke et al. (Acta Physica Polonica 114, 1193, 2008)



$$U_{\mathrm{E}}(V,T) = 3(s-1)G^3\ \hbar\omega_0\ f_{\mathrm{E}}\left(\frac{\Theta_{\mathrm{E}}}{T}\right) \quad \text{with} \quad f_{\mathrm{E}}(x) = \frac{1}{\exp(x)-1}, \tag{2}$$

$$U_{\mathrm{D}}(V,T) = 3G^3\ k_{\mathrm{B}}\Theta_{\mathrm{D}}\ f_{\mathrm{D}}\left(\frac{\Theta_{\mathrm{D}}}{T}\right) \quad \text{with} \quad f_{\mathrm{D}}(x) = 3\int_0^1 \frac{t^3}{\exp(tx)-1}\,\mathrm{d}t. \tag{3}$$

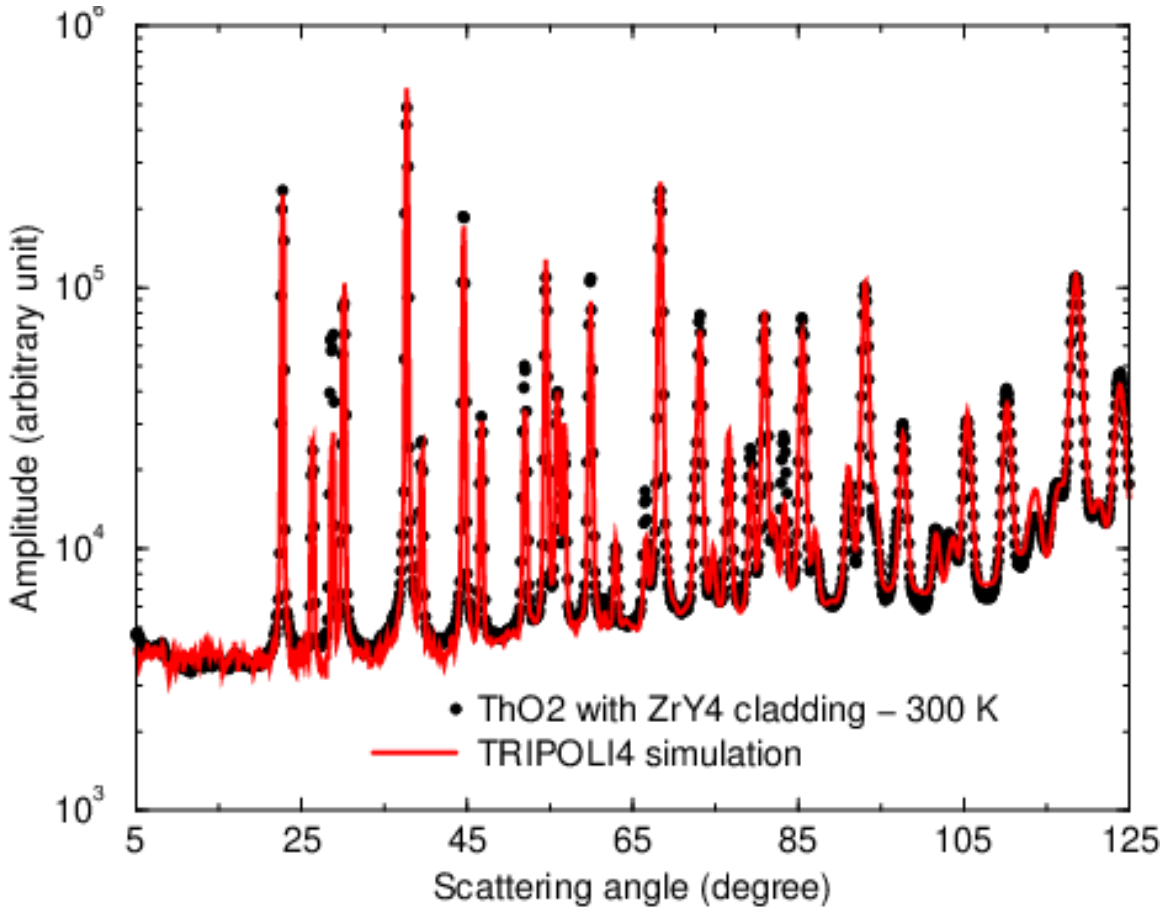
For the fitting of the measured data we identified the prefactors as macroscopic fit parameters, which leads to Eq. (4). Here a_0 is the lattice constant at 0 K, α is the high-temperature thermal expansion coefficient (TEC) and $\Theta_{\mathrm{E/D}}$ the characteristic Debye- or Einstein temperature

$$a(T) = a_0 + a_0\ \alpha\ \Theta_{\mathrm{E,D}}\ f_{\mathrm{E,D}}\left(\frac{\Theta}{T}\right). \tag{4}$$

- Debye temperature (626.5 K) deduced from the PDOS of Ghosh
- Lattice constant à 0 K is fixed to 5.59 Å
- Coefficient α is a free parameter

Diffraction experiments – THO2

Experimental diffraction pattern (300 K) compared to TRIPOLI4 simulations



- No background correction is introduced in the simulation
- Parameters of the angular response function has to be improved (Cagliotti formula) to better reproduce the amplitude of the diffraction structures
- Analysis of the data measured at low temperature is in progress…