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Generation of thermal scattering laws with the CINEL code

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Abstract. The thermal scattering laws (TSL) take into account the crystalline structure and atomic motions of isotopes bound in materials. This paper presents the CINEL code, which was developed to generate temperature-dependent TSL for solid, liquid and free gas materials of interest for nuclear reactors. CINEL is able to calculate TSL from the phonon density of states (PDOS) of materials under the Gaussian-Incoherent approximations. The PDOS can be obtained by using theoretical approaches (e.g., ab initio density functional theory and molecular dynamics) or experimental results. In this work, the PDOS presented in the ENDF/B-VIII.0 and NJOY-NCrystal libraries were used for numerical validation purposes. The CINEL results are in good agreement with those reported in these databases, even in the specific cases of TSL with the newly mixed elastic format. The coding flexibility offered by Python using the JupyterLab interface allowed to investigate limits of physical models reported in the literature, such as a four-site model for UO_2 , anharmonic behaviors of oxygen atoms bound in a Fm3m structure, texture in Zry4 samples and jump corrections in a roto-translational diffusion model for liquid water. The use of graphic processing units (GPU) is a necessity to perform calculations in a few minutes. The performances of the CINEL code is illustrated with the results obtained on actinide oxides having a Fm3m structure (UO_2 , ThO_2 , NpO_2 and PuO_2), low enriched fuel (UMo), cladding (Zry4) and moderators (H_2O with a specific emphasis on ice).

1 Introduction

Thermal Scattering Laws in the international libraries were all produced with the LEAPR module of NJOY [1]. However, the formalism in LEAPR is limited especially to account for the structure of the molecules. In addition, it is rather painful to accommodate new physics within the LEAPR formalism through its static input file. In our case, we encountered issues to analyse neutron scattering experiments for which some experimental corrections are needed, such as texture. It was also difficult to investigate the complexity of the dynamical structure factor for light water or to add an anharmonic correction to the nominal vibrations of the atoms. For these reasons, we started to develop the CINEL code to avoid using LEAPR in the generation of the TSL.

Models implemented in the CINEL code are presented in section 2. The interpretation of scattering experiments with CINEL is discussed in section 3. Section 4 highlights the needs of theoretical Phonon Density Of States (PDOS) calculated from ab initio simulations.

2 Presentation of the CINEL code

The CINEL code [2] is composed of three modules. The CUBIC module treats the coherent and incoherent neu-

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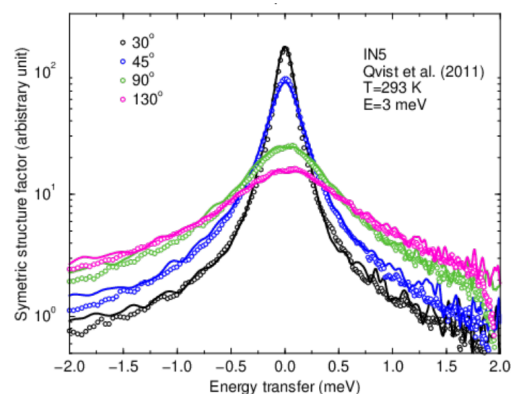


Figure 1. Experimental quasi-elastic neutron scattering (QENS) peaks for light water. The theoretical curves (solid lines) were obtained with the Monte-Carlo code TRIPOLI4.

tron scattering cross sections. The INELASTIC module calculates the inelastic scattering contribution. The third module, namely SVT, is devoted to study various Doppler models above the thermal cutoff energy. TSL provided by CINEL are stored in ENDF-6 format and tested thanks to the Monte-Carlo code TRIPOLI4 [3].

In the INELASTIC module, the theoretical description of the inelastic dynamic structure factor comes from the

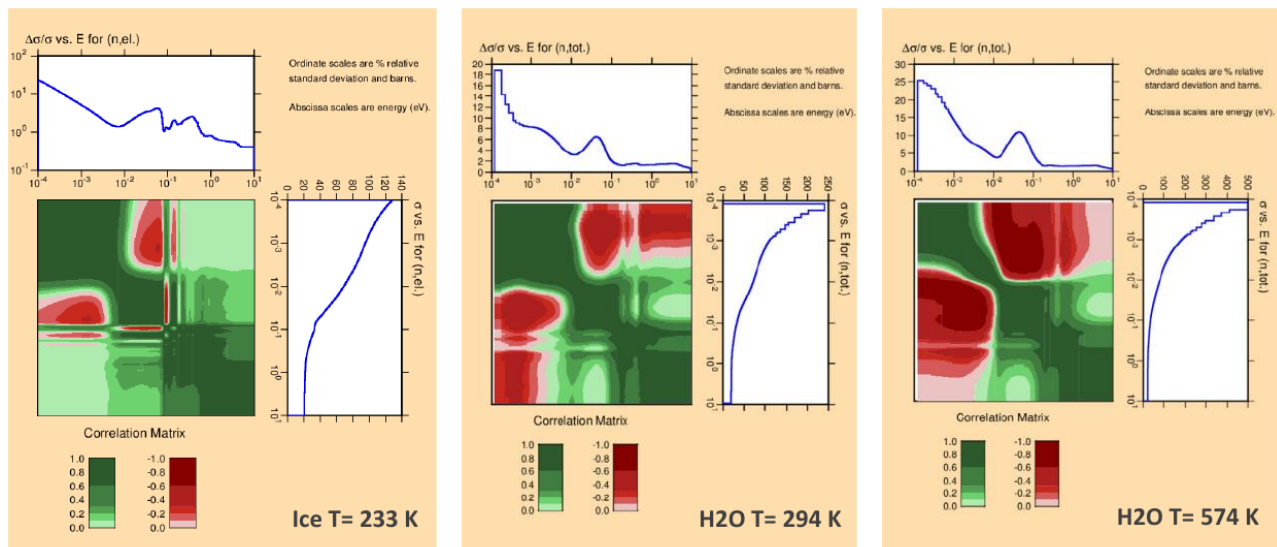


Figure 2. Covariance matrices of the elastic scattering cross sections for ice and light water generated via the CONRAD code.

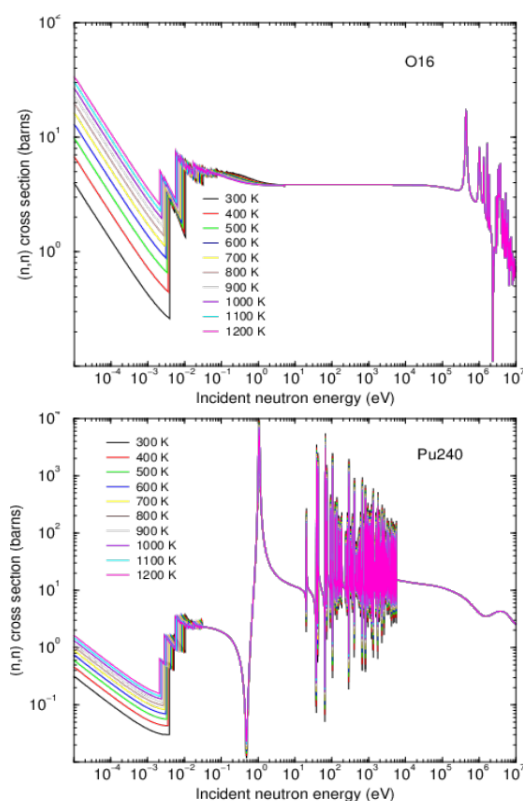


Figure 3. Elastic scattering cross sections of ^{16}O and ^{240}Pu in PuO_2 as a function of temperature.

LEAPR module. It relies on the phonon expansion method enriched by specific models for light water. In order to reproduce the experimental quasi-elastic neutron scattering (QENS) peaks, we implemented a roto-translational diffusion model with a random jump diffusion correction and a fluidity factor (Fig. 1).

In the CUBIC module, the incoherent elastic scattering cross sections rely on the LEAPR model. For the coherent contribution, a generalized formalism was implemented to account for structure information stored in CIF files, textured materials and anharmonicity via a third-cumulant coefficient as proposed by Willis [4]

For the propagation of the TSL uncertainties, we use the CONRAD code [5], which contains a mathematical framework for generating covariance information from model parameters. Figure 2 shows the results obtained for ice and light water.

Example of elastic scattering cross sections of ^{16}O and ^{240}Pu in PuO_2 are shown in Fig. 3. The so-called thermal cutoff energy delimits the low energy neutron cross section, which is reconstructed with the CINEL code and the resolved resonance range, which is reconstructed with a R-Matrix formalism.

3 Scattering experiments

Among the model parameters required for computing TSL, the lattice parameters and phonon density of states (PDOS) were the subject of experimental studies carried out at ILL.

Figure 4 summarizes the experimental work performed on ZrO_2 , UO_2 and ThO_2 by using the IN6-SHARP instrument. Results obtained for PuO_2 from inelastic X-ray scattering experiments reported in the literature are given for comparison. INS experiments required sample of large volume compared to X-ray experiments. If INS experiments can provide well-resolved experimental PDOS with low statistical uncertainties, its application is limited to stable or low radioactive nuclei. A good agreement between the data and the theoretical curves simulated with the TRIPOLI4 code was achieved by an iterative optimization of the PDOS.

For determining the lattice parameters as a function of temperature, experiments were performed on different

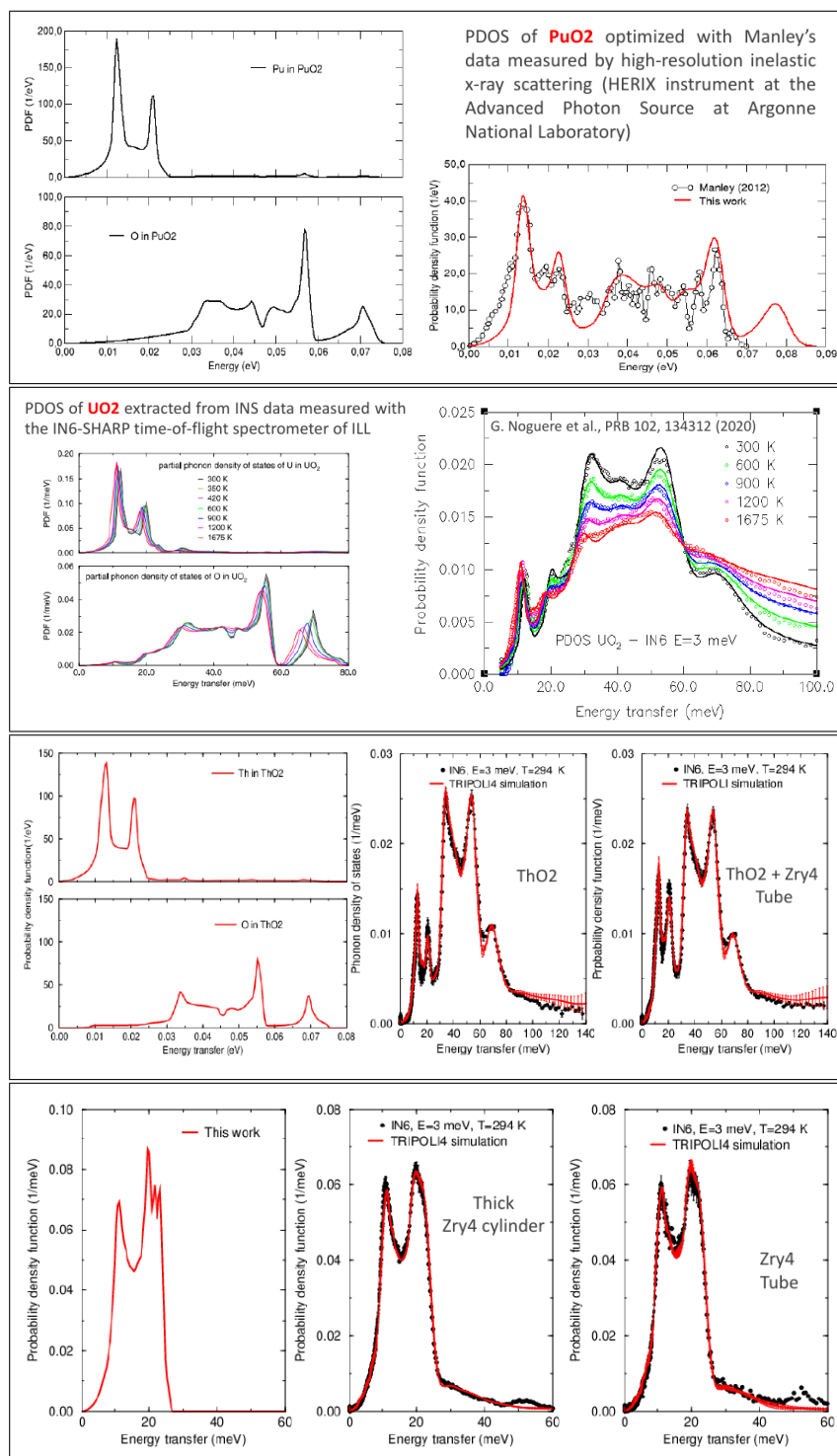


Figure 4. Examples of PDOS for PuO_2 , UO_2 , ThO_2 and Zry4 extracted from inelastic scattering measurements.

diffractometers available at ILL. For example, Fig. 5 compares the theoretical and experimental diffraction patterns for Zry4 measured with the D1B and D20 instruments. The Monte-Carlo code TRIPOLI4 was used to simulate the experimental results by adjusting the lattice parameters of the Zry4 hexagonal structure. Our results probe that the temperature-dependence of the obtained lattice parameters are well reproduced by a Debye model [6] from 40 K to 700 K, in which we used the Debye temperature of 261 K

deduced from the PDOS measured with the IN6-SHARP spectrometer (Fig. 4). Such a unique result is a good indication of the overall consistency of the CINEL models.

4 Theoretical Phonon Density Of States

The major limitation of the INS experiments relies on the response function of the spectrometers that depends on the time distribution of the neutron burst. As shown in

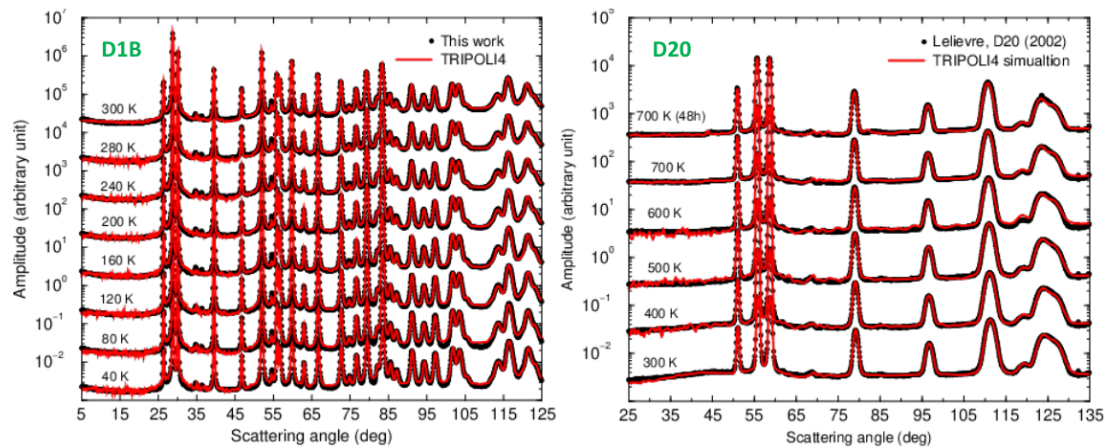


Figure 5. Examples of diffraction patterns for Zry4 measured with the D1B and D20 instruments of ILL.

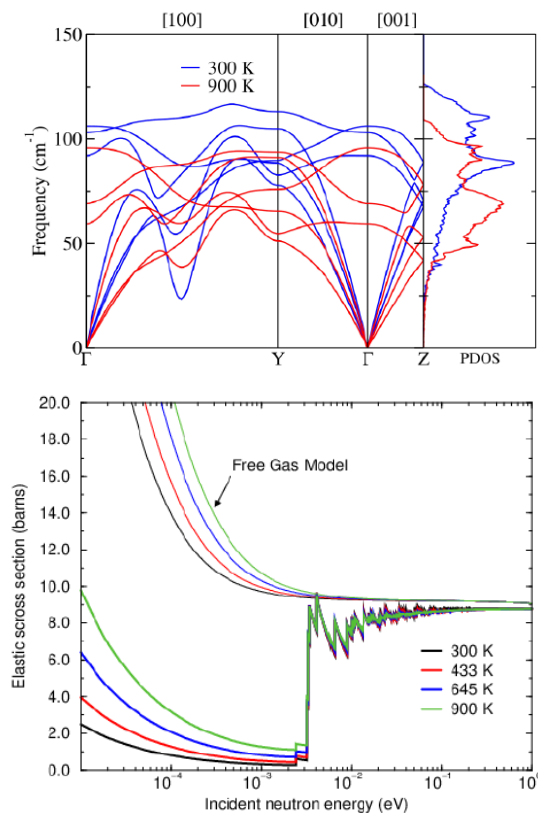


Figure 6. Ab initio phonon dispersion curves and phonon density of states of α -U taken from Ref. [7]. The neutron elastic cross sections were calculated with the CINEL code as a function of temperature.

Fig. 4, the response function of the IN6-SHARP spectrometer lumps fine substructures in the PDOS of Zry4 in a single broad peak, making it impossible to explore the full dynamic of the Zry4 crystal. Similarly, we can observe that the response function combined to the high-temperature effects smooths out the structures in the PDOS of UO₂. As a result, this measurement can only provide qualitative information on the anharmonic behavior of the

oxygen atoms relative to the uranium atoms. To overcome these experimental limitations, theoretical PDOS are needed to correctly interpret the experimental results. Future experimental works planned at the ILL facilities will deal with standard MTR nuclear fuel (UAl) and low-enriched nuclear fuel (U₃Si₂). This experimental program will benefit from the expertise of the Fuel Studies Department of CEA Cadarache, and from their knowledge on ab initio molecular dynamics (AIMD) simulations, as shown in Fig. 6 for α -U [7].

5 Conclusions

Results summarized in this paper show that the CINEL code is able to provide thermal scattering laws for materials of interest for nuclear applications with temperatures ranging from 40 K to 1000 K. The combination of INS and neutron diffraction experiments probes the consistency of the CINEL models. However, some experimental limitations make the use of ab initio simulations essential. Evaluations for Zry4, ThO₂ and α -U are still in progress. The UO₂ and PuO₂ files were delivered to the NEA data bank in the test version JEFF-4T1 [8]. The CINEL code and validation tests will be made available at the NEA data bank before the release of JEFF-4 in 2024.

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