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INVESTIGATION OF FREQUENCY SPECTRUM OF LIGHT WATER TO GENERATE THERMAL SCATTERING LAW

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ABSTRACT

Standard thermal neutron scattering cross-section libraries of light water generated using the LEAPR module of the NJOY code treat the cross-sections as an analytical function of the density of states, i.e. the frequency spectrum. Hence, an accurate estimation of the frequency spectrum is necessary to generate reliable thermal scattering cross-sections, as long as one chooses to use LEAPR. In this paper the experimental frequency spectrum of light water measured at several temperatures and pressures, using high resolution time-of-flight inelastic spectrometers, namely the IN4c and the IN6 at the Institut Laue-Langevin, Grenoble, France is presented. Within the scope of the experimental limitations of our measurements, an effort has been made to provide qualitative comparisons of the experimentally obtained frequency spectrum with that derived from the classical molecular dynamics simulations. The thermal scattering law present in the JEFF-3.1.1 library and a very recent test library developed at Centro Atomico Bariloche, Argentina, named as CAB library were chosen for our comparison. A study of the dependence of pressure and temperature on the experimental frequency spectrum was also carried out, majorly to predict the sensitivity of thermal scattering law to pressure.

KEYWORDS: Frequency spectrum, thermal scattering law, molecular dynamics, neutron cross-section

1. INTRODUCTION

The accuracy of criticality safety studies and reactor physics calculation depends on the nuclear data it uses. Existing thermal neutron scattering evaluations are inadequate for many nuclear systems and novel applications such as the design of small neutron sources, developing treatment plans for neutron and proton radiotherapy, etc [1]. There are needs for reviewing the existing thermal scattering data evaluations and support new experiments to improve them and expand their applicability. A commonly used tool by the reactor physics community, for thermal neutron scattering evaluation, is the LEAPR module of the NJOY code [2]. LEAPR calculates the thermal scattering law (TSL) using several approximations [3–5] and the entire physics behind the interaction of thermal neutrons and moderator material is turned into an analytical function of the frequency spectrum of the moderator material [4–6]. Following this, a majority of the existing TSL libraries still rely on the frequency spectrum based on the experiments performed in the sixties [7–9]. Experimental data are not always available for most of the necessary moderator materials to validate TSL libraries. Safety analysis and design of water moderated reactors like pressurized

water reactors (PWR) rely on the accurate estimation of thermal scattering cross-sections of water at high temperatures and pressures. This paper focuses on the investigation of light water thermal scattering cross-section data at PWR operating conditions. In order to reach better agreement with the experimental double-differential and total cross-section of light water, the existing TSL libraries need a proper estimation of the frequency spectrum for a broad range of temperatures and pressures. Inelastic neutron scattering (INS) measurements of light water with two time-of-flight spectrometers, namely the IN4c and IN6, at the Institut Laue-Langevin (ILL), Grenoble have been performed to generate the frequency spectrum at several high temperatures and pressures.

2. THERMAL SCATTERING LAW

A complete description of the thermal scattering process can be found in the literature [3–5]. Using the first Born approximation and Fermi pseudopotential, as formalized by Van Hove [3], the double-differential cross-section (DDXS) for thermal neutrons with an incident energy E , scattering angle θ , scattered with final energy E' and at temperature T can be expressed as a function of $S(\alpha, \beta)$, the thermal scattering law by:

$$\frac{d^2\sigma_T}{d\Omega dE'}(E \rightarrow E', \mu) = \frac{\sigma_b}{4\pi k_B T} \sqrt{\frac{E'}{E}} S(\alpha, \beta) \quad (1)$$

where α is the change in momentum of the neutron and β , the change in energy, with

$$\alpha = \frac{E + E' - 2\mu\sqrt{EE'}}{Ak_B T} \quad \beta = \frac{E' - E}{k_B T} \quad \text{and} \quad \sigma_b = \sigma_f \left(\frac{A + 1}{A} \right)^2 \quad (2)$$

Here, A is the ratio of the scatterer mass to that of the neutron mass, σ_b is the bound atom scattering cross-section, σ_f is the free atom scattering cross-section, μ is the neutron scattering cosine ($\mu = \cos\theta$) and k_B is the Boltzmann constant. Under the incoherent and the Gaussian approximation, the TSL can be expressed as a function of the frequency spectrum $\rho(\beta)$ of the excitations in the molecular system by [4]:

$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{i\beta\hat{t}} e^{-\gamma(\hat{t})} d\hat{t} \quad (3)$$

where t is the time measured in the units of $\hbar/(kT)$ seconds. $\gamma(\hat{t})$ is given by:

$$\gamma(\hat{t}) = \alpha \int_{-\infty}^{+\infty} P(\beta)(1 - e^{-i\beta\hat{t}}) e^{-\beta/2} d\beta \quad (4)$$

and

$$P(\beta) = \frac{\rho(\beta)}{2\beta \sinh(\beta/2)} \quad (5)$$

where $\rho(\beta)$ is the frequency spectrum. Neutrons scattering from light water can exchange energy and momentum from the motion of water molecules. Frequency spectrum defines the density function of available atomic vibrational energy states for energy exchange between a neutron and a water molecule [10]. These energy states correspond to the intramolecular and intermolecular vibrations, hindered rotations and molecular diffusion of the water molecule. The translation (diffusive properties) and hindered rotation mode describe the motion of the water molecule as a whole whereas the vibrational mode (bending and stretching) describes the motions of the atoms in the molecule.

3. TSL GENERATION METHODOLOGIES

Currently, available TSLs for light water in the evaluated thermal scattering libraries come from different physics models. These models depend on the frequency spectrum as an input to generate the TSL. One distinct feature in a very recent model named CAB model for water [11], involves molecular dynamics (MD) simulations of light water to generate the frequency spectrum. All the other models depend on the experimental frequency spectrum [5,6]. On the basis of the type of frequency spectrum used, the TSL libraries can be broadly classified into two types: The one obtained from the measurements and the other from the MD simulations.

3.1. TSL Based on Experimentally Measured Frequency Spectrum

The very first model for TSL generation based on an experimentally measured frequency spectrum was prepared by Koppel and Houston in 1978 [6] often referred as the General Atomics model. This model was based on the frequency spectrum measured by Haywood and Thorson [7]. Mean-

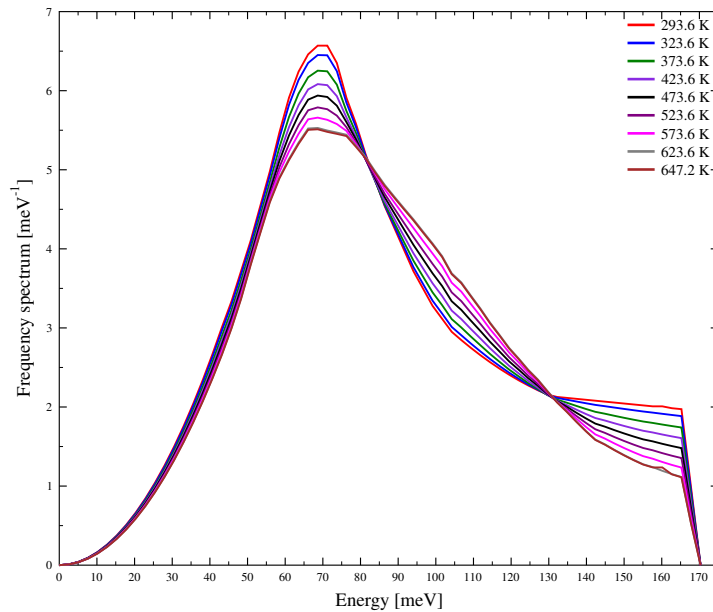


Figure 1: Frequency spectrum of light water based on the IKE model.

while, another model was developed by J. Keinert and M. Mattes in 1984 and then updated in 2005 which later became part of the JEFF-3.1 library [5]. This model is often referred as the IKE model and is based on the temperature dependent frequency spectrum collected by Haywood and Page at 300 and 550 K. For the intermediate temperatures between 300-500 K, this frequency spectrum is linearly interpolated. Hindered rotations of the water molecule are described as a solid type frequency spectrum and the molecular diffusion is treated as a free gas. Temperature dependent frequency spectrum of light water based on this IKE model is shown in Fig. 1. The energy band corresponding to the hindered rotation of the water molecule can be seen around 70 meV and is unaffected by the change in temperature. This IKE model was later adapted by the ENDF/B-VII library by slightly reducing the energy of the hindered rotation band to 60 meV to have better agreement with the experimental total cross-section in the energy range between 10-100 meV. It implies that the double-differential and total cross-section in the thermal energy range can be sensitive to the energy corresponding to the hindered rotations of the water molecule. It is thus important to perform new high-resolution measurements of the frequency spectrum of light water to properly characterize the molecular vibrations and hindered rotations.

3.2. TSL Based on MD Simulations

Until recently, standard data libraries have relied mostly on experimental data measured decades ago [7–9] and are based on physical models [4,5]. Consequently, some discrepancies are still observed between the evaluation and benchmark results at reactor normal operating conditions, even with the most recent TSL data libraries (JEFF-3.1.1, ENDF/B-VII.1). In the past few years, there has been a renewed interest in reviewing these models and in generating new thermal scattering libraries based on the recent advancement in the development of light water empirical potentials and improved computational capabilities. Several groups [11–13] have proposed different methodologies to produce TSL for light water. A promising technique which provides reliable TSL involves MD simulations of light water. This method allows the generation of the temperature-dependent frequency spectrum of light water based on the MD trajectories. This methodology is not completely approximation-free, due to the intrinsic limitation of the classical MD simulations at lower and higher energies, which are corrected by applying quantum corrections [13]. Other techniques rely on a combination of MD with available experimental data to calculate the parameters of TSL models like the one implemented in the LEAPR module of the NJOY code. The accuracy of the MD simulations also depends on the intra and interatomic water potentials used at high temperature and pressure. A bunch of water potentials, governed by empirical or semi-empirical intermolecular potentials exists [14]. A new TSL library for light water has been adapted by ENDF/B-VIII.β5 as a test library which is referred as the CAB model for light water. This new library is based on MD simulations of light water using the non-polarizable TIP4P/2005f [15] flexible light water potential. In this model, simulations are carried out at several temperatures [11] to generate the temperature-dependent frequency spectrum of light water. Instead of a free gas translation, as utilized in the IKE model, CAB models the molecular translation with the Egelstaff-Schofield diffusion model [16,17]. Fig. 2 shows the frequency spectrum of light water at different temperatures utilized by the CAB model in the ENDF/B-VIII.β5 data library. An interesting feature that is observed in the frequency spectrum based on the CAB model is that the energy of the hindered rotational band, centered around 65 meV at room temperature, shifts towards lower energies with increasing temperature. This shift of the hindered rotational band towards lower energy region is not in agreement with existing experimental data of frequency spectrum of light water [7–9].

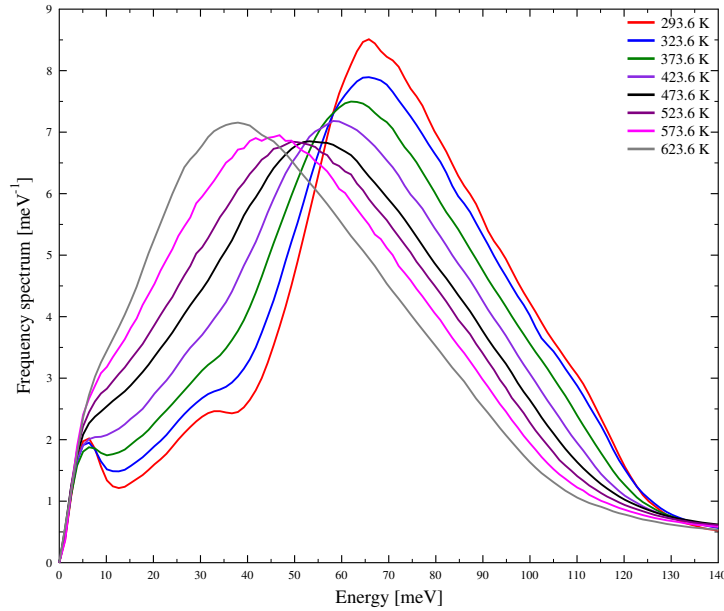


Figure 2: Frequency spectrum of light water based on the CAB model.

This difference, questions the accuracy of MD simulations of light water at high temperature and pressure using interaction potentials [15] that are parametrized to simulate bulk water properties at room temperature. It should be noted as a matter of fact that a flexible water potential partially accounts for the polarization of the water. However, although flexible water potentials provide convincing results at room temperature and pressure but fail to mimic the properties of water at high temperatures and pressure in agreement with the experiments [14]. A direct comparison of the MD result with the experimental data is important for the validation of these simulations and thus the water model/potential used.

4. TOTAL SCATTERING CROSS-SECTION

The total scattering cross-section for light water has been computed using the TSL based on the above-described models and the THERMR module of the NJOY code. Oxygen was treated as a free gas. The total scattering cross-section was obtained in the following manner:

$$\sigma_{\text{H}_2\text{O}} \simeq (2 \times \sigma_H) + \sigma_O \quad (6)$$

The upper part of Fig. 3 shows the total scattering cross-section of light water for 293.6 K, 423.6 K and 573.6 K using the TSL data from both, the CAB (right) and the IKE model (left). The lower part of this figure shows a ratio of the total scattering cross-section at temperature 293.6 K, 423.6 K and 573.6 K with respect to 293.6 K. This type of ratio serve as a quick analysis of the temperature dependence of the total scattering cross-section in the thermal energy range. An expected trend would show an increase in the ratio with increasing temperature due to increase in the probability of up-scattering of thermal neutrons. This behavior is observed with the IKE model (left), but on the other hand, the CAB model (right), between neutron energy 18-45 meV indicates that the

cross-section decreases with increasing temperature. Thermal reactors are sensitive in this energy region and any discrepancy in the total cross-section would lead to misleading results in criticality safety studies benchmarks.

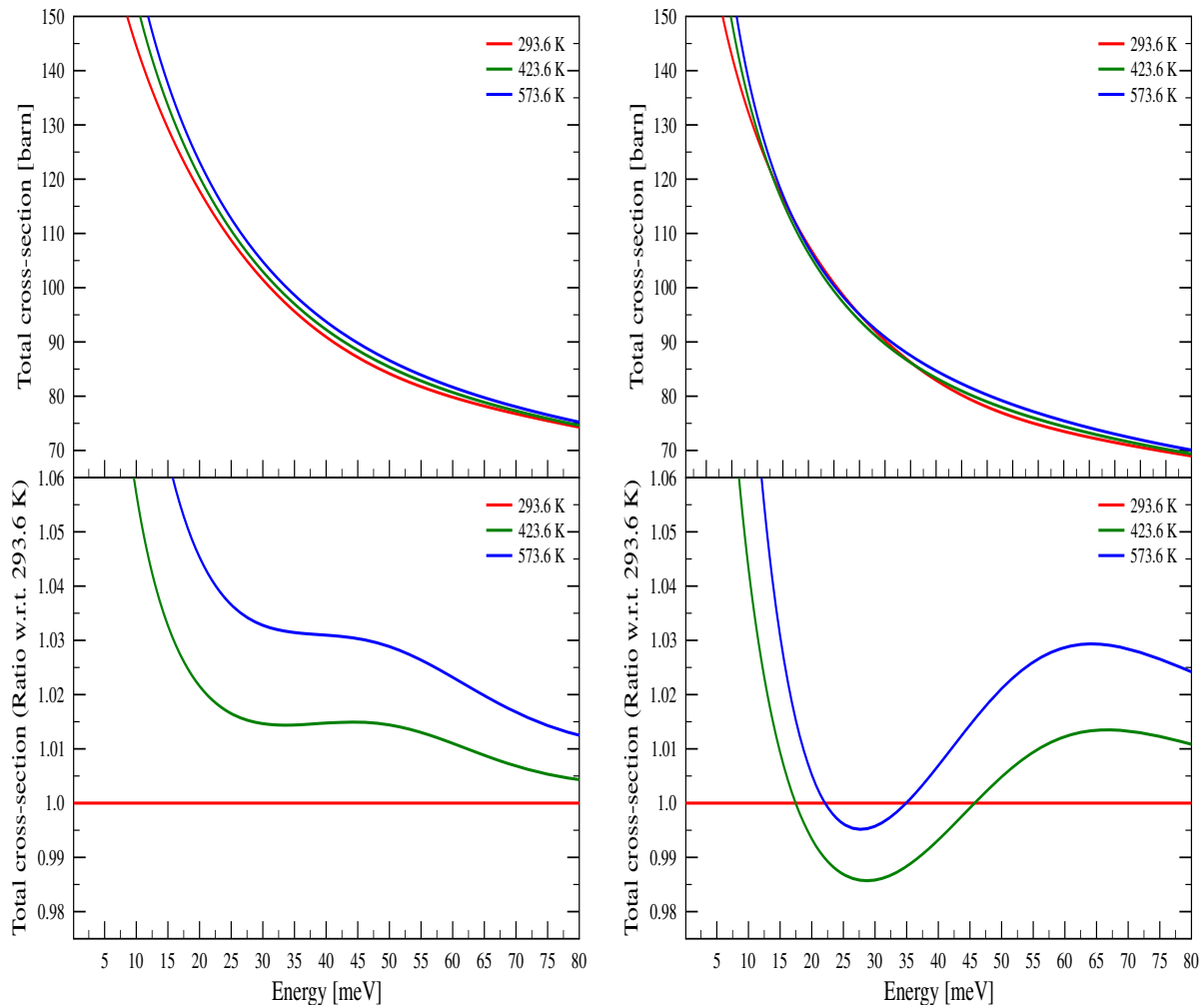


Figure 3: Total scattering cross-section obtained from the CAB (right) and the IKE model (left).

5. INELASTIC NEUTRON SCATTERING EXPERIMENT TO GENERATE FREQUENCY SPECTRUM OF LIGHT WATER

Inelastic neutron scattering (INS) spectra of light water were measured on two time-of-flight spectrometers, IN4c and IN6, at the Institut Laue-Langevin (ILL), Grenoble. The incident neutron energy for IN4c and IN6 were 14.2 meV and 3.15 meV respectively. These two spectrometers provide high resolution spectroscopy, allowing a flexible energy and momentum transfer range. Measurements were carried out in the temperature range of 300-517 K and pressure range of 1-

470 bar matching the normal operating conditions of nuclear power reactors as shown in Fig. 4. Measured INS spectra are in the up-scattering mode (neutron energy gain or sample energy loss) for both the spectrometers due to low incident neutron wavelength. The experimental resolution of both the spectrometers were derived by fitting the vanadium data. The data analysis was performed using the LAMP [18] data analysis tool. Dynamic structure factor (DSF) $S(q, \omega)$ was obtained after the necessary data reduction of the experimental data. Readers are suggested to refer a recently published conference paper [19] for a detailed explanation of the data reduction and analysis.

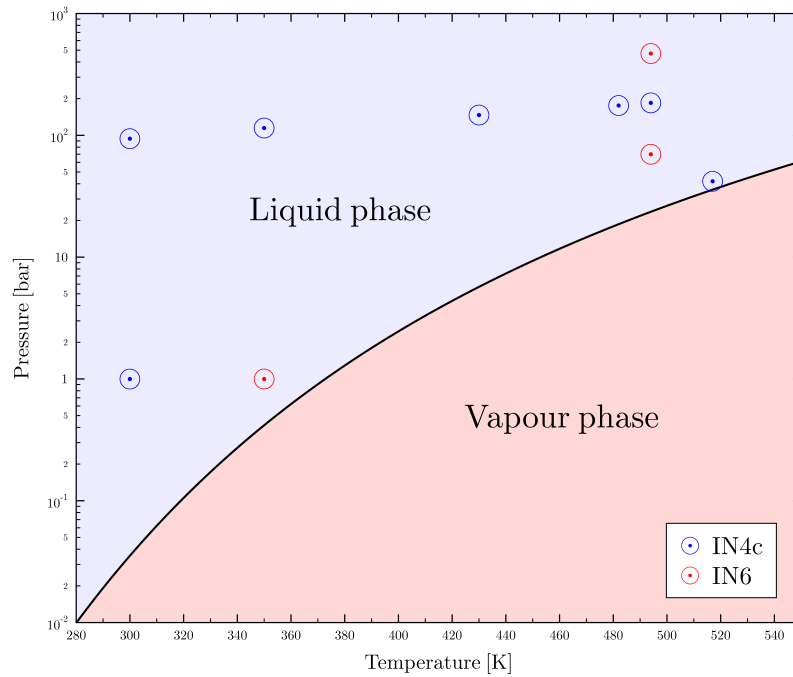


Figure 4: Thermodynamic conditions of INS measurements of light water at ILL.

Using LAMP, the DSF $S(q, \omega)$ was finally transformed to the frequency spectrum $\rho(\omega)$ based on equation 7 [20]:

$$\rho(\omega) \simeq \lim_{q \rightarrow 0} S(q, \omega) \frac{\omega^2}{q^2} \quad (7)$$

6. RESULTS

The neutron weighted frequency spectrum is derived from the INS spectra obtained from both the spectrometers and the results are presented in Fig. 5 and 6. Even though at this level, the experimental data is not corrected for multiple scattering, all other necessary corrections have been performed to provide qualitative results.

6.1. Pressure Dependence on the Frequency Spectrum

Two measurements at the same temperature but at different pressures have been carried out to study the impact of pressure on the TSL and also to validate our previous study of the impact of pressure on the experimental DDXS [19]. The results obtained from the INS spectra from IN4c

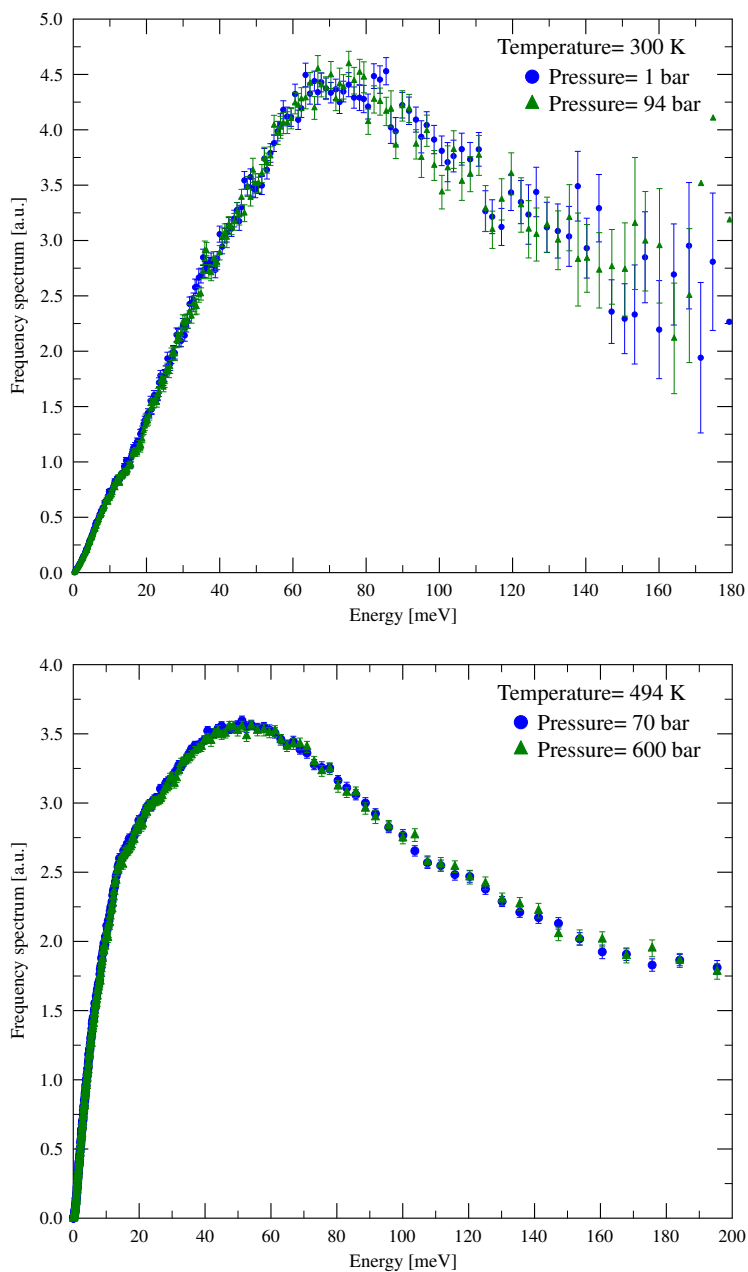


Figure 5: Frequency spectrum of light water from IN4c (top) and IN6 (bottom) measured at different pressures.

at 300 K and two different pressures, i.e., 1 and 94 bar are shown in Fig. 5 (top). The results from

the IN6 measurements at 494 K and two different pressures, i.e, 70 and 600 bar are shown in Fig. 5 (bottom). The results obtained from both the spectrometers indicate that there is a negligible change in the frequency spectrum of water with the change in pressure. This also confirms our previous study [19] of the negligible change in experimental DDXS with the change in pressure.

6.2. Temperature Dependence on the Frequency Spectrum

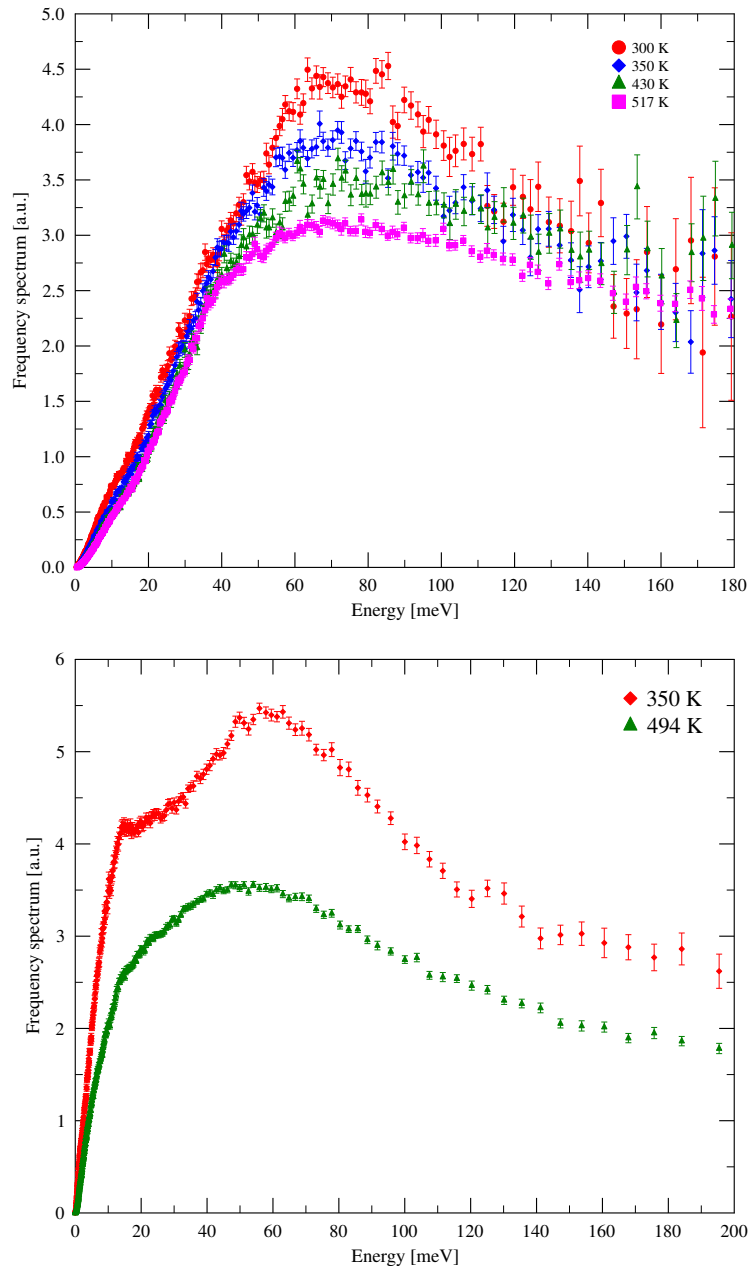


Figure 6: Frequency spectrum of light water from IN4c (top) and IN6 (bottom) measured at different temperatures.

Fig. 6 shows the variation of the frequency spectrum of water with respect to temperature. The energy corresponding to the hindered rotational band around 70 meV, observed in our IN4c data (top) at 300 K is in good agreement with the measurements by Haywood [8]. There is a slight shift in this rotational band towards lower energy with increasing temperatures. A possible reason for this shift might be the weakening of hydrogen bonds at higher temperatures. But, within the experimental limitations of the instrument resolution at higher energies it can be concluded that the hindered rotational band lies close to 70 meV. Due to the limitation in the experimental resolution, the translational band is missing in the IN4c spectra at all temperatures. In the IN6 spectra (bottom) this hindered rotational band is slightly shifted towards lower energy side, i.e., around 60 meV at 350 K and around 55 meV at 494 K. For both these temperatures, a peak around 12 meV is observed which might possibly correspond to the translation band of light water. A similar peak at this energy is observed in the frequency spectrum of Haywood and Page [9].

Our present results of the frequency spectrum from both the IN4c and IN6 spectra are limited to the hindered rotational band due to the limitation in the energy transfer range. This limited range of energy transfer from both the spectrometers makes it insufficient to be fully used as an input frequency spectrum to the LEAPR module of the NJOY code. In order to overcome this, new experiments are under considerations to extend these measurements at spallation sources like ISIS, SNS or PSI where higher neutron energies are available. The experimentally obtained frequency spectrum can then be further used by the NJOY code to generate new TSL evaluations.

7. CONCLUSIONS

This paper is in connection with reactor physics applications, where an accurate determination of the frequency spectrum at reactor operating conditions is important to generate the TSL. Measurement of the dynamic structure factor and hence the frequency spectrum of light water at several high temperatures and pressures were carried out at the Institut Laue-Langevin. These measurements were compared with the frequency spectrum of light water utilized by the existing TSL evaluations. Some discrepancies in the shape of the frequency spectrum obtained from the experiments and those utilized by the CAB model based on classical MD simulations with TIP4P/2005f flexible water potential were observed. In the frequency spectrum obtained from our experiment, a very slight shift in the energy of the hindered rotational band with increasing temperature is observed, as opposed to the CAB model where this shift is significantly larger. However, the energy of the hindered rotational band at room temperature, obtained from CAB model is in close agreement with our measurements. It has been demonstrated that choosing a water potential without testing its limitations for high temperature and pressure conditions is not a viable option for the reactor physics community. A given water model/potential is relevant only when it is able to reproduce water properties at high temperatures and pressures. Pointing out the best water potential is difficult as different potentials reflect various different properties at different thermodynamic conditions. Certain water potential exists which takes polarization effect into account which respond to the fluctuations in the electric field due to molecular motion. A non-polarizable model that is parametrized at ambient conditions might fail to predict the change in the hydrogen bond network at high temperatures and pressures. Using a water potential for reactor physics applications implies its test for all the thermodynamic properties of water at the required temperatures and pressures, failure to do so would lead to misleading results and conclusions. Work is underway to investigate these features. Experimental measurement of the frequency spectrum is a promising solution to

generate frequency spectrum at high temperatures and pressures.

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