

Generation of thermal scattering libraries for liquids beyond the Gaussian approximation using molecular dynamics and NJOY/LEAPR



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ABSTRACT

In this paper we review the existing methodologies for the generation of thermal scattering libraries for liquids from molecular dynamics simulations, either by Fourier transform or developing models in the Gaussian approximation. To overcome the limitations of the Gaussian approximation in the quasielastic range, we present a modification introduced to the LEAPR module of NJOY to include a random jump diffusion model, and a methodology to obtain the required parameters. This methodology is applied to light water with good results.

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1. Introduction

In the thermal neutron scattering libraries the double differential scattering cross section is not stored itself, but as a function S of the change in momentum and energy known as the *thermal scattering law* (TSL) (MacFarlane and Kahler, 2010). The thermal scattering law is a function of two non dimensional quantities, α and β :

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

$$\alpha = \frac{E' + E - 2\sqrt{EE'} \cos \theta}{m/m_n k_B T}; \quad \beta = \frac{E' - E}{k_B T} \quad (2)$$

where σ_b is the bound scattering cross section of the atom, m is the mass of the scattering atom, m_n is the mass of the neutron, T is the temperature of the medium, k_B is Boltzmann's constant, μ is the cosine of the scattering angle, E is the incident neutron energy and E' is the outgoing neutron energy.

In neutron scattering and condensed matter physics, the thermal scattering law is called *dynamic structure factor* or *scattering function* (Squires, 2012) and is usually written as a function of the change in wave vector² $Q = |\vec{k}' - \vec{k}|$ and the excitation $\omega = (E' - E)/\hbar$:

$$\frac{d^2\sigma}{dE'd\mu} = \sigma(E \rightarrow E', \mu) = \frac{\sigma_b}{2\hbar} \sqrt{\frac{E'}{E}} S(Q, \omega) \quad (3)$$

where E and \vec{k} are the energy and wave vector of the incident neutron, and E' and \vec{k}' are the energy and wave vector of the outgoing neutron. It is important to note that both functions represent the same physical quantity, and

$$S(\alpha, \beta) = \frac{k_B T}{\hbar} S(Q, \omega) \quad (4)$$

$$\alpha = \frac{\hbar^2 Q^2}{2mk_B T}, \quad \beta = \frac{\hbar\omega}{k_B T} \quad (5)$$

In the past few years there has been a renewed interest on the evaluation of thermal neutron scattering laws and their uncertainties. This interest generated the creation of a new subgroup within the Working Party on International Nuclear Data Evaluation Cooperation (WPEC, 2015), SG-42, dedicated to the development of thermal scattering libraries, and several research projects. For liquids, particularly for light water, there are projects at Kyoto University (Abe et al., 2014; Abe and Tasaki, 2015), the NAUSICAA collaboration (Farhi et al., 2015), and here at Centro Atómico Bariloche (Marquez Damian et al., 2014). While independent, all these projects share one thing: they all seek to produce TSL for water from molecular dynamics results, in some cases in combination with experimental data.

Molecular dynamics is a computational chemistry technique used to calculate equilibrium and transport properties of classical molecular systems (Frenkel and Smit, 2002). The molecular system is simulated as a set of classical particles representing the atoms,

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² For liquids isotropy is considered, therefore the dependency is on Q , the modulus of \vec{Q} .

with inter and intra molecular forces represented as classical potentials. Starting from an initial configuration of positions and velocities, the equations of motion are integrated in time to calculate the trajectories of the particles: $\vec{r}(t)$ and $\vec{v}(t)$. Barostat and thermostat algorithms can be used to simulate the system at specific thermodynamics conditions.

2. Review of calculation schemes

The output of the molecular dynamics simulation can be used directly to compute the TSL, or can be used to calculate the parameters of thermal scattering models such as the models implemented in LEAPR. Thus, to analyse the calculation methodologies of these projects we divide them into two categories:

- *thermal scattering models*, with parameters computed from molecular dynamics results, and
- *direct calculations*, on which the TSL is calculated directly from trajectories computed from molecular dynamics.

Thermal scattering models are discussed in Section 2.1, and direct calculations in Section 2.2.

2.1. Thermal scattering models

The methodologies presented by Abe, Tsuboi and Tasaki at Kyoto University (Abe et al., 2014), and the CAB Model from Centro Atómico Bariloche (Marquez Damian et al., 2014) use molecular dynamics calculations to compute the parameters of thermal scattering models.

In the calculation scheme developed at Kyoto University, a generalized frequency spectrum is computed from the velocity autocorrelation function (VACF):

$$g(\omega) = \frac{2m}{3\pi k_B T} \int_0^\infty d\tau \text{VACF}(\tau) \quad (6)$$

with:

$$\text{VACF}(\tau) = \langle \vec{v}(t) \cdot \vec{v}(t + \tau) \rangle \quad (7)$$

where $\vec{v}(t)$ is the velocity vector at time t for a particular atom type, extracted from the molecular dynamics trajectory file, and $\langle \rangle$ is an average over all atoms of that type. The generalized frequency spectrum is used to compute the intermediate scattering function in the Gaussian approximation (Abe et al., 2014; Squires, 2012):

$$I(Q, t) = e^{-\gamma(t)Q^2} \quad (8)$$

with

$$\gamma(t) = \frac{\hbar}{m} \int_0^\infty d\omega \frac{g(\omega)}{\omega} \left\{ \coth\left(\frac{\hbar\omega}{2k_B T}\right) (1 - \cos \omega t) - i \sin \omega t \right\} \quad (9)$$

The scattering law is then computed as the Fourier transform of the intermediate scattering function:

$$S(Q, \omega) = \frac{1}{2\pi} \int_0^\infty dt e^{-i\omega t} I(Q, t) \quad (10)$$

In the CAB Model, Eq. (6) is used to compute a generalized frequency distribution $g(\omega)$, which is then separated into two components:

- a diffusion component computed using the Egelstaff-Schofield diffusion model (Egelstaff and Schofield, 1962), which describes a diffusion behavior at low (Q, ω) , and an asymptotic free-gas behavior at high (Q, ω) . The frequency spectrum for the Egelstaff-Schofield diffusion model is given by:

$$\rho_{\text{diff}}(\omega) = \frac{4c w_t \hbar}{\pi k_B T} \sqrt{c^2 + 1/4} \cdot \sinh(\hbar\omega/(2k_B T)) \cdot K_1 \left\{ \hbar\omega/(k_B T) \sqrt{c^2 + 1/4} \right\} \quad (11)$$

with $w_t = m/m_{\text{diff}}$ and $c = \frac{mD}{w_t \hbar}$, where m_{diff} is the diffusion mass and D is the self-diffusion coefficient and K_1 is the modified Bessel function of the second kind, and

- a solid-like continuous spectrum obtained by subtracting the diffusion spectrum from the generalized frequency distribution:

$$\rho(\omega) = g(\omega) - \rho_{\text{diff}}(\omega) \quad (12)$$

The thermal scattering law is then computed using a standard version of the LEAPR module of NJOY (MacFarlane and Kahler, 2010). In LEAPR, the calculation is performed in two parts: a diffusional component $S_{\text{diff}}(Q, \omega)$, and a non-diffusional component $S_{\text{non-diff}}(Q, \omega)$.

The diffusion component is calculated using the Egelstaff-Schofield diffusion model:

$$S_{\text{diff}}(Q, \omega) = \frac{\hbar}{k_B T} \frac{DQ^2}{\pi} e^{\frac{MD^2Q^2}{w_t k_B T} - \frac{\hbar\omega}{2k_B T}} \times \frac{\sqrt{c^2 + 1/4}}{\sqrt{\omega^2 + (DQ^2)^2}} K_1 \left[\frac{\hbar}{k_B T} \sqrt{c^2 + 1/4} \sqrt{\omega^2 + (DQ^2)^2} \right] \quad (13)$$

and the non-diffusional component is calculated using a phonon expansion in the Gaussian approximation from the solid-like continuous spectrum $\rho(\omega)$.

The TSL is then calculated as a convolution in energy of the two parts:

$$S(Q, \omega) = S_{\text{diff}}(Q, \omega) \otimes S_{\text{non-diff}}(Q, \omega) \quad (14)$$

The thermal scattering law computed with the Kyoto University model is inherently Gaussian, because it is entirely computed in the Gaussian approximation. The thermal scattering law computed with the CAB Model is also Gaussian, because (as we will see in Section 3) the Egelstaff-Schofield diffusion model can be expressed as a Gaussian intermediate scattering function.

2.2. Direct calculations.

As a contrast to models, direct calculations attempt to obtain $S(Q, \omega)$ directly from the atoms positions ($\vec{r}(t)$), extracted from the trajectory obtained by molecular dynamics. The general scheme to this is to calculate the Van Hove autocorrelation function³:

$$\tilde{G}(r, t) = \langle \delta[\vec{r} + \vec{r}_i(0) - \vec{r}_i(t)] \rangle \quad (15)$$

and then the thermal scattering law is calculated by Fourier transform in space and time:

$$\tilde{S}(Q, \omega) = \frac{1}{2\pi} \int_{-\infty}^\infty e^{i(\vec{Q} \cdot \vec{r} - \omega t)} \tilde{G}(r, t) \quad (16)$$

The problem of this scheme is that $\tilde{S}(Q, \omega)$ is not the quantum thermal scattering law $S(Q, \omega)$, but its classical counterpart, which is only valid for motions with times and distances far larger than the quantum scale:

$$1/\omega \gg \hbar/k_B T, \quad 1/Q \gg \hbar/\sqrt{2Mk_B T} \quad (17)$$

³ In the following derivation we include only the self component of the autocorrelation function, which is dominant in incoherent scatterers like light water. The coherent component, if needed, can be computed in the Sköld approximation (Sköld, 1967).

This restricts the validity of this kind of classical calculation to the region of low energy and momentum transfer (for water at room temperature, $\hbar\omega \ll 25.3$ meV, $Q \ll 3.5 \text{ \AA}^{-1}$).

The projects that use the direct approach -namely, the NAUSICAA project and the group at Kyoto University- overcome this limitation and use classical results beyond their limits using so called *quantum corrections*.

In the case of light water calculations in the NAUSICAA project (Farhi et al., 2015), the classical $\tilde{S}(Q, \omega)$ is used as a replacement of the symmetric quantum mechanical thermal scattering law:

$$S(Q, \omega) = e^{-i\hbar\omega/(2k_B T)} S_{\text{sym}}(Q, \omega) \simeq e^{-i\hbar\omega/(2k_B T)} \tilde{S}(Q, \omega) \quad (18)$$

thus applying the *quasiclassical* or Schofield approximation (Schofield, 1960). This approximation performs a correction on the order of \hbar , which is still not enough to calculate a $S(Q, \omega)$ valid in the whole (Q, ω) range needed for reactor calculations (Sears, 1985). To fix this, the NAUSICAA project replace the high (Q, ω) region of the TSL by ENDF/B-VII data computed in the Gaussian approximation.

In the GAAQC (Gaussian Approximation Assisted Quantum Correction) proposed by Abe and Tasaki from Kyoto University (Abe and Tasaki, 2015), the classical intermediate scattering function calculated from the classical Van Hove autocorrelation function:

$$\tilde{I}(Q, t) = \int d\vec{r} e^{i\vec{Q}\cdot\vec{r}} \tilde{G}(r, t) \quad (19)$$

is corrected by applying a correction function $R(Q, t)$:

$$I(Q, t) = \tilde{I}(Q, t)R(Q, t) \quad (20)$$

$R(Q, t)$ is computed as the ratio of two Gaussian intermediate scattering functions: one with its width computed in the quantum mechanical version (Eq. 9) and and one computed in the classical approximation:

$$R(Q, t) = \frac{I^G(Q, t)}{\tilde{I}^G(Q, t)} = \frac{e^{-\gamma(t)Q^2}}{e^{-\tilde{\gamma}(t)Q^2}} \quad (21)$$

$$\gamma(t) = \frac{\hbar}{m} \times \int_0^\infty d\omega \frac{g(\omega)}{\omega} \left\{ \coth\left(\frac{\hbar\omega}{2k_B T}\right) (1 - \cos \omega t) - i \sin \omega t \right\} \quad (22)$$

$$\tilde{\gamma}(t) = \frac{2k_B T}{m} \int_0^\infty d\omega \frac{g(\omega)}{\omega^2} (1 - \cos \omega t) \quad (23)$$

One important note on these direct methods they still require calculations in the Gaussian approximation, either to correct the classical results or to replace them in the high (Q, ω) region where they are not valid.

3. Diffusion models

3.1. Random jump diffusion

The Gaussian approximation, used in the models presented in Section 2.1 is strictly valid for some particular cases (free gas, harmonic oscillator and simple diffusion), and results on a good description for liquids in the high energy region ($E \sim 1$ eV), where internal vibrations and the asymptotic free gas behavior are dominant.

In the low energy region, for a system with self diffusion coefficient D , the Gaussian approximation can only represent simple diffusion, i.e.:

$$I_{\text{diff}}(Q, t) = e^{-Q^2 D |t|} \quad (24)$$

which corresponds to a Van Hove correlation function that satisfies Fick's law.

The Fourier transform in time of Eq. (24) is a Lorentzian:

$$S_{\text{diff}}(Q, \omega) = \frac{1}{\pi} \frac{DQ^2}{\omega^2 + (DQ^2)^2} \quad (25)$$

with half-width at half maximum (HWHM) $\Gamma(Q) = DQ^2$.

Approximating $K_1(x)$ as $1/x$ for $x \rightarrow 0$, the Egelstaff-Schofield model (Eq. (13)) reduces to Eq. 25 in the $(Q, \omega) \rightarrow 0$ limit, reproducing the same DQ^2 HWHM in the quasielastic region.

In liquids, when the dynamics is observed over a time and length scale that reveals the non-continuous character of the particles' motion, its description by the Fick's law (Egelstaff, 1994) is inadequate. This is observed by quasielastic neutron scattering (QENS), where the dynamic structure factor is measured at low (Q, ω) and it is found that the HWHM of the quasielastic peak is not a linear function of Q^2 as predicted by Eq. (25) (Fig. 1).

The actual mechanism behind this behavior is still a matter of discussion within the community of water chemical physics (Qvist et al., 2011; Teixeira, 2012), and several models have been proposed to explain it (Egelstaff, 1994). One of the simplest is the random jump diffusion model, on which the scattering law is still Lorentzian, but the HWHM has a non-linear dependence in Q^2 :

$$S_{\text{RJD}}(Q, \omega) = \frac{1}{\pi} \frac{\Gamma(Q)}{\omega^2 + \Gamma(Q)^2} \quad (26)$$

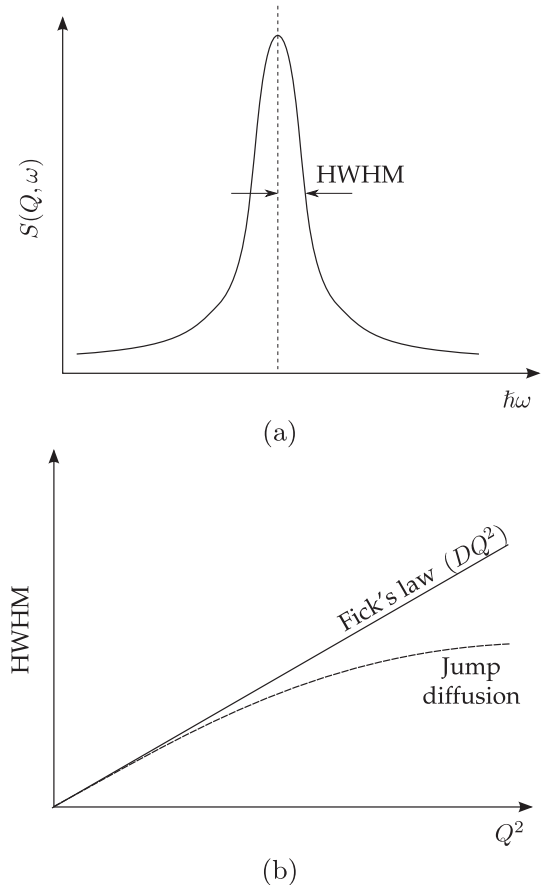


Fig. 1. (Top) half-width at half maximum of the quasielastic peak for a system where Fick's law is satisfied and for a system with jump diffusion (bottom).

with

$$\Gamma(Q) = \frac{DQ^2}{1 + DQ^2\tau_0} \quad (27)$$

where τ_0 is the residence time between jumps.

3.2. Implementation in LEAPR

Eqs. (26) and (27) are equivalent to a simple diffusion model with a variable diffusion coefficient, $\mathcal{D}(Q) = \Gamma(Q)/Q^2$. Since LEAPR evaluates $S(\alpha, \beta)$ at constant α , the Egelstaff-Schofield diffusion model can be rewritten to include this dependence in Q :

$$S_{\text{diff}}(Q, \omega) = \frac{\hbar}{k_B T} \frac{\mathcal{D}(Q)Q^2}{\pi} e^{\frac{m\mathcal{D}(Q)^2Q^2}{w_1 k_B T} - \frac{\hbar\omega}{2k_B T}} \times \frac{\sqrt{c(Q)^2 + 1/4}}{\sqrt{\omega^2 + \mathcal{D}(Q)^2Q^2}} K_1 \left[\frac{\hbar}{k_B T} \sqrt{c(Q)^2 + 1/4} \sqrt{\omega^2 + \mathcal{D}(Q)^2Q^2} \right] \quad (28)$$

with:

$$c(Q) = \frac{\mathcal{D}(Q)m}{w_1 \hbar} \quad (29)$$

$$\mathcal{D}(Q) = \frac{D}{1 + DQ^2\tau_0} \quad (30)$$

A similar methodology, although in the time domain, was utilized by Edura and Morishima (2004) to introduce non-Gaussian diffusion into their water model.

The utilization of this diffusion model requires the introduction of a new parameter, the relaxation time τ . This relaxation time can be computed from molecular dynamics simulations, using a direct calculation in the classical approximation:

- The classical Van Hove self correlation function, $\tilde{G}(r, t)$, is computed from the trajectory file.
- The classical scattering law, $\tilde{S}(Q, \omega)$ is calculated as the Fourier transform in time and space of the Van Hove self correlation function.
- The width of the quasielastic peak is calculated from $\tilde{S}(Q, \omega)$, plotted against Q^2 , and fitted with Eq. (27) to calculate the diffusion coefficient D (if an experimental value is not used) and the relaxation time τ . This can be done because the conditions presented in Eq. (17) are satisfied.

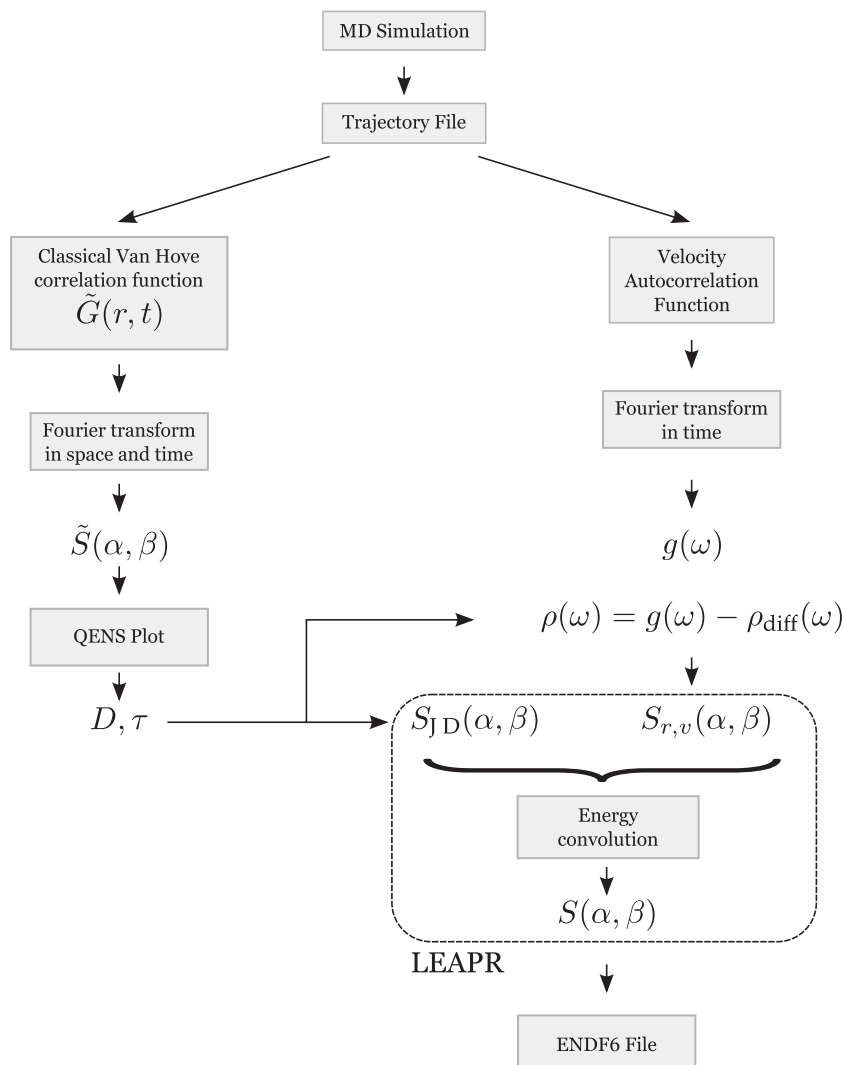


Fig. 2. Calculation methodology to evaluate thermal scattering cross sections in LEAPR for liquids with the jump diffusion model.

The user provides in the input of LEAPR three parameters: w_t , $c_0 = c(Q = 0) = Dm/(w_t h)$ and τ_0 . This allows for the following cases:

- $w_t = 0$, no translational component (solid).
- $w_t > 0$, translational component:
 - $c_0 = 0$, translation is modeled as free gas.
 - $c_0 = 0$, diffusion:
 - $\tau_0 = 0$, simple diffusion (Egelstaff–Schofield model).
 - $\tau_0 > 0$, random jump diffusion.

The random jump diffusion model is implemented in the outer loop of the subroutine `trans`, which iterates over α . For each α value in the grid, we combine Eqs. (29) and (30) to calculate the value of c :

$$c = \frac{c_0}{1 + k_B T w_t \alpha c_0 \tau_0 / \hbar} \quad (31)$$

and Eq. (28) is evaluated using the Egelstaff-Schofield model implemented in LEAPR.

4. Application to light water

The calculation methodology requires the calculation of the frequency spectrum and the jump diffusion parameters. Starting from a molecular diffusion simulation:

- The velocity autocorrelation function (VACF) is computed from the trajectory file.
- The generalized frequency distribution, $g(\omega)$ is calculated as the Fourier transform of the VACF.
- A solid-like continuous spectrum, $\rho(\omega)$ is obtained by subtracting the diffusion spectrum calculated with the Egelstaff-Schofield model (Eq. (11)).
- With this we prepare an input for our modified version LEAPR. In LEAPR:
 - The solid-like continuous spectrum is used to compute the non-diffusional component, $S_{\text{non-diff}}(\alpha, \beta)$.
 - The diffusion component, $S_{\text{JD}}(\alpha, \beta)$, is calculated using the jump diffusion model.
 - Both components are convolved in energy to obtain the thermal scattering law and produce an ENDF6 formatted file.

A flowchart of this methodology is shown in Fig. 2.

This methodology was applied to light water at 293.6 K, using as a base the CAB Model for light water. Fitting Eq. (27) to the width of the quasielastic peak calculated in the classical approximation (Section 2.2), and using a diffusion constant $D = 0.2 \text{ \AA}^2/\text{ps}$ measured by Mills (1973), a relaxation time $\tau = 1.05 \text{ ps}$ was obtained.

As expected, the modified water model reproduces well the HWHM measured by Teixeira et al. (1985) for $E_0 = 3.14 \text{ meV}$ (Fig. 3). This improvement is similar to the results obtained by Abe and Tasaki using the GAAQC method (Abe and Tasaki, 2015). The total cross section does not change significantly with this change of diffusion models and it is only slightly reduced in the cold neutron energy range (Fig. 4).

5. Conclusions

The evaluation of thermal scattering cross sections requires the utilization of models which are as accurate as possible and include all the physical phenomena present in the material. The usual approximation for the evaluation of the incoherent component of the thermal scattering law, the Gaussian approximation, is known

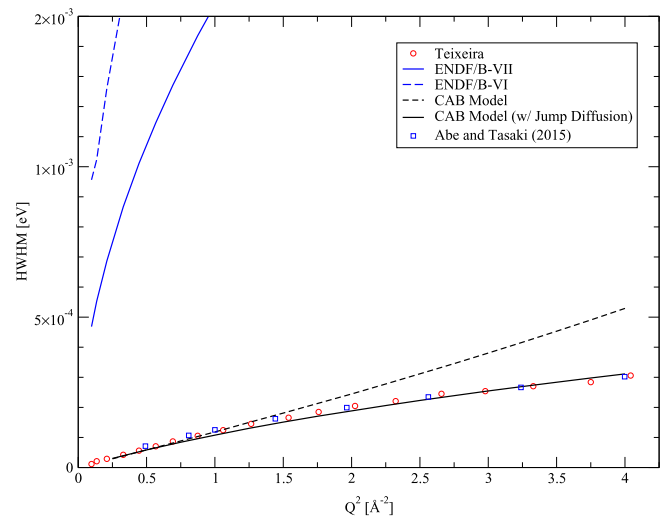


Fig. 3. Half-width at half maximum of the quasielastic peak for $E_0 = 3.14 \text{ meV}$ neutrons incident in light water, compared with measurements by Teixeira et al. (1985), and calculations by Abe and Tasaki using the GAAQC method (Abe and Tasaki, 2015).

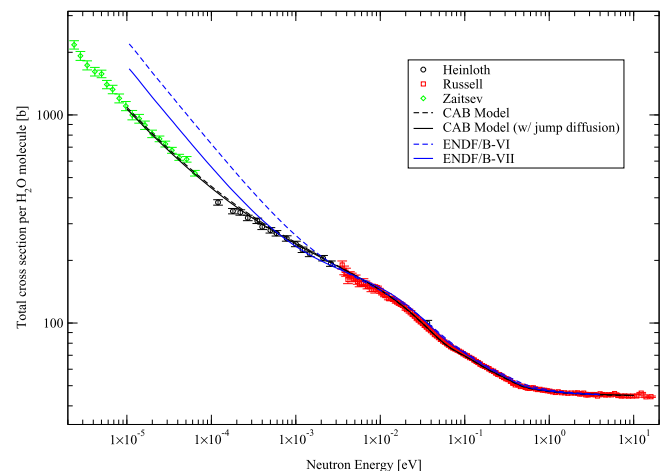


Fig. 4. Total cross section for light water, compared with measurements by Zaitsev et al. (1991), Heinloth (1961), and Russell et al. (1966).

to fail in the quasielastic region. This approximation is part of the module LEAPR of NJOY.

As an alternative to the Gaussian approximation, several groups have developed direct methods for the calculation of the thermal scattering law from molecular dynamics simulations. Due to the intrinsic limitation of classical molecular dynamics simulations, these methods are not completely model-free, and require calculations in the Gaussian approximation to correct and/or complement the results obtained their results.

To overcome the limitations of the Gaussian approximation in LEAPR, we modified the code to introduce a random jump diffusion model. The resulting calculation methodology is a synthesis of modeling and direct calculations: a direct calculation in the classical approximation is used to compute the residence time τ , which is then introduced in the modified model. As an example, we applied this method to light water. The results show that this modification preserves the overall good results obtained with the CAB Model, and extends its validity to the quasielastic region allowing LEAPR to calculate thermal scattering cross sections comparable to the results obtained with the newest developments by Kyoto University.

This method could be applied to any liquid moderator with diffusive translational motion: light and heavy water, liquid methane, liquid ortho and para hydrogen, liquid ortho and para deuterium, etc. Although, in the case of liquid hydrogen and deuterium classical molecular dynamics will not be enough, and other techniques such as centroid molecular dynamics as should be applied (Hone and Voth, 2004).

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