



Molecular dynamics analysis of incoherent neutron scattering from light water via the Van Hove space–time self-correlation function with a new quantum correction



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ABSTRACT

In this paper, we propose a general method for evaluating the neutron incoherent scattering cross-section of light water by molecular dynamics (MD) analysis of the Van Hove space–time self-correlation function (STSCF) with a newly developed quantum correction named as Gaussian approximation-assisted quantum correction (GAAQC). The self-intermediate scattering function (SISF) by GAAQC satisfies the detailed balance condition and sum rules up to second order adequately, and approaches for long times the one obtained directly from MD trajectory data. These features are desirable for the evaluation of the neutron scattering cross-section over a wide range of energy and momentum transfer. From the analysis of quasi-elastic scattering, the self-scattering law by GAAQC shows the jump-diffusive behavior of the molecular translational motion, which is not reproduced by Gaussian approximation (GA). As compared with GA, double differential and total cross-sections by GAAQC show better agreement with experimental data, particularly below the cold neutron region where the non-Gaussian property of the SISF becomes apparent. Thus, the present method, namely, the direct analysis of the STSCF with GAAQC will serve for improving the evaluation of the neutron scattering cross-section for light water.

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1. Introduction

Neutron scattering cross-section data of light water have been extensively utilized for the design and analysis of reactor cores and cold neutron sources. In recent years, they are also referred for the analysis of neutron imaging data from various water-containing materials (Biesdorf et al., 2014; Kiyonagi et al., 2012; Josic et al., 2012; Muhrer et al., 2012; Kawabata et al., 2005). Owing to large incoherent scattering from hydrogen, cold and thermal neutron scattering cross-sections for these materials are dominated by the motion of hydrogen. Thus, any variations of the total neutron cross-section from bulk water, which could be measured by neutron time-of-flight (TOF) imaging (Kardjilov et al., 2003; Kiyonagi et al., 2005), may provide information on the dynamical state of water. Such use of the neutron imaging may bring a new possibility of the nondestructive inspection for water-containing materials. However, dynamical information, which appears principally in the double differential cross-section, tends to be featureless in the total cross-section. Thus, for the better

identification and understanding of the dynamical state of water from the variation of the total cross-section, theoretical analysis of the total cross-section for water would also be necessary as a complementary approach.

According to the Van Hove theory (Van Hove, 1954), the thermal neutron scattering cross-section is represented as the Fourier transformation of the space–time correlation function (STCF) of the particle-density operator for the target system. But in general, because of the many-body problem, determining the exact form of the STCF is difficult. Hence, the existing thermal neutron scattering libraries for various materials (Chadwick, 2006; Mattes and Keinert, 2005) have been evaluated on the basis of physical models, which inevitably involve errors coming from assumptions of the models, and difficulties in determining the value of model parameters over a wide range of temperatures. Therefore, in the previous study (Abe et al., 2014), we applied molecular dynamics (MD) to the analysis of the neutron scattering cross-section for light water for the purpose of improving the evaluation method, and found better agreement with experiments on the total cross-section for cold neutrons in comparison with ENDF/B-VII (Chadwick, 2006). Around the same time, similar results were reported independently by the group at the Bariloche Atomic Centre (Marquez Damian et al., 2013;

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Márquez Damián et al., 2014), and their results also confirmed the effectiveness of the MD-based evaluation.

In both the existing nuclear data libraries and MD-based evaluations mentioned above, Gaussian approximation (GA) is utilized in representing the self-intermediate scattering function (SISF), which is defined as the Fourier transformation in space of the space–time self-correlation function (STSCF). The grounds for GA are based on the fact that the SISF is exactly represented by the Gaussian function of the scattering vector κ for recoil scattering from free atoms at large κ and for quasi-elastic neutron scattering (QENS) from diffusing atoms at small κ . On GA, the width function, which gives the standard deviation of the Gaussian function, is formulated by the frequency distribution function for the target system, allowing the SISF to satisfy the detailed balance condition and sum rules up to second order (Rahman et al., 1962). These properties of the SISF by GA are desirable for applying over a wide region in the κ – t plane, and the calculation of the scattering cross-section by GA is relatively straightforward once the frequency distribution for the target system is given by physical models or MD simulations. Thus, GA has been used for the evaluation of thermal neutron scattering cross-sections.

However, the SISF is not assured to be Gaussian for all ranges of κ . Moreover, as hydrogen bonds among water molecules are relatively strong, the diffusion of water cannot be explained by the continuous diffusion model, but by the jump-diffusion model (Singwi and Sjölander, 1960). In the jump-diffusion model several water molecules are assumed to form a transient network, oscillating around their residence sites between successive jumps which is caused by the collapse of the network due to the molecular thermal motion. Such jump-diffusional motion of water was observed by QENS (Teixeira et al., 1985), where the half width at half maximum (HWHM) of QENS peaks showed suppression from the linear relationship with κ^2 (In case of the continuous diffusion, the HWHM of QENS peaks is proportional to κ^2). The SISF represented by the jump-diffusion model contains a decay constant of the network and becomes a non-Gaussian function of κ . This non-Gaussian behavior of the SISF may influence on the neutron scattering cross-section below cold neutron energies where QENS is significant. Therefore, in order to consider the non-Gaussian property of the SISF adequately, the neutron scattering cross-section for light water should be analyzed directly from the Van Hove STSCF by statistical processing of MD trajectory data.

But there is an essential problem that the STSCF from MD simulations cannot be identified with the one in the Van Hove theory. This is because MD simulation is based on classical mechanics while the Van Hove theory on quantum mechanics. As a result, the SISF from MD simulations do not reproduce quantum effects such as the detailed balance condition and sum rules, and hence must be corrected to satisfy them for the evaluation of neutron scattering cross-sections. Although several quantum corrections (QCs) have been developed in order to recover the detailed balance condition (Schofield, 1960; Egelstaff, 1962; Aamodt et al., 1962), to our knowledge, none of them satisfy high-order sum rules simultaneously. Sum rules up to second order are particularly important for the evaluation of deep inelastic neutron scattering above several eV because the peak position and width of recoil scattering depend on the first and second moments of the self-scattering law.

Thus, the purposes of the present study are to develop a new QC which satisfies both the detailed balance condition and high-order sum rules adequately, and to examine the effectiveness of the direct analysis of the STSCF with the new QC by comparing with the previous results based on GA. The present paper is organized as follows. Chapter 2 describes the method for the evaluation of the neutron scattering cross-section by MD and proposes a new QC. In Chapter 3, calculated results are shown and discussed as

compared with GA and conventional QCs. Finally in Chapter 4, a few concluding remarks are mentioned.

2. The method for the evaluation of the scattering cross-section

2.1. The framework of the present method

Owing to the large incoherent scattering cross-section of hydrogen, the double differential scattering cross-section per molecule can be represented by the incoherent approximation:

$$\frac{d^2\sigma_s}{d\Omega d\varepsilon} = \frac{2}{\hbar} \frac{\sigma_b^H}{4\pi} \sqrt{\frac{E}{E_0}} S_s^H(\kappa, \omega) + \frac{1}{\hbar} \frac{\sigma_b^O}{4\pi} \sqrt{\frac{E}{E_0}} S_s^O(\kappa, \omega), \quad (1)$$

where \hbar is the Planck constant divided by 2π , σ_b^H and σ_b^O are the bound-atom cross-sections for hydrogen and oxygen, $S_s^H(\kappa, \omega)$ and $S_s^O(\kappa, \omega)$ are self-scattering laws for hydrogen and oxygen, and E_0 and E are the energies of the incident and scattered neutrons. The energy and momentum transfers of neutrons are defined as $\hbar\omega = E_0 - E$ and $\hbar\kappa = \hbar|\mathbf{k}_0 - \mathbf{k}|$. Here, \mathbf{k}_0 and \mathbf{k} are the wave vectors of the incident and scattered neutrons. The self-scattering law is defined by

$$S_s^X(\kappa, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{-i\omega t} F_s^X(\kappa, t), \quad (2)$$

where $F_s^X(\kappa, t)$ is the SISF. Note that X denotes hydrogen by X = H or oxygen by X = O.

Instead of GA used in the previous study (Abe et al., 2014), we return to the STSCF $G_s^X(r, t)$ for the evaluation of $F_s^X(\kappa, t)$, where $F_s^X(\kappa, t)$ is defined as the Fourier transformation of $G_s^X(r, t)$ in space \mathbf{r} :

$$F_s^X(\kappa, t) = \int d\mathbf{r} e^{i\kappa \cdot \mathbf{r}} G_s^X(r, t). \quad (3)$$

Based on classical mechanics, $G_s^X(r, t) d\mathbf{r}$ means the probability of finding a X-atom in the thin shell $d\mathbf{r}$ ($= 4\pi r^2 dr$) at a distance r from the origin at time t , provided the same atom exists at the origin at $t = 0$. Therefore, the classical STSCF $\tilde{G}_s^X(r, t)$ can be calculated from MD trajectory data as below:

$$\tilde{G}_s^X(r, t) = \langle \delta(\mathbf{r} - \mathbf{R}_i^X(t + \tau) + \mathbf{R}_i^X(\tau)) \rangle_c, \quad (4)$$

where $\mathbf{R}_i^X(\tau)$ is the position of the i -th X-atom at time τ and the notation $\langle \dots \rangle_c$ denotes the average over time τ and atoms i with keeping the lag-time t constant. Note that the target system is assumed to be isotropic, thus in the left-hand side of Eq. (4) the vector \mathbf{r} is reduced to the scalar r . As $G_s^X(r, t)$ in Eq. (3) cannot be identified with the classical one $\tilde{G}_s^X(r, t)$ in Eq. (4), a QC of $\tilde{G}_s^X(r, t)$ must be carried out for the reconstruction of the scattering cross-section through Eqs. (1)–(3). This will be discussed in the next subsection.

2.2. The quantum correction for the SISF

We begin with the QC of the classical SISF $\tilde{F}_s^X(\kappa, t)$ proposed by Sears (1985), which was intended to fulfill sum rules higher than first order for almost-classical liquid systems. In this correction, $F_s^X(\kappa, t)$ is represented by

$$F_s^X(\kappa, t) = R(\kappa, t) \tilde{F}_s^X(\kappa, t), \quad (5)$$

where $R(\kappa, t)$ is a correction function. From Eq. (2), $S_s^X(\kappa, \omega)$ is represented by the convolution integral between $Q(\kappa, \omega)$ and $\tilde{S}_s^X(\kappa, \omega)$ if we define $Q(\kappa, \omega)$ and $\tilde{S}_s^X(\kappa, \omega)$ as the Fourier transformations of

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