Thermal Neutron Scattering Cross Section Measurements of Heavy Water

G. Li[1], G. Bentoumi[1], Z. Tun[2], L. Li[1] and B. Sur[1]

[1] Nuclear Science Division, Atomic Energy of Canada Limited, Chalk River, Ontario, K0J1J0 Canada

(Corresponding author: G. Li, E-mail: lig@aecl.ca)

[2] Canadian Neutron Beam Centre, Chalk River, Ontario, K0J1J0 Canada

Abstract

In fission reactor design, high accuracy nuclear cross section data for moderator and coolant materials are essential to perform neutronic transport calculations. For the conceptual Gen-IV supercritical water reactor, only very few and sparse experimental nuclear cross section data for water are available in supercritical states. As the first step towards cross section measurements of light and heavy water in the supercritical states, we have established an experimental setup and tested the feasibility of measuring cross sections using a triple-axis Spectrometer at the NRU reactor, by performing measurements on heavy water (D_2O) under ambient conditions. The total cross section σ_{tot}, as well as single ($d\sigma / d\Omega$) and double differential ($d^2\sigma / d\Omega dE$) scattering cross sections were measured. Incident neutron energies from 15 meV to 50 meV, and scattering angles from 10° to 110° were covered. The experimental techniques and the data analysis method to obtain the absolute cross sections will be discussed and the resulting cross sections are compared with the Evaluated Nuclear Data File.

1. Introduction

In a GEN-IV supercritical water reactor (SCWR), operating conditions would be completely different from previous CANDU reactors[1]. The light water coolant will be well above its thermodynamic critical point (T_c ~ 374 °C, p_c ~ 22 MPa), at a pressure of ~25 MPa and a temperature varying from 350 °C (inlet) to 625 °C (outlet). Although the light water is primarily the coolant it also takes part in moderating fission-generated neutrons, especially in the thermal energy range. Therefore the SCWR design needs more experimental data under conditions in where the neutron cross section data have not been measured previously.

At high neutron energies, the cross section is calculated by assuming that nuclei of the propagating medium are isolated particles initially at rest. At neutron energies less than ~1 eV, however, it is necessary to take into account the nucleus' initial energy and the effects of molecular binding. Under supercritical conditions, the experimental data for neutron scattering with water are rare, and their reliability is questionable. Currently, neutronic transport simulations involving supercritical water assume that the hydrogen nucleus is unbound, an approximation known as the "free gas model". New experimental data are undoubtedly required to test the range of validity of that model. Simulations have found that the cross section data of deuterium has a significant impact on the neutron multiplication factor and reactivity coefficient, for the conceptual Canadian Supercritical Water Reactor[2]. Even under normal conditions, new

experimental data are still desirable today, in order to investigate the discrepancy between evaluations and existing data[3].

The majority of the work on thermal neutron scattering was performed in the last century during the fifties and sixties. Since then very few measurements have been done to re-evaluate neutron cross sections. This paper describes an experiment measuring the neutron scattering cross section of D_2O under ambient conditions using a triple-axis spectrometer (TAS). The experimental procedure, data normalization method and systematic uncertainties of the TAS will be discussed. The results will be presented and some discrepancies between existing data and models are investigated.

2. Experimental Setup

The experiment was carried out at the neutron scattering facilities of AECL's NRU reactor using the TAS on the N5 beamline. The schematic layout of the instrument configuration is shown in Figure 1. The "three axes" refer to a set of rotation axes at the monochromator, at the sample, and finally at the analyzer. The angular settings for all axes are accurate to within ±0.01°.

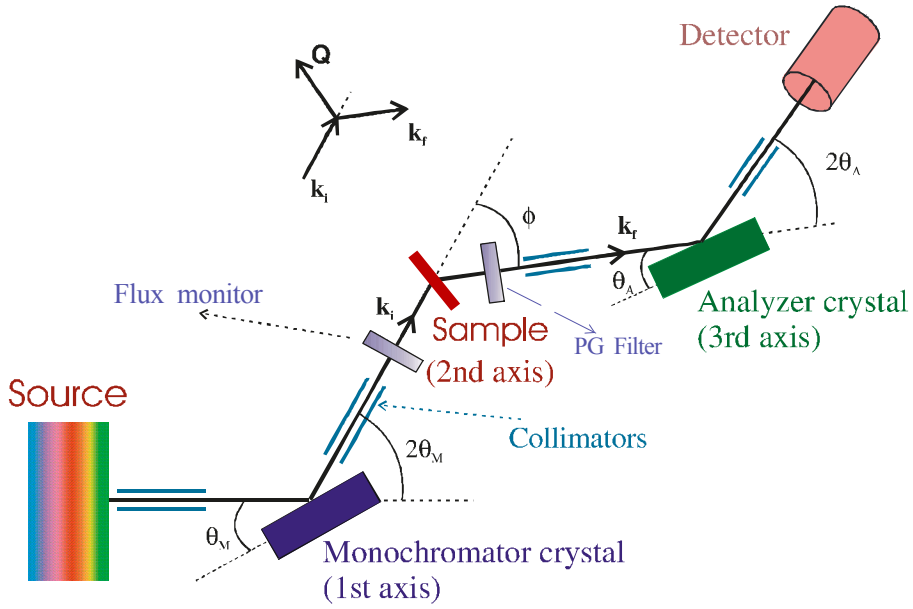

Figure 1: Schematic drawing of the triple-axis spectrometer. The monochromator crystal at the first axis selects neutrons with a specific energy, providing a monochromatic beam. The analyzer crystal at the third axis defines the final energy. Figure is taken from Ref.[4] and modified to show the actual set up.

The incoming beam energy is determined by the Bragg angle of the monochromator crystal. The monochromator was single-crystal silicon oriented for Bragg diffraction from (111) planes. To monitor the neutron flux, the incident beam passed through a ^{235}U fission chamber operated in

pulse mode. The detection efficiency of the monitor is on the order of 10^{-4}, and is inversely proportional to the neutron speed throughout the energy range of interest.

The heavy water sample was held between two pure silicon plates of 1 mm thick and 10 cm in diameter, sealed with a rubber O-ring. The silicon plates are virtually transparent to neutrons. The heavy water sample was kept at the thickness of 2.9 mm and at this thickness, the neutron beam has about 85~90% transmission. The selection of the sample thickness is a trade-off between gaining signal intensity and avoiding multiple scattering.

The scattered neutron energy was defined by Bragg reflection from the analyzer crystal, which was pyrolytic graphite (PG) oriented for Bragg diffraction from (002) planes. The scattered neutrons from the analyzer were detected with a ^{3}He detector, which has a detection efficiency >95% at all energies of interest.

3. Experimental Procedure and Results

3.1 Total Cross Section σ_{tot}

Neutron total cross sections were measured for heavy water with beam energies from 12 meV to 50 meV. Within this energy region, there is a known discrepancy between the evaluated total cross section and existing data[5]. The total cross section can be calculated from the transmission T_{tran} by equation

$$T_{tran} = \exp(-\sigma_{tot} \rho \, x) \qquad (1)$$

in which ρ is the number density of the sample and x is the sample thickness. The measured cross sections are shown in Figure 2, along with previously published data[6] and the line shape of the total cross section modeled at Institute for Nuclear Technology and Energy Systems (IKE)[5]. The uncertainty of this measurement is dominated by the sample-thickness measurement and relative uncertainty is estimated to be within 4%. This measurement confirms the discrepancy between the IKE evaluation and the existing data.

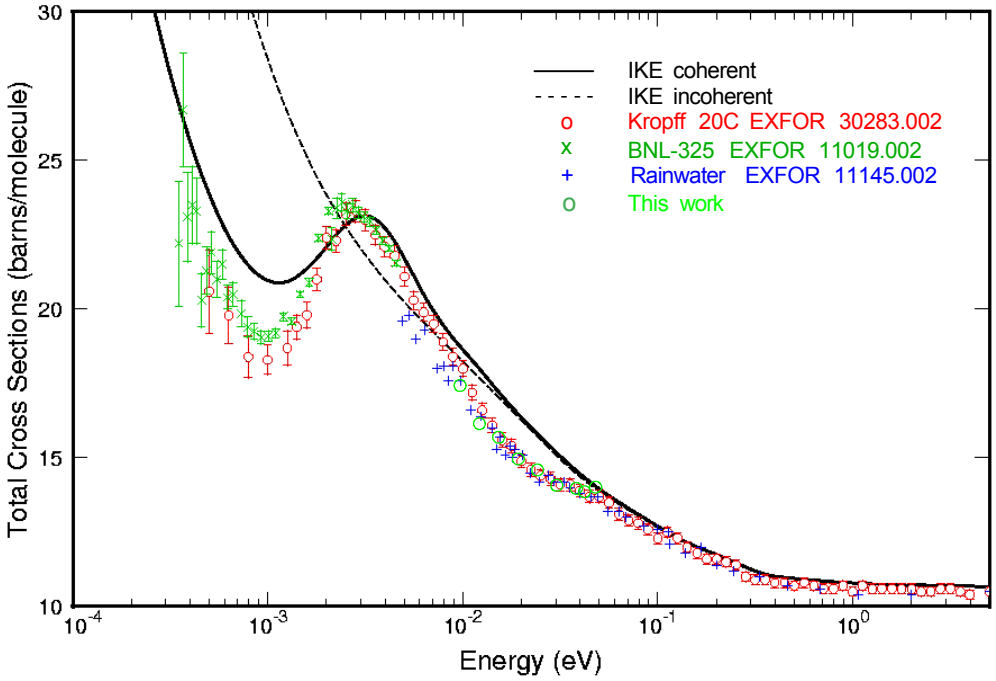

Figure 2. Total cross section of D_2O. The solid and dashed lines are the IKE evaluations[5], the green circles are the data from this experiment, and other data points are from Ref.[6]. The figure of previous data and IKE evaluations are from Ref.[5].

3.2 Single Differential Scattering Cross Section $d\sigma / d\Omega$

The single differential cross sections for heavy water were measured at beam energies of 41.44 meV and for scattering angles of 10° to 110°, in 5° steps. By definition, the count rate C is related to the cross section as follows:

$$C = \frac{d\sigma}{d\Omega} I \cdot \rho \cdot S \cdot x \cdot A_T \, \Delta\Omega \qquad (2)$$

in which I is the incident beam intensity (per unit time), ρ is the number density of the target material, S is the area of the beam, x is the sample material thickness along the beam direction, A_T is the attenuation factor due to all interactions between neutrons and the target material, and $\Delta\Omega$ is the detector acceptance angle. Vanadium was used as the standard because of its complete elastic incoherent scattering property, and the cross section of water can be calculated as follows:

$$\frac{d\sigma_{D2O}}{d\Omega} = \frac{\sigma_V}{4\pi} \frac{\rho_V}{\rho_{D2O}} \frac{x_V}{x_{D2O}} \frac{A_{TV}}{A_{TD2O}} \frac{C_{D2O}}{C_V}, \qquad (3)$$

The subscript V and $D2O$ refer to vanadium and D_2O, respectively. The attenuation factor A_T which describes attenuation of the beam passing through the sample is:

$$A_T = \exp(-\sigma_{tot} \cdot \rho \cdot x) \, . \qquad (4)$$

The results for the differential scattering cross section of D_2O at beam energy of 44.0 meV are shown in Figure 3, compared with the previous measurement[7] and the IKE evaluation[5]. The result has good agreement with the data measured by Springer[7]. The uncertainties are dominated by (1) the background subtraction, and (2) the finite beam size and detector acceptance angle. This correction is small if the differential cross section changes slowly with scattering angle. However, around the coherent peak of $d\sigma/d\Omega$, this correction is significant.

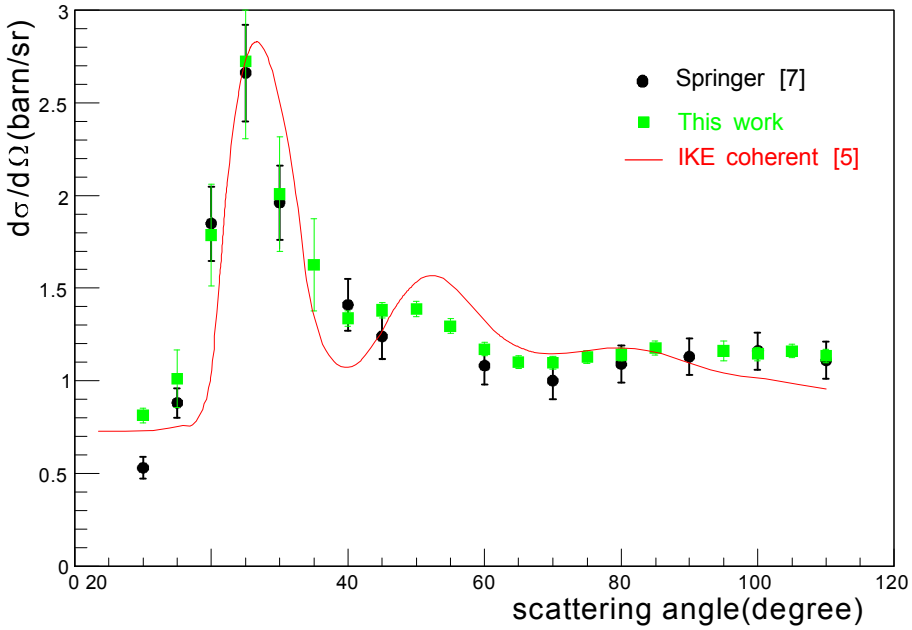

Figure 3. Differential scattering cross section of D_2O. Beam energy is 44.0 meV. To compare our results with other data, the measurement by Springer[7] and the IKE evaluation[5] are also plotted.

3.3 Double Differential Scattering Cross Section $d^2\sigma/d\Omega dE$

The neutron scattering cross section of water is usually represented by the scattering law $S(\alpha,\beta)$ [8], which is related to the double-differential cross section as follows:

$$\frac{d^2\sigma_D(E_1 \to E_2,\Omega)}{d\Omega dE} = \frac{\sigma_b}{4\pi kT}\sqrt{\frac{E_2}{E_1}}e^{-\frac{\beta}{2}}S(\alpha,\beta). \qquad (5)$$

Where α and β are dimensionless quantities related to momentum and energy transfer respectively, defined as

$$\alpha = \frac{\Delta p^2}{2MkT} = \frac{m(E_1 + E_2 - 2\sqrt{E_1E_2}\cos\theta)}{MkT}, \quad \beta = \frac{E_2 - E_1}{kT} \qquad (6)$$

in which m is the neutron mass, M is the mass of the principal scattering atom (the deuterium nucleus, in the case of D_2O), θ is the scattering angle, σ_b is the bound total scattering cross section of the principal scattering atom, k is the Boltzmann constant, T is the sample temperature, E_1 and E_2 are the incident beam energy and scattered neutron energy, respectively. In the scattering law representation, the values of $S(\alpha,\beta)$ depend only on the neutron energy transfer

and momentum transfer, which facilitates the comparison between various experiments with different beam energies.

The double differential cross section measurements were performed for energy transfer from 42.00 meV to 14.56 meV ($\beta = -1.1$), and for scattering angles ranging from 10° to 110°. The absolute scattering cross section of water was measured relative to the vanadium, which is a pure incoherent elastic scatterer. Using vanadium as the standard, the scattering law of the deuterium atom in the heavy water can be expressed as[9]

$$S(\alpha,\beta) = \frac{1}{\sqrt{2\pi}} \frac{kT}{\Delta E} \frac{\sigma_V n_V C_{\mathrm{D_2O}(E_1 \to E_2)}}{\sigma_{Db} n_D C_{V(E_1=E_2)}} e^{\frac{\beta}{2}} \tag{7}$$

in which $C_{\mathrm{D_2O}(E_1 \to E_2)}$ and $C_{V(E_1=E_2)}$ are the counts from the water and vanadium scattering, respectively, and ΔE is the energy resolution of the system measured by vanadium. The validity of Eq. (7) assumes that the resolution function of the system is Gaussian, and the variation of the inelastic scattering cross section is small over the system resolution. The resolution was measured by varying the incident neutron beam energy while fixing the scattered neutron energy. The resulting spectrum can be approximated by a Gaussian distribution, as shown in Figure 4. The overall energy resolution of the spectrometer was 1.2 meV (FWHM) at the scattered neutron energy of 14.6 meV.

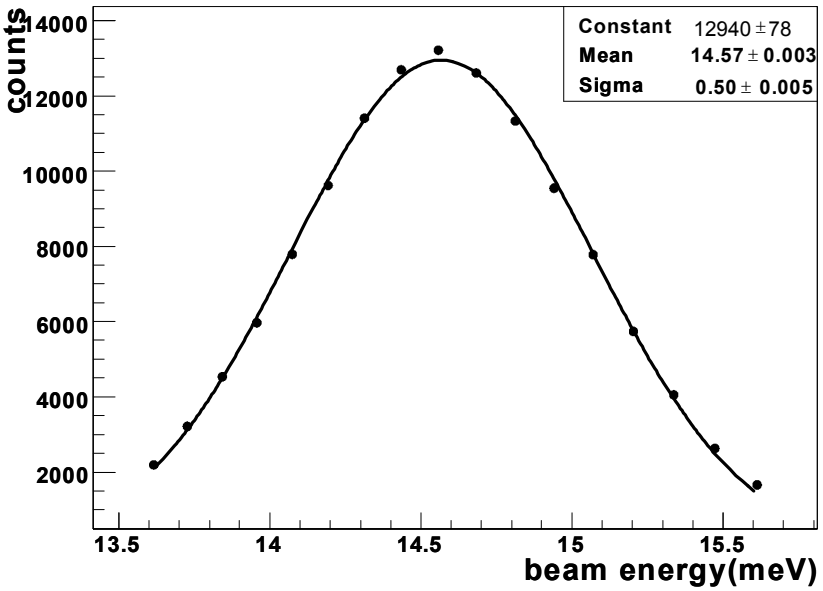

Figure 4: Vanadium spectrum, measured by scanning the incident beam energy while keeping the analyzer set at 14.6 meV. The resolution of the spectrum represents the overall resolution of the triple-axis spectrometer. The parameters of the fitted Gaussian distribution are shown in the text box.

The scattering law measurement of D_2O at the energy transfer of 42.00→14.56 meV is presented

in Figure 5. The results shown in this figure are calculated directly using Eq. (7). Our measurement is generally consistent with the measurement performed by Harling[10], but there is a noticeable discrepancy between these measurements and the ENDF/B-VII.1 evaluation[11]. Note that multiple scattering and oxygen scattering corrections have not been applied in these three measurements. The final results from Harling[10] did not include these corrections, although Harling recognized that multiple scattering could contribute as much as 25% for small angle and large energy transfer scatterings[10]. In this paper, the multiple scattering and the oxygen scattering are considered and will be discussed in the following sections.

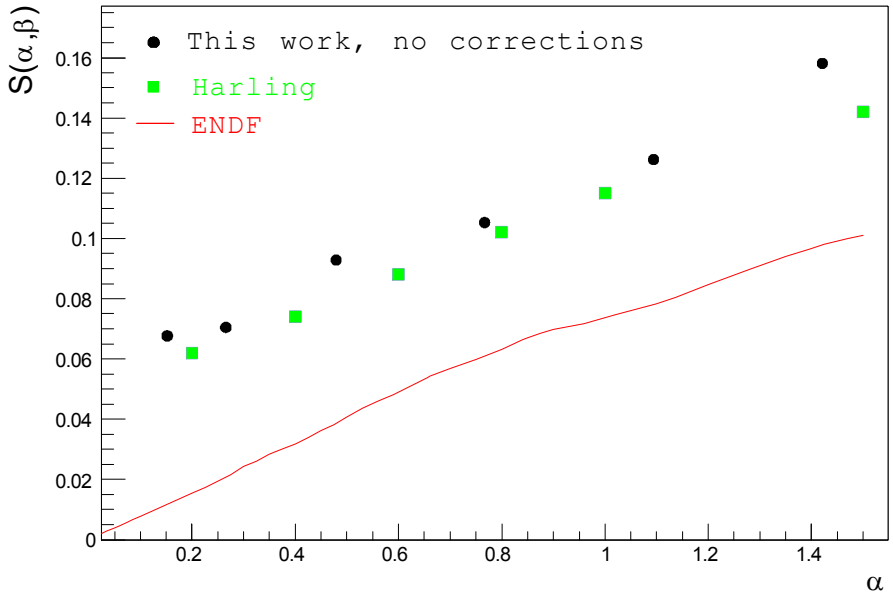

Figure 5. Measured scattering law of D atoms in D_2O, compared with ENDF. The black circle is the result from N5, the green square is the measurement by Harling[10], and the red line is the ENDF/B-VII.1 evaluation[11]. Note that multiple scattering correction and contribution from oxygen has not been considered.

Multiple scattering

The transmission is about 85%-90% in this experiment, which is a typical value used in inelastic scattering cross section measurements. However, the multiple scattering could still contribute a significant correction for measurements at some particular energy transfers, in the sense that the cross section with large energy transfer could be orders of magnitude lower than that with lower energy transfer, so the multiple scattering from small energy transfer could be comparable with real signals from large energy transfer scatterings.

Geant4[12] is a Monte Carlo program simulating particle interactions and transportations. In the Geant4 simulation, the information of how many scatterings occurred before neutrons reach the detector is available. A Geant4 model was built to study the multiple scattering effect. In this model, a D_2O target with the same geometry as the experiment was built and oriented 45°

relative to the neutron beam of 42 meV. For all outgoing neutrons with energy around 14.6 meV, the number of scattering which occurred in the target was retrieved and plotted in the histogram in Figure 6. The simulation indicates that at this energy transfer, which is the same as the energy transfer in Figure 5, about 32% events are from multiple scattered neutrons. Since multiple scattering tends to have no structure, the 32% events coming from multiple scattering are assumed to have a flat distribution, and subtracted from the signals before applying Eq. (7).

The version 9.6p0 of Geant4 was used in this study. In Geant4, the $S(\alpha,\beta)$ matrix has been recently included in the "G4NeutronHPThermalScattering" model for thermal neutron scattering simulations with common moderator materials [12], [13]. However, the reliability has not been widely evaluated. To account for the uncertainty of the multiple scattering correction using Geant4, another physical model of Geant4 (G4NeutronHPElastic) was tested. The difference between the calculated corrections using these two models is about 8%, and is quoted as the uncertainty from the Geant4 simulation.

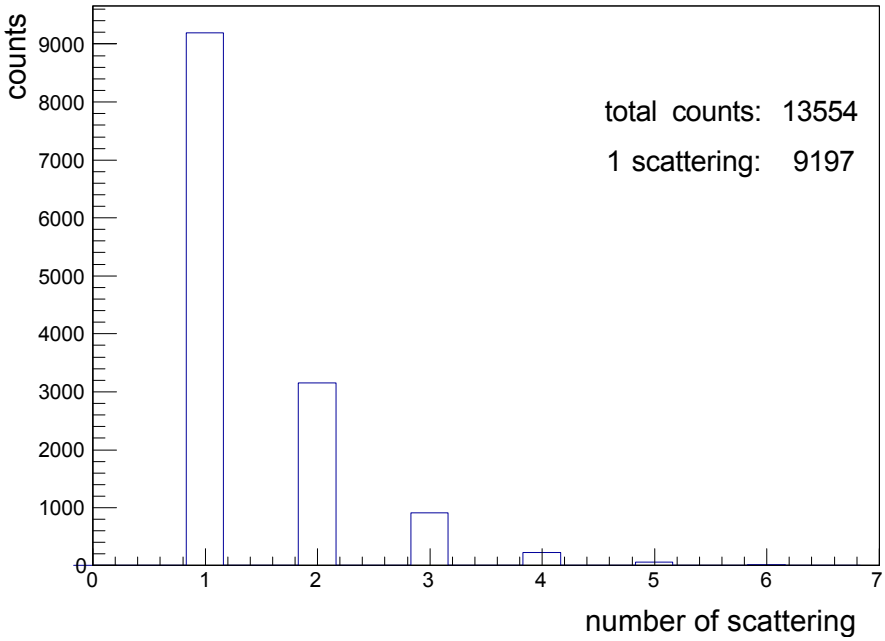

Figure 6. The histogram of the number of neutron scatterings in sample before entering detectors, simulated using Geant4. The simulation uses a 42 meV neutron beam, scattered into 14 ~ 15 meV, by a 2.9 mm D_2O sample.

Oxygen scattering

In the D_2O, the total scattering cross sections of oxygen ($\sigma_O = 4.23\ barn$) and deuterium ($\sigma_D = 7.64\ barn$) are comparable. However, most of previous experiments ignore the effect of oxygen scattering, as Egelstaff stated that the major contribution will be from the deuterium scattering, although there will be some contributions from oxygen atoms[14]. ENDF/B-VII uses

the free gas approximation to calculate the scattering from oxygen atoms in D_2O[5]. This is the approach to be used in the analysis of our experiment, since our results are to be compared to ENDF/B-VII evaluation. In the free gas approximation, the frequency distribution is given by a delta function, and the scattering law is written as follows[5]:

$$S(\alpha,\beta)=\frac{e^{-\frac{\alpha^2+\beta^2}{4\alpha}}}{\sqrt{4\pi\alpha}}. \qquad (8)$$

The scattering from oxygen is calculated using Eq. (8), and this contribution is subtracted from the results calculated directly using Eq.(7).

After applying the corrections of the multiple scattering and the oxygen scattering, the final results, presented in Figure 7, are generally consistent with the ENDF evaluation. The uncertainty includes the statistical uncertainties (typically $10\%-20\%$), the sample thickness uncertainty (typically 3%) and the uncertainty from the multiple-scattering correction (typically 8%). It is noticeable that the results are too high at small α, and too low at large α, relative to the ENDF. This is most probably due to the simplified assumption that the multiple scattering is totally isotropic, which is not true for a slab sample geometry. For the slab geometry, the multiply scattered neutrons tend to be detected along the normal direction of the slab surface, which should give a larger correction at small angles.

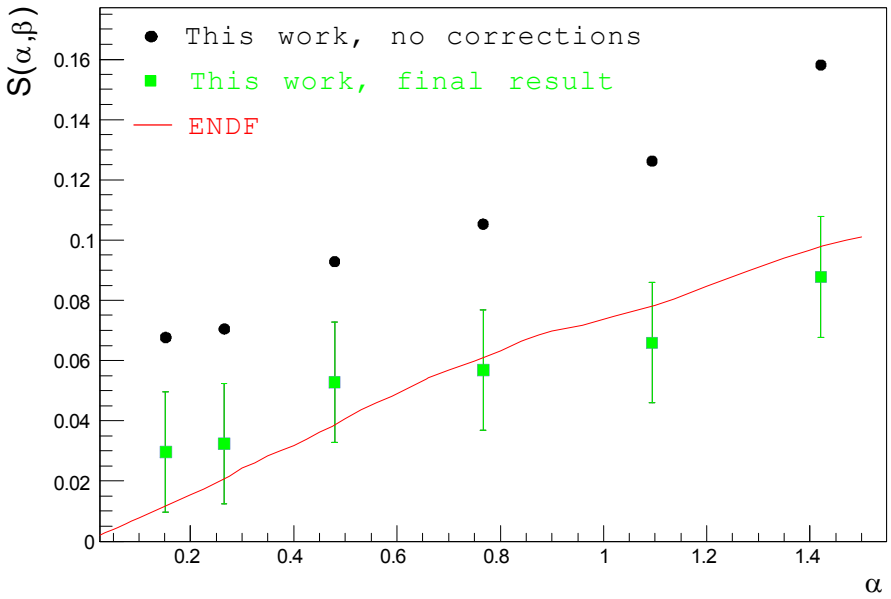

Figure 7. Final results of the scattering law of D_2O, compared with ENDF/B-VII.1. The black circles are the result measured without correcting multiple scattering and oxygen scattering. The squares are the result with multiple scattering and oxygen scattering subtracted.

4. Conclusion and outlook

The thermal neutron scattering cross section measurements of heavy water under ambient conditions have been performed. The measured total cross section σ_{tot}, differential scattering cross section $d\sigma/d\Omega$ and double differential scattering cross section (in the S(α,β) representation) are consistent with published data. The feasibility of using the triple-axis spectrometer to measure neutron scattering cross sections is demonstrated. The multiple-scattering effect is found to be the most important correction in the data analysis. The thermal neutron transportation simulation using Geant4 is to be further evaluated and the multiple scattering studies using other Monte Carlo codes (e.g. MCNP) are desirable. Future experiments should avoid too much multiple scattering by using a thinner sample and proper selection of beam energies and scattered neutron energies.

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