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A Computer Code DEUKER for D₂O Scattering Cross Section

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Abstract

Based on the Butler scattering kernel for D_2O, a computer code DEUKER has been developed to compute the scattering laws, differential scattering cross sections and total scattering cross sections. Interference scattering between any two atoms of a D_2O molecule is important in resolving the distribution of scattered neutrons in thermal energy region. Energy-transfer scattering cross sections are, therefore, studied in the various incident neutron energies. This study may be put in practice to utilize the kernel in determining the neutron spectrum in a reactor system. The study also shows that the scattering process in D_2O is somewhat different from that in H_2O.

요   약

D_2O에 대한 Butler의 scattering kernel에 관해 조사하였다. 이 kernel을 사용한 전자계산 코드 DEUKER를 개발하였으며 이를 이용하여 산란법칙, 미분산란 단면적 및 전산란 단면적을 계산하였다. D_2O 분자내의 어떤 두개 원자간의 산란 영향은 열에너지 영역에서의 산란 중성자 분포를 결정하는데 있어서 중요한 인자가 된다. 따라서 여러 입사 중성자 에너지에 따른 에너지 전달 산란 단면적에 관해 조사하여 이 kernel의 원자로 내의 중성자 분포 계산을 위한 사용 가능성을 연구하였다. 또한 본 연구를 통하여 D_2O와 H_2O에서의 중성자 산란 파장의 차이점을 밝혔다.

I. Introduction

In solving the Boltzmann equation of neutron transport, a closed form of the neutron scattering kernel in a moderator system is required to calculate thermal neutron spectrum and a large number of physical properties. Therefore, there are many scattering kernels, for example, Nelkin model for light water system and syn-thetic gas kernels concerning with gas dynamics. Before utilizing a scattering kernel proposed for a typical moderator system, the kernel should be examined whether the total scattering cross section can be properly estimated in terms of the incident neutron energy. Especially in a chemically bound molecule such as D_2O, the computation of thermal neutron scattering cross sections requires a wide range of physical knowledge pertaining to the dynamics of the

bound atoms.

Butler[1] proposed a tractable model for thermal neutron scattering by D_2O molecule. Unlike in the case of the light water approximation, the scattering in deuterium is largely coherent, that is, $\sigma_{sb}=7.6b$ and $\sigma_{coh}=5.4b$, which are bound and coherent scattering cross section respectively. Therefore, interference effect between any two atoms in D_2O molecule may play an important role in scattering process.

Due to the complexity of the Butler model for D_2O scattering with neutrons, there is little investigation on the kernel in order to apply to the Boltzmann equation of neutron transport. Before making use of the kernel into neutron transport equation, a detailed study on the kernel has been carried out in this paper and a computer code DEUKER is developed to produce numerical values for the physical properties of D_2O such as the scattering laws. Differential scattering cross sections are evaluated to examine the scattering behavior in D_2O moderator. Total scattering cross sections computed from the code are compared with the experimental results of BNL-325[2]

The scattering model is presented with physical parameters and the definitions of quantities in section Ⅱ. The subsequent section details numerical methods and DEUKER program. The computed results are discussed in the last section.

Ⅱ. Scattering Kernel

Scattering process of thermal neutron in a chemically bound molecule involves various modes associated with energy exchange and transport properties of bound atoms. In describing neutron scattering cross section in D_2O, three basic modes of the Butler model are based on the following assumptions:

1) translation is considered to be the similar process to the case of free gas model

2) vibration is analyzed into D_2O molecular modes and frequencies are experimentally taken from spectrographic data

3) rotation is approximated by torsional oscillation, according to the experimental result[3], the phenomenon is appeared to be severely hindered.

As a neutron with the incident energy E_0 is scattered into D_2O molecule, the differential scattering cross section is expressed[1] by

$$\sigma_{\nu\nu'}(E_0 \rightarrow E, \theta) = \frac{(A_\nu A_{\nu'} + \delta_{\nu\nu'} C_\nu^2)}{T}$$

$$\frac{\sin b_{\nu\nu'}\sqrt{\alpha}}{b_{\nu\nu'}\sqrt{\alpha}} \left(\frac{E}{E_0}\right)^{\frac{1}{2}} \exp\left(-\frac{\beta}{2}\right) S_{\nu\nu'}(\alpha, \beta) \tag{1}$$

where E is the final neutron energy and θ the scattering angle between the incident and scattered neutron wave vectors. The subscripts, ν and ν', indicate the individual nuclei in scattering system of a single molecule, and A_ν and C_ν represent the coherent and incoherent scattering amplitudes of nucleus ν, respectively. T is the system temperature measured in units of energy.

The conversion factor $b_{\nu\nu'}$ is expressed in terms of the equilibrium position vector of the ν-th and ν'-th nucleus with respect to the molecular center of mass:

$$b_{\nu\nu'} = \left(\frac{2MT}{\hbar}\right)^{\frac{1}{2}} |\vec{b}_\nu - \vec{b}_{\nu'}| \tag{2}$$

where $\hbar$ is Plank's constant divided by 2π, and M the mass of a single molecule. The other quantities are:

$$\alpha = \frac{m}{MT}(E + E_0 - 2\sqrt{EE_0} \cos \theta):$$

momentum transfer   (3)

$$\beta = \frac{E - E_0}{T}: \text{ energy transfer} \tag{4}$$

where m is the mass of neutron. Together with the quantities α and β, $S_{\nu\nu'}(\alpha, \beta)$ in Eq. (1) is representative of the scattering function expressed as:

$$S_{\nu\nu'}(\alpha, \beta) = \frac{1}{2\sqrt{\pi\alpha}} \exp\left[-\left(\lambda_{\nu\nu'}+\frac{1}{4}\right)\alpha\right] \times$$

$$\sum_{n1=-\infty}^{\infty} \sum_{n2=-\infty}^{\infty} \sum_{n3=-\infty}^{\infty} \times$$

$$\exp\left[-\frac{\left(\beta-\sum_{r=1}^{3} n_r\beta_r\right)^2}{4\alpha}\right] \prod_{r=1}^{3} I_{nr}\left(\frac{\alpha}{B_r^{\nu\nu'}}\right) (5)$$

The $\lambda^{\nu\nu'}$ and $B_r^{\nu\nu'}$ are defined as

$$\lambda^{\nu\nu'} = \frac{M}{m}\sum_{r=1}^{3} P_r^{\nu\nu'}\frac{\coth\frac{1}{2}\beta_r}{\beta_r} \qquad (6)$$

and

$$\frac{1}{B_r^{\nu\nu'}} = \frac{M}{m}\frac{Q_r^{\nu\nu'}}{\sinh\frac{1}{2}\beta_r} \qquad (7)$$

where $\beta_r = \frac{\hbar\omega_r}{T}$ and $I(x)$ is modified Bessel function. The subscript $r=1$ indicates the rotational mode, and 2 and 3 stand for the vibrational modes. The $P_r^{\nu\nu}$ and $Q_r^{\nu\nu}$ are rotational and vibrational parameters.

In computing the total scattering cross section at the incident neutron energy E_0 in D₂O, there are four distinct nucleus combinations such as D-D, O-O, D-D' and O-D. Thus define the individual contributions as

$$\sigma_{\nu\nu'}(E_0) = 2\pi \int dE \int \sigma_{\nu\nu'}(E_0 \rightarrow E, \theta) d(\cos\theta) \qquad (8)$$

and the total scattering cross section at E_0 is

given by

$$\sigma(E_0) = \sum_{\nu\nu'}\sigma_{\nu\nu'}(E_0) \qquad (9)$$

II. Numerical Calculations

In principle, the cross section can be computed by an explicit summation over contributions from all the scattering processes that occur. But the large number of states and energetically permissible quantum states of the molecule will make such a procedure impractical. Thus implicit methods of performing the summatons required must be sought with taking the contribution of all transitions permitted by the conservation laws.

The following parameters are adopted[1] for estimating D₂O scattering cross sections:

—the distance between the oxygen atom O and a deuteron D or $D'=1.01$Å

—the angle between the two OD bonds=105°

—bound scattering cross sections are $4\pi(A_D^2+C_D^2)$ =7.6 barns, $4 A_O^2=4.24$ barns, $C_O=0$, $4\pi A_O A_D=4.76$ barns and $4\pi A_D^2=5.4$ barns,

—the three excitation levels are taken to be 0.048 eV, 0.146 eV, and 0.3385 eV.

Table 1. Numerical Values of $P_r^{\nu\nu'}$ and $Q_r^{\nu\nu'}$

Quantities	r=1	r=2	r=3
P_r^{DD}	0.22800	0.741354×10^{-1}	0.147868
P_r^{OO}	0.550086×10^{-2}	0.229948×10^{-2}	0.469966×10^{-2}
P_r^{OD}	0.116749	0.382175×10^{-1}	0.762839×10^{-1}
$Q_r^{DD'}$	-0.519691×10^{-1}	-0.551989×10^{-3}	0.252109×10^{-2}
Q_r^{OD}	-0.220035×10^{-1}	-0.919793×10^{-2}	-0.187987×10^{-1}

Table 2. Numerical Values of $\lambda^{\nu\nu'}$ and $B_r^{\nu\nu'}$

Quantities	DD	OO	DD'	OD
$\lambda^{\nu\nu'}$	3.731213	9.349647×10^{-2}	3.731213	1.912355
$B_1^{\nu\nu'}$	0.456603	1.892503×10	-2.003190	-4.731258
$B_2^{\nu\nu'}$	3.474508×10	1.120183×10^3	-4.666473×10^3	-2.800456×10^2
$B_3^{\nu\nu'}$	1.818918×10^3	5.722916×10^4	1.066842×10^5	-1.430740×10^4

The calculations are performed at the system temperature $T=0.0253$ eV, and the calculated values of rotational and vibrational parameters $P_r^{\nu\nu'}$ and $Q_r^{\nu\nu'}$ are presented in Table 1.

From these values, the $\lambda^{\nu\nu'}$ and $B_r^{\nu\nu'}$ are also obtained and given in Table 2.

Evaluation of $S(\alpha, \beta)$ in Eq. (5) is suited to break down the threefold sum into a double sum over a quantity $S(n_2, n_3; \alpha, \beta)$:

$$S(\alpha, \beta) = \sum_{n_2=-\infty}^{\infty} \sum_{n_3=-\infty}^{\infty} S(n_2, n_3: \alpha, \beta) \quad (10)$$

with

$$S(n_2, n_3: \alpha, \beta) = \frac{1}{2\sqrt{\pi\alpha}}$$
$$\times \exp\left(-(\lambda+\tfrac{1}{2})\alpha\right) In_2\left(\frac{\alpha}{B_2}\right) In_3\left(\frac{\alpha}{B_3}\right)$$
$$\times \sum_{n_1=-\infty}^{\infty} \exp\left[-\frac{(\beta-n_3\beta_3-n_2\beta_2-n_1\beta_1)^2}{4\alpha}\right] In_1\left(\frac{\alpha}{B_1}\right)$$

(11)

The subscripts and superscripts, ν and ν', in the previous equations are dropped from now on for convenience. Although individual sums in the scattering function run over from $-\infty$ to ∞, the practical sum will be truncated in several terms judging by the significance. After making a summation up to a certain number n_1, it is found that the quantity $S(n_2, n_3: \alpha, \beta)$ oscillates monotonically and dies out rapidly for increasing n_1. Likely, only a few $S(n_2, n_3: \alpha, \beta)$ terms are found to be significant in the sum over n_2 and n_3 to form $S(\alpha, \beta)$. All the values of the scattering law $S(\alpha, \beta)$ are calculated with the accuracy of relative error 10^{-4}.
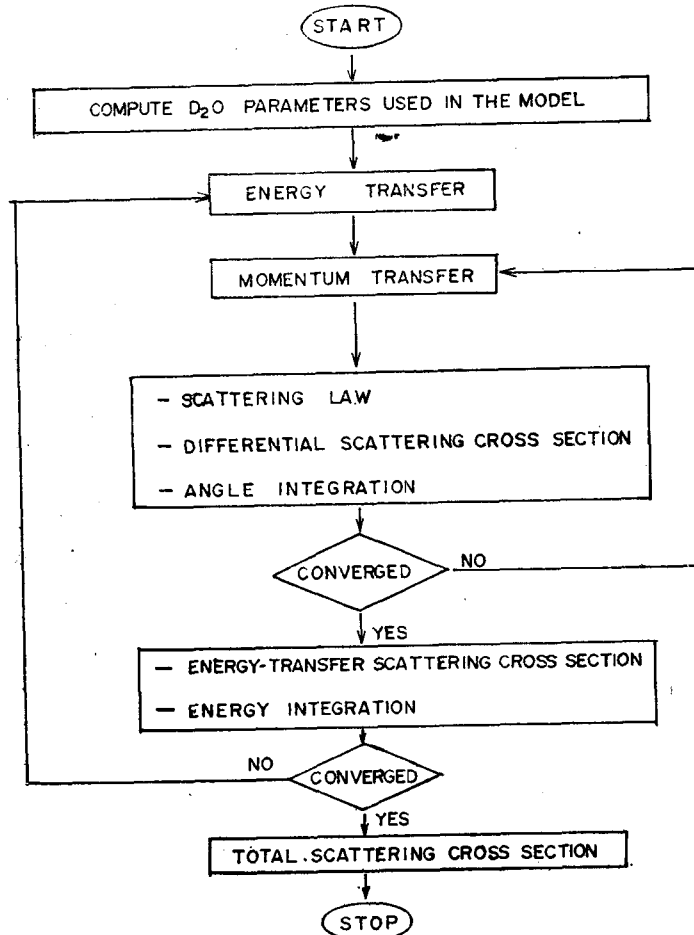
For the convenience of numerical computa-

Fig. 1. Flow Diagram of the Program DEUKER

Table 3. Comparisons of Scattering Cross Secations in D_2O between Calculation and Experiment

Incident Neutron Energy (E_0) eV	$\sigma_t(E_0)$ from BNL-325	$\sigma_t(E_0)$ (barns)	$2\sigma_{DD}$ (barns)	σ_{OO} (barns)	$2\sigma_{DD}'$ (barns)	$4\sigma_{OD}$ (barns)	Remark
0.0253	14.6	15.61	11.04	3.92	0.19	0.46	Present
		15.64	11.02	3.94	0.23	0.45	Butler
0.2277	11.2	11.51	7.62	3.79	0.02	0.08	Present
		11.4	7.62	3.78	—	—	Butler

tions, the neutron energy is treated in terms of neutron speed v in dimensionless unit. In doing so, when neutron energy differs from the system temperature, neutron energy can be described by the following equation,

$$E = Tv^2 \qquad (12)$$

where v is a dimensionless variable and the system temperature T is measured in unit of energy. It can be readily seen from Eq. (12) that neutron speed v is unity when neutron energy is equal to the system temperature.

The energy-transfer differential scattering cross section can be obtained by

$$\sigma(E_0 \to E) = 2\pi \int_{-1}^{+1} \sigma(E_0 \to E, \mu) d\mu \qquad (13)$$

where $\mu = \cos\theta$. The angle integral in Eq. (13) can be approximated by using the Gaussian Legendre quadrature[4,5]. Most of the relative error is in the range of 10^{-4} at the number of nodes taken up to 16. Therefore, the maximum node is adopted to be 16 in computing the values of $\sigma(E_0 \to E)$.

The scattering cross section for a given initial neutron energy E_0 is given by

$$\sigma(E_0) = \int_0^\infty \sigma(E_0 \to E) dE. \qquad (14)$$

The numerical evaluation of the integral in Eq. (14) is made by Simpson's rule[5,6]. Though the range of the scattered neutron energy is from 0 up to ∞, it is convenient that the adequate upper and lower limits are determined by truncating the negligible values of σ $(E_0 \to E)$ in the numerical evaluation of the integral. Likely, after evaluating the values of $\sigma(E_0 \to E)$

with a given neutron energy interval, the integrations are repeated with the interpolation of the computed values $\sigma(E_0 \to E)$ by halving of an initial width until the results satisfy a convergence criterion of 10^{-3}. The calculation procedure is given in Fig. 1, which is the flow diagram of the computer program DEUKER.

V. Results and Discussions

In computing thermal neutron scattering cross sections in D_2O moderator, individual cross sections are estimated at the incident neutron energies of 0.0253 and 0.2277 eV and the results are presented in Table 3.

It can be seen from the table that the effects of the direct terms, $D—D$ and $O—O$ bond, arising from deuteron and oxygen atom to the total scattering cross section are appeared to be very significant. On the other hand, the interference terms formed by any two atoms of the same molecule make a little contribution to the total cross section. It can also be deduced from the calculation that at low neutron energy the interference effects would be no longer possible to be ignored to compute the total scattering cross section. Furthermore, it should be noted[1] that the interference effects should be properly treated to evaluate the average cosine of the scattering angle.

The normalized differential scattering cross sections for thermal deuteron and oxygen atom are computed and illustrated in the figures 2 and
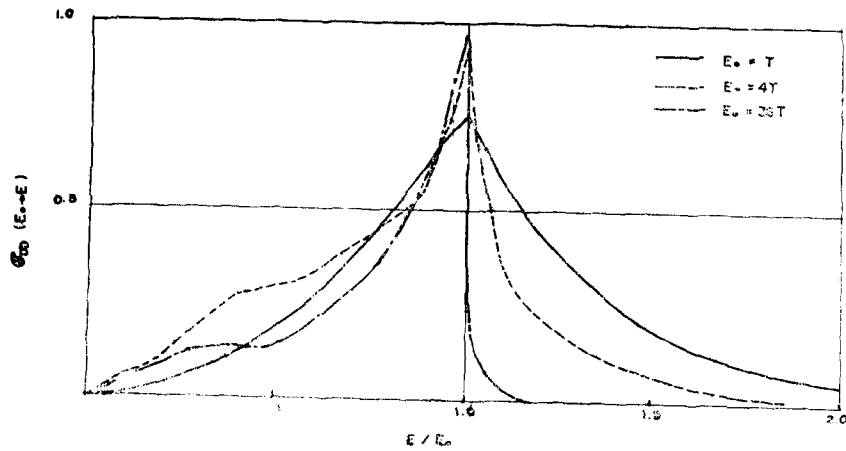
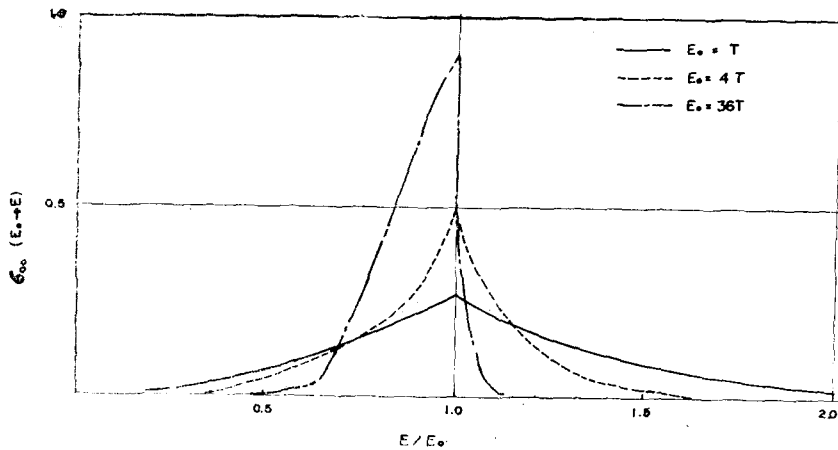Fig. 2. The Normalized Energy-Transfer Cross Section of a Deuteron Atom

Fig. 3. The Normalized Energy-Transfer Cross Section of an Oxygen Atom

3, respectively.

At the low incident neutron energy, $E_0 = T$, in Fig. 2, the differential scattering cross section is almost evenly distributed throughout up-and down-scattering region. As increasing incident neutron energy, the distribution is somewhat shifted toward down-scattering region, that is, it is much less probable for a neutron to be up-scattered than down-scattered. In the case of the light water system[7], thermal hydrogen, however, is noticeable in down-scattering region in which neutron collision with hydrogen atom will end up with energy loss and the probability for collisions with an energy gain becomes smaller.

The differential scattering cross section for thermal oxygen atom is also well distributed at the low incident neutron energy, $E_0 = T$, as shown in Fig. 3. Up-scattering of neutron changes very rapidly as neutron energy increases, but the distribution is appeared to be densed in the range $0.5 \sim 1.0$ at $E_0 = 36T$.

The calculations of the total scattering cross section of D_2O are carried out over the range of incident neutron energy E_0 from 0.01 eV up to 1.0 eV. Fig. 4 illustrates the numerical results compared with experimental values and the computed cross sections from heavy gas model[8].
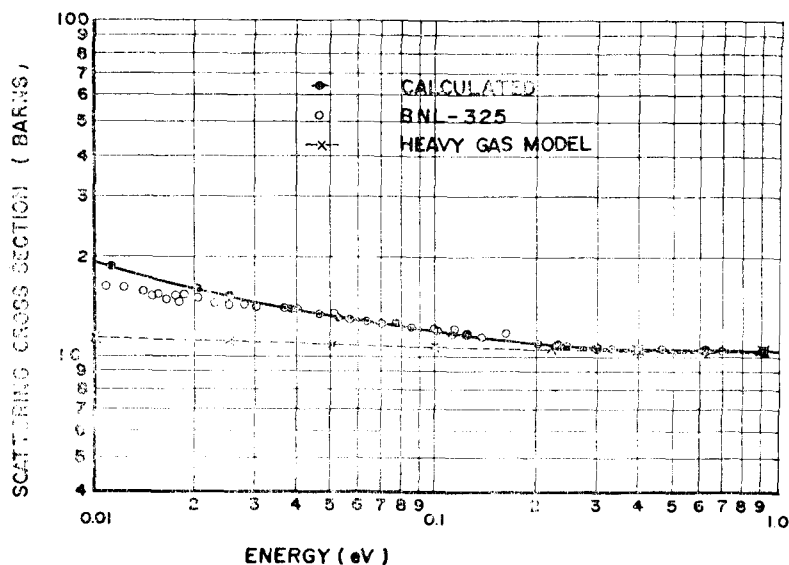
The experimental points are those of Rain-

Fig. 4. Scattering Cross Section of D₂O

water[9] et al, as read from the cross section compilation BNL-325[2]. It can be easily recognized from Fig. 4 that heavy gas model cannot be used to predict thermal neutron spectrum in D_2O moderator system. Scattering cross sections between Butler model and experiment in the energy range of interest is in good agreement as shown in Fig. 4. The discrepancies at the low neutron energy are considered to be due to the single molecular assumption of the model with hinderance of the molecular motion which might be important for values of energy transfer corresponding to small momentum transfer value.

Although the kernel evaluates the total scattering cross section of D_2O with some discrepancies in low neutron energy range 0.01eV~0.03 eV, it is believed from this study that the Butler model can be applied to predict thermal neutron behavior in a heavy water moderated system.

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