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MODEL FOR CALCULATING
THE SLOW NEUTRON SCATTERING BY LIQUID D₂O

H. L. McMurry

RELEASED FOR ANNOUNCEMENT
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MODEL FOR CALCULATING THE SLOW NEUTRON SCATTERING BY LIQUID D_2O

H. L. McMurtry

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H. L. McMurry

SUMMARY

The Nelkin (N) model for room temperature water considers each H₂O molecule to possess free translational motion and assumes the intermolecular interaction produces single torsional oscillation of 0.06 eV. The McMurry-Russell (MR) model is a modification which allows for aggregates of H₂O molecules which exhibit low frequency modes below 0.025 eV associated with group translation, together with intermediate modes with energies up to 0.125 eV associated with group rotations. Both the N and MR models assume that each H₂O molecule retains its three internal modes of 0.2, 0.46 and 0.46 eV.

These models have been scaled to suit D₂O by reducing the energies of the modes with characteristic energies in the range $0.025 < E \leq 0.125$ eV by $1/\sqrt{2}$, and by introducing the known internal D₂O frequencies. In addition, the scaled MR model (called MD in this work) has been altered by selecting the fraction of free D₂O molecules to give a least squared fit to the experimental data.

Seventeen percent of free D₂O molecules gives the scaled MR model which optimizes the fit to the 22°C scattering law data of Haywood. This agreement is quite good and is superior to that obtained with the scaled N model. Agreement with recent double differential scattering data obtained by Whittemore using 0.157, 0.206 and 0.300 eV neutron at 22°C is quite good for both the scaled MR and scaled N models. However, Whittemore's data show structure

which is not predicted by either model.

The scaled MR model with 53 percent free D₂O molecules optimizes agreement with Haywood's 150°C data. The agreement is fair at $\beta \leq 0.5$ and is superior to the scaled N model. For $\beta > 0.5$ both models give rather poor agreement.

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MODEL FOR CALCULATING THE SLOW NEUTRON SCATTERING BY LIQUID D₂O

H. L. McMurry

I. INTRODUCTION

This report describes a scaling of the parameters in the McMurry-Russell (MR) model¹ for water so that it applies to D₂O. The parameters are scaled according to reasonable theoretical criteria. Subsequent to this, a least squares fit to Haywood's room temperature data² for D₂O is obtained by adjusting the fraction of free D₂O molecules in the mixture. A similar adjustment of the free D₂O fraction is made to optimize agreement with the 150°C data.² It is found that 17 and 53 percent of free molecules are required at 22°C and 150°C, respectively. However, this results from an empirical process which omits consideration of many factors and there is little theoretical significance in these numbers.

The Nelkin model for water³ is also scaled to fit D₂O. Comparison of calculations using the two models are made with Haywood's data.

II. THE MR MODEL FOR WATER

The MR model¹ treats water as a mixture of 10 percent of free H₂O molecules, together with 45 percent each of two types of molecular aggregates in which the effective scattering masses for H atoms are 150 and 75 atomic units. The free H₂O molecules exhibit the two internal stretching vibrations near 0.46 eV, and the deformation mode of 0.2 eV energy.⁴ The aggregates are treated as large molecules exhibiting three types of normal modes. One type has characteristic vibrational energies E_{τ} in the range $0 < E_{\tau} \leq 0.025$ eV which are assumed to involve mainly translational motions of the H₂O molecules.

A second type with characteristic energies in the range $0.025 < E_T \leq 0.125$ eV are modes in which torsional oscillations of the H_2O molecules dominate. Finally, each H_2O molecule in an aggregate is assumed to exhibit the same internal modes as occur in the free molecules. Characteristic energies of both internal stretching modes were taken to be 0.46 eV which is near the experimental values, and the experimental value of 0.2 eV was used for the deformation mode. The vibrational amplitudes, which appear in the neutron scattering equations, were calculated from a normal coordinate analysis.

The characteristic energies and vibration amplitudes for the low energy modes ($E_T \leq 0.125$ eV) in the aggregates were selected to bring the calculated scattering into agreement with the data. However, the vibration amplitudes are subject to certain sum rules which will be discussed later since they are also used in determining the scaled parameters in D_2O .

Table I presents the parameters which characterize the MR model for H_2O . Separate entries are given for the free molecules and each aggregate. Molecule 1 is the free H_2O and the $f_1 = 0.1$ denotes the fraction of H_2O molecules in the system which are free. The M_{H1} and M_{O1} are the effective scattering masses for H and O atoms, respectively, in molecule 1. Aggregates 2 and 3 contain bonded H_2O molecules in which the effective scattering masses M_{H2} and M_{H3} for H atoms scattering are 150 and 75, respectively. The masses M_{O2} and M_{O3} are taken to be 150 and 75 also. The numbers $f_2 = f_3 = 0.45$ indicate that each aggregate contains 45 percent of the H_2O molecule in the system.

The rows headed by E_T lists the characteristic energies for the normal modes for that aggregate. Included in each list is a mode of zero frequency which corresponds to the lumped sum of the translational and rotational degrees of freedom of the entire aggregate.

TABLE I
MR MODEL PARAMETERS ^a

A. Molecule 1; $f_1 = 0.1$; $M_{H1} = 1.89$; $M_{O1} = 17.0$

$E_{\tau 1}(\text{eV})$	0.0	0.2	0.46
$\bar{\delta}_{H\tau 1}$	1.590	0.5	0.91
$\bar{\delta}_{O\tau 1}$	0.1760	0.0045	0.0070

B. Molecule 2; $f_2 = 0.45$; $M_{H2} = 150$; $M_{O2} = 150$

$E_{\tau 2}(\text{eV})$	0.0	0.005	0.01	0.015	0.02	0.025	0.05	0.075	0.1	0.125	0.2	0.46
$\bar{\delta}_{H\tau 2}$	0.02	0.013	0.018	0.020	0.018	0.047	0.35	0.45	0.3	0.354	0.5	0.91
$\bar{\delta}_{O\tau 2}$	0.02	0.0005	0.005	0.01	0.01	0.015	0.02	0.025	0.03	0.0405	0.0045	0.007

C. Molecule 3; $f_3 = 0.45$; $M_{H3} = 75$; $M_{O3} = 75$

$E_{\tau 3}(\text{eV})$	0.0	0.0075	0.0125	0.0175	0.0225	0.0375	0.0625	0.0875	0.1125	0.2	0.46
$\bar{\delta}_{H\tau 3}$	0.04	0.027	0.019	0.022	0.024	0.19	0.69	0.54	0.038	0.5	0.91
$\bar{\delta}_{O\tau 3}$	0.04	0.0005	0.005	0.01	0.015	0.02	0.025	0.030	0.0305	0.0045	0.007

^aAll aggregates contain a vibrational mode of zero energy. The $\bar{\delta}_{v0\ell}$ for this mode determines $M_{v\ell}$ through the relation $M_{v\ell} = 3/\bar{\delta}_{v0\ell}$.

The rows headed by $\bar{\delta}_{H\tau}$ and $\bar{\delta}_{O\tau}$ contain effective reciprocal masses for the atoms in each normal mode. Physically these quantities are related to the average squared vibration amplitudes $\langle \Delta r_{v\tau}^2 \rangle$ by the relation⁵

$$\langle \Delta r_{v\tau}^2 \rangle = \frac{\bar{\delta}_{v\tau} \hbar^2}{2E_\tau} = 0.208 \times 10^{-2} \frac{\bar{\delta}_{v\tau}}{E_\tau} (\text{\AA}^2) \quad (1)$$

In Eq. (1) subscript v refers to either the H or O atom and τ refers to the mode. For semi-rigid molecules with known force constants, the $\bar{\delta}_{v\tau}$ can be obtained from a normal coordinate analysis⁵. They must obey the sum rule⁶

$$\sum_\tau \bar{\delta}_{v\tau} = \frac{3}{m_v} \quad (2)$$

In Eq. (2) m_v is the mass of atom v in atomic units and the sum is over all modes, including the mode with zero frequency. In addition, the present application uses approximate sum rules for sums over the translational and rotational modes in the aggregates. These are discussed in the next section.

III. SCALING OF THE MR MODEL

1. Choice of Masses and Characteristic Energies.

The model for D_2O assumes the existence of free D_2O molecules and two aggregates corresponding to those with $M_H = 150$ and $M_H = 75$ in the MR model. In the free D_2O molecule the effective scattering masses $M_{D1} = 3.598$ and $M_{O1} = 18.012$ were obtained from the molecular masses and moments of inertia in the usual way.^{6,7} In the aggregates the masses were $M_{D2} = M_{O2} = 167$ and $M_{D3} = M_{O3} = 83$ which are just $\frac{20}{18}$ times 150 and 75, respectively, of the corresponding masses in

the MR model. This slight alteration in masses has little significance and is used only to conform to a consistent scheme for scaling parameters.

The frequencies in the two aggregates were obtained from those in the H_2O aggregates on the assumption that modes involving translational motions of the molecules are not significantly affected, while those involving rotational oscillations are reduced by about $1/\sqrt{2} = 0.707$. Thus, those modes in the range $0 < E_\tau \leq 0.025$ eV in the MR model for H_2O have been used unaltered in the D_2O model, while those MR modes with energies in the range $0.025 < E_\tau \leq 0.125$ have been multiplied by 0.707 to give values suited to the D_2O aggregates. The stretching and deformation modes internal to the D_2O molecules were given their experimental values of 0.146 eV (symmetrical deformation), 0.331 eV (symmetrical stretching) and 0.346 eV (unsymmetrical stretching).

2. Determination of $\bar{\delta}_{v\tau}$

The $\bar{\delta}_{v\tau}$ values ($v = D$ or O) for the free D_2O molecules were obtained from a D_2O normal coordinate analysis.

The $\bar{\delta}_{v\tau}$ for the mode of zero energy in the aggregates were determined from the requirement¹

$$\bar{\delta}_{v\tau, \ell} = \frac{3}{M_{v\ell}} \quad (3)$$

where τ now denotes the mode of zero frequency.

Here $M_{v\ell}$ is the effective scattering mass of atom v in the ℓ th aggregate ($\ell = 1, 2$ or 3). The masses are, of course, $M_{D1} = 3.598$, $M_{O1} = 18.012$, $M_{D2} = M_{O2} = 167$, $M_{D3} = M_{O3} = 83$.

For modes in the range $0 < E_\tau \leq 0.025$ eV in aggregates 2 and 3

the sum condition for pure translational modes is used.¹

$$\sum_t \bar{\delta}_{vt,\ell} = 3 \left[\frac{1}{18} - \frac{1}{M_{v\ell}} \right] \quad \text{for } H_2O \quad (4a)$$

$$\sum_t \bar{\delta}_{vt,\ell} = 3 \left[\frac{1}{20} - \frac{1}{M_{v\ell}} \right] \quad \text{for } D_2O \quad (4b)$$

The subscript t is used in place of τ to denote modes to emphasize that in Eqs. (4a, 4b) the sums are over modes involving group translations. Also, the sums exclude the mode of zero frequency.

The $\bar{\delta}_{v\tau,\ell}$ for the D_2O aggregates were obtained from the $\bar{\delta}_{v\tau,\ell}$ for the corresponding MR mode by multiplying the MR value by the ratio of the sums in 4b and 4a.

For modes in the range $0.025 < E \leq 0.125$ eV in H_2O , or $0.025 < E \leq 0.0883$ in D_2O , the sum rules corresponding to Eqs. (4a, 4b) are

$$\sum_r \bar{\delta}_{Hr,\ell} = \frac{3}{m_H} - \frac{3}{18} - 1.41 \quad \text{MR Model} \quad (5a)$$

$$\sum_r \bar{\delta}_{Or,\ell} = \frac{3}{m_O} - \frac{3}{18} - 0.0115$$

$$\sum_r \bar{\delta}_{Dr,\ell} = \frac{3}{m_D} - \frac{3}{20} - 0.6662 \quad \text{MD Model} \quad (5b)$$

$$\sum_r \bar{\delta}_{Or,\ell} = \frac{3}{M_O} - \frac{3}{20} - 0.02094$$

Again, the subscript r replaces τ to emphasize that the sum is over modes associated with rotational motions. The constants 1.41 and 0.6662 are the sums of $\sum_{\tau} \bar{\delta}_{H\tau}$ and $\sum_{\tau} \bar{\delta}_{D\tau}$ over the three internal modes in H_2O and D_2O , respectively. They come from the normal coordinate analyses. The constants 0.0115 and 0.02094 are the corresponding sums

for the O atom.

The $\bar{\delta}_{DT,\ell}$ were obtained from the corresponding $\bar{\delta}_{HT,\ell}$ values by multiplying the $\bar{\delta}_{HT,\ell}$ by the ratio of the sum in Eq. (5b) to that in (5a). Small adjustments were used to insure that Eq. (2), which is rigorous, holds.

The $\bar{\delta}_{OT,\ell}$ values were determined by taking the value given by Eq. (5b) and distributing it evenly among all the modes.

IV. DETERMINATION OF f_1 , f_2 AND f_3

It was discovered that to optimize the agreement between results calculated for the D₂O model and Haywood's data² the values of f_1 , f_2 , and f_3 had to be altered from those used in the MR model. It proved adequate to retain the assumption $f_2 = f_3$ and to alter f_1 subject to the requirements

$$f_1 + f_2 + f_3 = 1.0$$

The final f_1 was arrived at by making a least squares fit to Haywood's data. These data can be presented in terms of the scattering law⁸ $S(\kappa^2, \beta)$ defined from the equation

$$S(\kappa^2, \beta) = \sqrt{\frac{E_0}{E}} e^{\beta/2} \sigma(E_0, E, \mu) \quad (6)$$

In Eq. (6) E_0 and E denote the energies of the incident and scattered neutron, respectively, $\beta = (E - E_0)/kT$, κ^2 the square of the neutron momentum change divided by $\hbar^2$ and μ is the cosine of the scattering angle. The $\sigma(E_0, E, \mu)$ is the partial differential scattering cross section for a neutron of energy E_0 scattered to energy E through an angle whose cosine is μ . Units here are barns/eV x sr x D₂O.

The least square analysis was as follows. Let $S_H(i)$ denote Haywood's observed value for a particular κ^2 and β . Let $S_1(i)$, $S_2(i)$ and $S_3(i)$ denote $S(\kappa^2, \beta)$ values calculated for this κ^2 and β for the free molecules, and molecules in aggregates 2 and 3, respectively. Then the calculated $S(i)$ is given by

$$\begin{aligned} S(i) &= f_1 S_1(i) + \frac{1-f_1}{2} [S_2(i) + S_3(i)] \\ &= f_1 \left[S_1(i) - \frac{S_2(i) + S_3(i)}{2} \right] + \frac{S_2(i) + S_3(i)}{2} \end{aligned} \quad (7)$$

It is convenient to consider the ratio $[S(i) - S_H(i)] / (S_H(i))$ and to choose f_1 so the following sum is minimized

$$\begin{aligned} \sum_i \left[\frac{S(i) - S_H(i)}{S_H(i)} \right]^2 &= \sum_i \left[f_1 \phi_1(i) + \phi_2(i) \right]^2 \\ \phi_1(i) &= \frac{S_1(i) - [S_2(i) + S_3(i)]/2}{S_H(i)} \\ \phi_2(i) &= \frac{[S_2(i) + S_3(i)]/2 - S_H(i)}{S_H(i)} \end{aligned} \quad (8)$$

This leads to

$$f_1 = \frac{-\sum_i \phi_1(i) \phi_2(i)}{\sum_i \phi_1^2(i)} \quad (9)$$

In applying Eq. (9) values of $\kappa^2 = 2, 4, 6, 8, 32, 64 \text{ \AA}^{-2}$ were used for each β value for which Haywood supplied data. The computing program³ gave the $S_1(i)$, $S_2(i)$ and $S_3(i)$ so the $\phi_1(i)$ and $\phi_2(i)$ could be obtained. The converged value for f_1 was 0.17 so that $f_2 = f_3 = 0.415$.

TABLE II

PARAMETERS FOR MODEL MD FOR CALCULATING SCATTERING BY LIQUID D₂OA. Molecule 1; $f_1 = 0.17$; $M_{D1} = 3.598$; $M_{O1} = 18.012$

E_T	0	0.146	0.331	0.346
$\bar{S}_{DT,1}$	0.8338	0.2190	0.2310	0.2162
$\bar{S}_{OT,1}$	0.16656	0.00774	0.00476	0.00844

B. Molecule 2; $f_2 = 0.415$; $M_{D2} = 167$; $M_{O2} = 167$

E_T	0.0	0.005	0.01	0.015	0.02	0.025	0.0354	0.053	0.0707	0.0884	0.146	0.331	0.346
$\bar{S}_{DT,2}$	0.01792	0.01170	0.01957	0.02137	0.01957	0.04567	0.1680	0.2160	0.1440	0.1700	0.2190	0.2130	0.2162
$\bar{S}_{OT,2}$	0.01792	0.00045	0.0045	0.0090	0.0090	0.01350	0.0280	0.0280	0.0280	0.02818	0.00774	0.00476	0.00844

C. Molecule 3; $f_3 = 0.415$; $M_{D3} = 83$; $M_{O3} = 83$

E_T	0.0	0.0075	0.0125	0.0175	0.0225	0.0265	0.0442	0.0619	0.07950	0.1460	0.331	0.346
$\bar{S}_{DT,3}$	0.03614	0.02430	0.01710	0.01980	0.02160	0.0930	0.3380	0.2650	0.01886	0.2190	0.2310	0.2162
$\bar{S}_{OT,3}$	0.03614	0.00045	0.0045	0.00899	0.0135	0.0257	0.0257	0.0257	0.02588	0.00744	0.00476	0.00844

Table II summarizes all the parameters for the D_2O model (called the MD (for McMurry - D_2O) including the f values.

V. SCALED NELKIN MODEL (ND MODEL)

The Nelkin model for water^{1,3} uses $\bar{\delta}_{vT}$ values for the internal modes computed from a normal coordinate analysis. A single mode of 0.06 eV energy characterizes the interaction of the H_2O molecules and the $\bar{\delta}_{vT}$ is just the sum of the values which would be associated with the rotational degrees of freedom of the free H_2O molecules. The $\bar{\delta}_{vT}$ for the mode of zero frequency is just $3/18$ where 18 is the molecular mass.

This model is easily scaled to fit D_2O by using the observed frequencies¹⁰ for the internal modes of D_2O and calculated values for the $\bar{\delta}_{vT}$ for these modes. The mode which has the characteristic energy 0.06 eV is scaled by multiplying by 0.707. The $\bar{\delta}_{vT}$ for this mode and the mode of zero frequency can be computed in the same way as for H_2O . The resulting parameters are shown in Table III.

TABLE III

PARAMETERS FOR THE ND (NELKIN - D_2O) MODEL

$$M_D = 20.0, M_O = 20.0$$

$E_T(\text{eV})$	0.0	0.0424	0.146	0.331	0.346
$\bar{\delta}_{DT}$	0.15	0.6838	0.2190	0.2310	0.2162
$\bar{\delta}_{OT}$	0.15	0.01656	0.00774	0.00476	0.00844

VI. MODELS FOR D₂O AT ELEVATED TEMPERATURES

Haywood also has measured the scattering of D₂O at 150°C. The MD model was altered by varying f_1 to optimize agreement with those data using the technique already described for obtaining a least squares fit. The result was $f_1 = 0.53$, $f_2 = f_3 = 0.235$.

No adjustments were made in the ND model.

VII. RESULTS

1. Room Temperature Results

Fig. 1 compares the $S(\kappa^2, \beta)$ curves calculated on the MD and ND models with Haywood's data² for water at 22°C. The MD model gives better over-all agreement, especially at the lower β values. Since the calculated functions do not include coherence, they cannot reproduce some of the minima such as those which are quite apparent in the data for $\beta = 0.2 - 0.5$. Recent work on CD₄ illustrates how such minima arise from interference.¹¹

Figs. 2-4 compare with double differential cross section data obtained by W. L. Whittemore¹² using incident neutrons of energy 0.157, 0.206 and 0.304 eV. Only at the forward angles are there really significant differences between the models. The pronounced maxima in the ND curves arise because of the single low energy mode of 0.0424 eV with high $\bar{\delta}_{Dr}$. If these are disregarded, the general features of the ND and MD calculations are similar even at forward angles.

Dr. Whittemore's data indicates a number of maxima which are not reproduced by either model. The strong peak near 0.06 eV outgoing energy (0.146 eV energy transfer) in the data for 0.206 eV neutrons scattered at 60° is not predicted by either model. The data

indicate a similar situation for 0.157 eV neutrons scattered at 60° (peak at ~ 0.06 eV outgoing energy, or 0.097 eV energy transfer) and 0.304 eV neutrons scattered at 60° (possibly 0.1 eV outgoing energy, or 0.2 eV energy transfer). Some of these effects may arise from coherence (neglected in the calculations). If this is true, the intensities contributed by the coherent effects are quite high (especially for the peak in the scattering of 0.206 eV neutrons at 60°).

Table IV gives results for total cross sections in comparison with experimental data and Table V compares average scattering cosines. Table VI gives the ratio of the average energy after collision to E_0 .

2. Comparison With High Temperature Data

Fig. 5 compares the calculated results with Haywood's data at 150°C . The agreement is not nearly as good as for the low temperature data. The MD model is in somewhat better agreement at low β values, but there is not much to choose between models for the larger β values.

VIII. DISCUSSION

The MD models for water at 22°C and 150°C must be looked on as empirical. The fact that f_1 for the 22° case is 0.17 for D_2O as compared to 0.1 for H_2O is not readily understood on physical grounds. Indeed, the fraction of free molecules might be expected to be less in D_2O .¹³

It is reasonable to expect f_1 to increase as the water temperature rises, but the increase is much larger for D_2O than for H_2O .¹⁴

No attempt has been made to take account of the looser binding

which may prevail at higher temperatures, and diffusive effects are neglected entirely.

Finally, some recent calculations of multiple scattering in H_2O suggest that the data in D_2O may contain significant contribution from multiple scattering.¹⁵

TABLE IV

COMPARISON OF OBSERVED AND CALCULATED TOTAL CROSS SECTIONS
FOR D_2O AT 295°K

E_o (eV)	$\sigma(E_o)$ (barns)		
	Obs'd	MD	ND
0.005	20.5	20.5	20.7
0.05	13.3	13.1	13.2
0.15	11.6	11.3	11.7

TABLE V

COMPARISON OF CALCULATED AVERAGE SCATTERING COSINES
FOR D_2O AT 295°K

E_o	$\bar{\mu} = \sigma_i(E_o)/\sigma_o(E_o)$	
	MD	ND
0.005	0.028	0.041
0.05	0.14	0.14
0.15	0.20	0.19

TABLE VI

CALCULATED RATIO OF AVERAGE ENERGY AFTER SCATTERING
TO INITIAL ENERGY FOR D₂O AT 295°K

E_o (eV)	$\bar{E}/E_o$	
	MD	ND
0.005	2.44	2.65
0.05	1.01	1.04
0.15	0.04	0.83

Fig. 1 Comparison of calculated and experimental scattering laws for D₂O at 295°K. Data from B. C. Haywood, Ref. 2

Fig. 2 Partial differential cross sections of D_2O for 0.157 eV neutrons at 295°K

Fig. 3 Partial differential cross sections of D_2O
for 0.206 eV neutrons at 295°C K

Fig. 4 Partial differential cross sections of D_2O for 0.304 eV neutrons at 295°K

Fig. 5 Comparison of calculated and experimental scattering laws for D₂O at 423°K. Data are from B. C. Haywood, Ref. 2

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