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Absolute vibrationally elastic cross sections for electrons scattered from water molecules between 6 eV and 50 eV

W M Johnstone and W R Newell

Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

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Abstract. In this work we report absolute vibrationally elastic differential cross sections for electrons scattered from water in the energy range 6–50 eV. From an extrapolation of the differential cross section data, absolute integral and momentum transfer cross sections have also been obtained. For incident electron energies above 20 eV and below 100 eV there exists only one previous report of differential cross sections (Danjo and Nishimura). Significant discrepancies have been found in this region between the cross sections reported by Danjo and Nishimura and those obtained in the present work.

1. Introduction

The interaction of electrons with water molecules has been found to be significant in a wide variety of naturally occurring phenomena and in many practical applications. Two areas of particular relevance are the study of the effects of ionizing radiation on the human body, whose water content is approximately 60% (Turner *et al* 1982), and the modelling of plasmas produced in magnetohydrodynamic power generators (Norcross and Collins 1982).

Several absolute measurements of electron–water cross sections have been reported. Absolute total cross sections were first obtained by Brüche (1929) and more recently by Sueoka *et al* (1986), Szmytkowski (1987) and Nishimura and Yano (1988). The first comprehensive set of absolute elastic differential cross sections (DCS) was reported by Danjo and Nishimura (1985) at incident electron energies between 4 eV and 200 eV and for scatterng angles from 15° to 120°. Measurements have also been reported by Katase *et al* (1986) at incident energies between 100 eV and 2100 eV for scattering angles between 5° and 130° and by Shyn and Cho (1987) at energies between 2.2 eV and 20 eV and scattering angles between 15° and 150°. Jung *et al* (1982) also published absolute elastic differential cross sections as part of their observations on the rotational excitations of water at incident energies of 2.2 eV and 6 eV.

A large number of theoretical studies have also been reported for electron scattering from H_2O. Several Born or Born-like calculations have been performed (e.g. Itikawa 1972, Fujita *et al* 1983), but have only had limited success at high energies (>100 eV) and low scattering angles ($\theta < 30°$). Gianturco and Thompson (1980) were the first to attempt an *ab initio* calculation of the cross sections for electron scattering from water using the single centre expansion method and model potentials to describe exchange

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and polarization effects. Recently, Gianturco and Scialla (1987) have reported results for electron-water collisions using similar single centre expansion techniques but employing improved exchange and polarization potentials. Sato *et al* (1988) have also published electron scattering cross sections using a 'modified' version of the continuum multiple scattering method. Finally, Brescansin *et al* (1986) have performed calculations based on the Schwinger variational principle where an exact static-exchange potential was employed to account for short-range aspects of the collision governed by the low angular momentum partial waves.

In the present work, absolute vibrationally elastic differential cross sections for electron scattering from water for incident energies between 6 eV and 50 eV have been measured. These results are presented here in addition to integral and momentum transfer cross sections derived from an extrapolation of the DCS data.

2. Experimental details

The apparatus used in this work has already been described in detail (Johnstone and Newell 1991) and so only a brief description will be given here. The apparatus used consisted of a conventional electron spectrometer which can be considered in three parts. In the first part, the monochromator, electrons emitted from a thoriated tungsten filament were focused into a 180° hemispherical selector. The resultant electron beam was transported to an interaction region where it was crossed with a gas beam whose central axis was perpendicular to that of the incident electron beam. The full width at half maximum (FWHM) of the electron beam was typically 40 meV for a current of $\simeq$15 nA. Electrons scattered from the gas beam were detected by the second part of the spectrometer, the analyser. In the analyser, the energy distribution of the scattered electrons was determined using a second smaller hemispherical selector and the resolution of the analyser was typically 45 meV. The resolution of the spectrometer was determined from a measurement of the FWHM of the energy distribution of the electrons elastically scattered from helium and found to be $\simeq$60 meV. The angular range of the analyser was $-10°$ to 120° and the angular resolution of the spectrometer was 2°. The third part of the spectrometer, the reference detector, was used to measure the scattered electron flux at a fixed scattering angle of $-90°$. It was constructed from a three-element electrostatic lens followed by a channeltron electron multiplier (CEM). By applying a sufficiently negative voltage to the centre element of the lens it was possible to allow only those electrons which had been elastically scattered from the gas beam to reach the CEM. In comparison to the analyser, the reference detector had a much lower resolution, $\simeq$100 meV.

To determine the contact potential, the position of the 19.37 eV He resonance was measured and compared to the value given by Brunt *et al* (1977). The gas beams were produced by flowing gas through a capillary tube 25 mm long with an internal diameter of 0.6 mm. The helium gas used was commercially available and of research grade purity while the water vapour was obtained from a cell containing de-ionized water. To purge the air from the cell and to remove any gases dissolved in the water, the following procedure was employed. The cell was first evacuated to 'boil' off any gases dissolved in the water, e.g. carbon dioxide, and then, while continuing to evacuate the cell, the water was frozen using liquid nitrogen. The cell was then sealed and the ice

allowed to melt. This procedure was repeated two or three times to reduce the amount of dissolved gases in the water to a minimum.

3. Experimental procedure

The absolute differential cross sections presented in this work were obtained using a two-step process. First, the relative differential cross section, normalized at $\theta = 90°$, was found. The absolute value of the cross section was then measured at $\theta = 90°$ and this value was then used to normalize the corresponding relative differential cross section. This procedure has been described in detail by Johnstone and Newell (1991) and so only a brief outline of the important points will be given.

3.1. Measurement of the relative differential cross sections

Two specific problems are often encountered when making differential cross section measurements. First, it is important to ensure that the intensities of the gas and electron beams remain stable during the course of the angular measurements or that any fluctuations are accounted for, otherwise a false angular dependence will be introduced into the values of the differential cross sections. The second problem concerns the overlap volume of the two beams and the proportion of this overlap 'seen' by the analyser as a function of angle. If the view cone of the analyser does not encompass the entire overlap volume then as the analyser is rotated, the fraction of the total overlap volume seen will vary and this too will introduce a false angular dependence into the differential cross section measurements.

In this work, any variations in beam stability were removed by determining the ratio of the scattered electron count rate measured by the analyser at an angle θ, to the scattered count rate measured by the reference detector. Thus if $D(\theta)$ is the count rate recorded by the analyser and $R(\theta)$ is the count rate simultaneously recorded from the reference detector, the relative differential cross section (normalized to 90°) is given by

$$\sigma(\theta)/\sigma(90°) = \frac{D(\theta)R(90°)V_{\text{eff}}(90°)}{D(90°)R(\theta)V_{\text{eff}}(\theta)} \tag{1}$$

where $V_{\text{eff}}(90°)/V_{\text{eff}}(\theta)$ is the ratio of the overlap volumes seen by the analyser at 90° and θ. Any change in $D(\theta)/D(90°)$ due to variation in the electron beam current, for example, was corrected for in the ratio $R(90°)/R(\theta)$. In addition, by taking two sets of measurements, the first with the gas beam present and the second without the gas beam but the same background pressure, it was also possible to eliminate the contribution due to electrons scattered from sources other than the gas beam by use of a subtraction method (Johnstone and Newell 1991).

The ratio $V_{\text{eff}}(90°)/V_{\text{eff}}(\theta)$, which can also be written as $F_{\text{eff}}(\theta)$, was determined by measuring the relative differential cross section for electron scattering from helium. $F_{\text{eff}}(\theta)$ was then found by comparing the measured differential cross sections with the true value. By suitable adjustment of the helium flow rate through the capillary tube it was possible to arrange for the electron–gas beam overlap in the helium measurements to be the same as that in the water measurements. The helium cross sections of Nesbet (1979) were used for incident energies below 20 eV. For energies of 20 eV and above the cross sections measured by Register *et al* (1980) were used. At the energies where these two sets of data overlap they are seen to agree to within 5%.

3.2. Absolute measurements of the differential cross section at 90°

To normalize the relative differential cross section to an absolute scale the absolute value of the elastic DCS at 90° was measured using the relative flow technique. This method, first described by Srivastava *et al* (1975), has recently been reviewed by Nickel *et al* (1989). In brief, the procedure employed is as follows. The gas for which the DCS is to be determined is admitted to the vacuum system via the capillary tube. The intensity of the incident electron beam I_1 is recorded and the count rate of electrons scattered through an angle θ by the gas beam $I_1^s(E, \theta)$ is measured. A second gas is then admitted to the system and its flow rate through the capillary tube adjusted such that the angular distribution of the second beam is the same as the first. The electron beam current I_2 and the scattered electron count rate $I_2^s(E, \theta)$ are again measured. The relationship between the two measured count rates and the cross sections is then given by

$$\frac{I_1^s(E, \theta)}{I_2^s(E, \theta)} = \frac{\sigma_1(E, \theta) N_1 I_1 m_1^{1/2}}{\sigma_2(E, \theta) N_2 I_2 m_2^{1/2}} \tag{2}$$

where $\sigma_1(E, \theta)$ and $\sigma_2(E, \theta)$ are the DCS for electrons scattered from the two gases, N_1 and N_2 are the flow rates of the two gases through the capillary tube and m_1 and m_2 are the molecular/atomic masses of the two gases. If the DCS for the second gas is known, it is then possible to determine $\sigma_1(E, \theta)$.

In the present work, the DCS at 90° was determined using the procedure described above and the reference detector was used to measure the scattered electron count rate. The flow of gas through the capillary tube was determined using a method described by Khakoo and Trajmar (1986). Helium was used as the second gas and as in the case of the volume correction measurements, the helium cross sections of Nesbet (1979) and Register *et al* (1980) were used as a reference standard.

3.3. Errors

The final error in the values of the DCS quoted in this work arose from two sources. These were the errors associated with the measurement of the relative differential cross sections and those associated with the determination of the absolute cross section at 90°. The errors associated with the measurement of the relative DCS had two contributions, the measurement of the DCS (4.5%) and the measurement of the volume correction (7%). For the absolute cross section measurements made at 90°, the error was calculated to be between 8% and 9.5%. These values included not only those associated with the electron beam current, flow rates and counting errors but also those from the absolute helium cross sections used as a reference standard. In addition, as the resolution of the reference detector is low, an error contribution has been included due to possible detection of inelastically scattered electrons detected by the reference detector. Thus the final error in the differential cross sections quoted in this work was $\simeq 13\%$.

4. Results and discussion

4.1. Differential cross sections

Results obtained in the present work for absolute elastic differential cross sections for electron scattering from water at incident energies between 6 eV and 50 eV are shown

in figures 1, 2 and 3 and these have also been tabulated in table 1. The experimental data of Jung *et al* (1982), Danjo and Nishimura (1985) and Shyn and Cho (1987) and the theoretical data of Brescansin *et al* (1986), Gianturco and Scialla (1987) and Sato *et al* (1988) are shown for comparison. The experimental results of Shyn and Cho (1987) were obtained using a similar two step procedure to that used in the present work. Shyn and Cho (1987) first obtained the relative differential cross sections before normalization to the absolute value at 90° found in a second experiment. In this case the value of the absolute cross section at 90° was determined from the relative intensities of the scattered signal from water and He measured in a gas cell experiment using the absolute helium cross sections of Shyn (1980) as the normalization standard. The measurements of Danjo and Nishimura (1985) were made using the relative flow technique with the helium elastic scattering data of Register *et al* (1980) used as the normalization standard. The helium data of Register *et al* (1980) have also been used as a normalization standard in the present work, therefore the data of Shyn and Cho (1987) have been renormalized to those of Register *et al* (1980) to eliminate any discrepancies arising from the use of different normalization standards. This has resulted in an increase of appoximately 10% in the published values of Shyn and Cho (1987).

Examination of the experimental data presented in figures 1, 2 and 3 reveals two features. At an incident energy of 6 eV, a minimum is observed at 120° which can be seen to migrate to an angle of 95° as the incident energy is increased to 50 eV. Secondly, an inflection is observed between 50° and 80° at 6 eV and, less distinctly, at 10 eV between 40° and 70°. In general, agreement between the present work and that of Shyn and Cho (1987) is good, except at the smaller scattering angles ($\theta < 40°$) and scattering

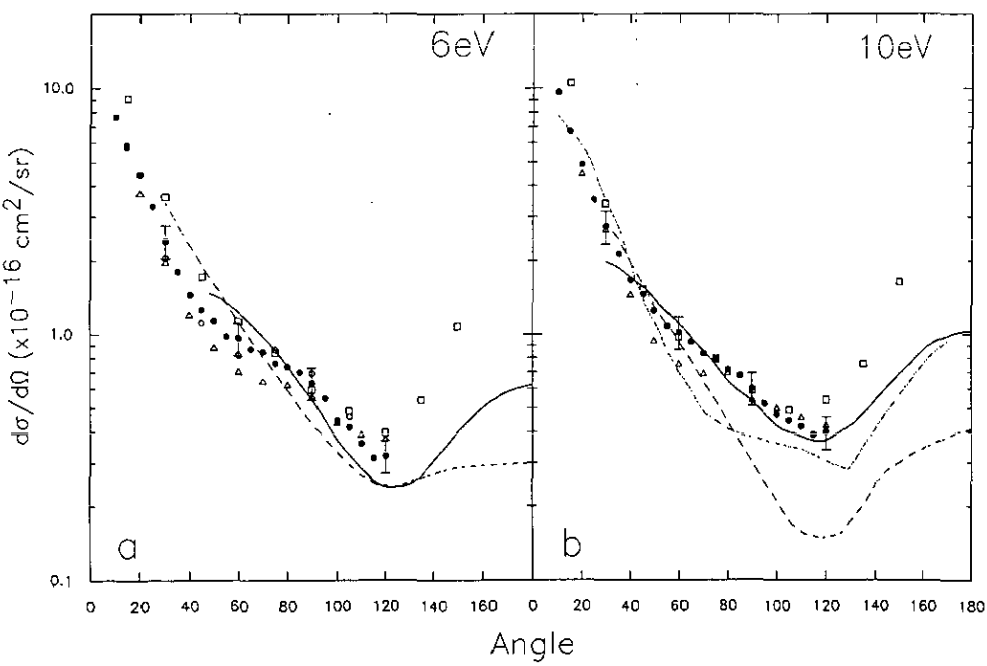

Figure 1. Absolute elastic differential cross section for electron scattering from H_2O for incident energies of: (a) 6 eV, (b) 10 eV. Experimental data: ●, present work; △, Danjo and Nishimura (1985); □, Shyn and Cho (1987); ○, Jung *et al* (1982). Theory: ——, Brescansin *et al* (1986); — — —, Gianturco and Scialla (1987); — · · —, Sato *et al* (1988).

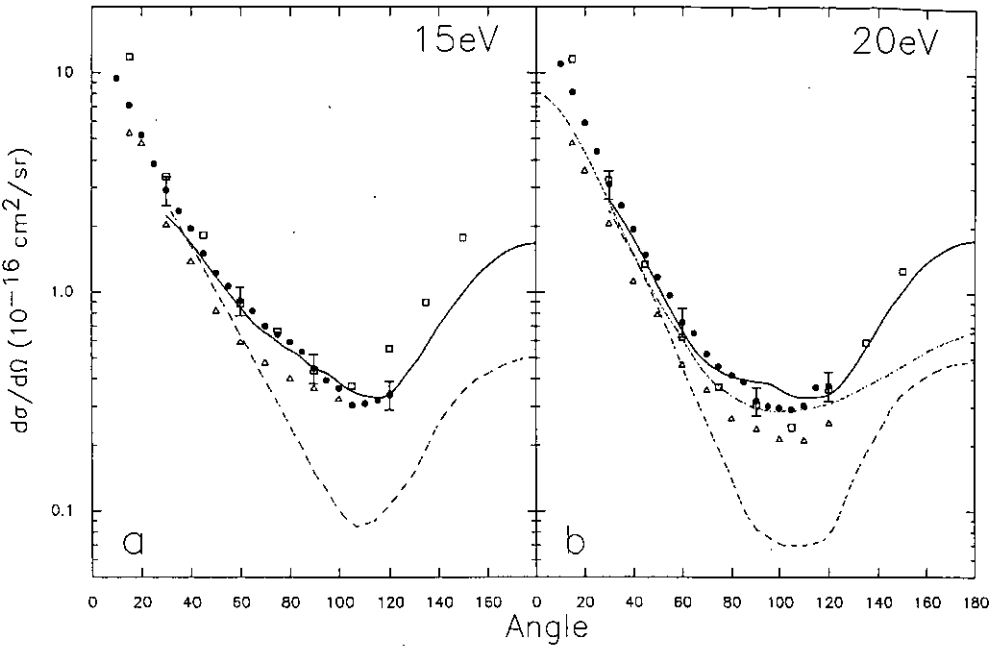

Figure 2. Absolute elastic differential cross section for electron scattering from H_2O for incident energies of: (a) 15 eV, (b) 20 eV. Experimental data: ●, present work; △, Danjo and Nishimura (1985); □, Shyn and Cho (1987). Theory: ——, Brescansin *et al* (1986); — — —, Gianturco and Scialla (1987); — · · —, Sato *et al* (1988).

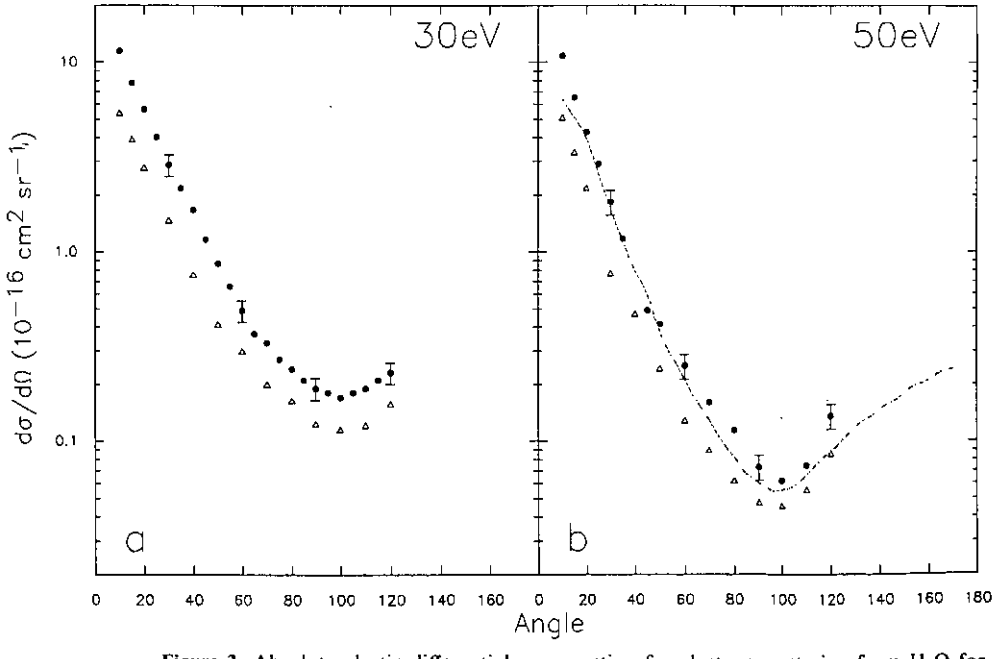

Figure 3. Absolute elastic differential cross section for electron scattering from H_2O for incident energies of: (a) 30 eV, (b) 50 eV. Experimental data; ●, present work; △, Danjo and Nishimura (1985). Theory: — · · —, Sato *et al* (1988).

Table 1. Absolute elastic differential cross sections for electron scattering from H_2O ($\times 10^{-16}$ cm^2 sr^{-1}).

	Incident energy (eV)					
θ (deg)	6	10	15	20	30	50
0						
10	7.63	9.62	9.45	11.02	11.44	10.76
15	5.86	6.69	7.10	8.15	7.80	6.55
20	4.46	4.89	5.17	5.91	5.66	4.30
25	3.31	3.52	3.85	4.38	4.04	2.92
30	2.40	2.75	2.91	3.12	2.88	1.84
35	1.80	2.13	2.34	2.48	2.17	1.17
40	1.45	1.67	1.95	1.94	1.67	
45	1.25	1.46	1.50	1.48	1.17	0.49
50	1.14	1.25	1.23	1.17	0.87	0.41
55	0.98	1.08	1.07	0.97	0.66	
60	0.96	1.02	0.91	0.73	0.49	0.25
65	0.87	0.93	0.82	0.65	0.37	
70	0.85	0.83	0.70	0.52	0.33	0.16
75	0.76	0.79	0.64	0.46	0.27	
80	0.74	0.71	0.59	0.42	0.24	0.11
85	0.70	0.68	0.53	0.39	0.21	
90	0.63	0.60	0.45	0.32	0.19	0.073
95	0.55	0.52	0.40	0.30	0.18	
100	0.45	0.47	0.36	0.30	0.17	0.062
105	0.42	0.44	0.30	0.29	0.18	
110	0.36	0.42	0.31	0.30	0.19	0.074
125	0.31	0.39	0.32	0.37	0.21	
120	0.32	0.40	0.34	0.37	0.23	0.14
error	13.0%	13.0%	13.0%	13.0%	13.0%	13.0%

angles larger than 110°, where their data are consistently higher. There is also reasonable agreement between the present work and that of Danjo and Nishimura (1985), but as the incident energy is increased, their data are seen to be consistently lower than the present work and those of Shyn and Cho (1987). This is particularly noticeable for incident energies of 20 eV and above where the data of Danjo and Nishimura (1985) are lower at all scattering angles. A comparison between the measurements of Katase *et al* (1986) and Danjo and Nishimura (1985) for an incident energy of 100 eV also reveals a similar behaviour. In figure 1(a) the data of Jung *et al* (1982) is shown and this is the sum of the pure elastic and rotational excitation cross sections. The excellent agreement between these results and the present work indicates that the cross sections measured in this work (and also by Danjo and Nishimura (1985) and Shyn and Cho (1987)) contain a significant contribution from rotational excitation and de-excitation cross sections for the lower incident electron energies. Unfortunately, the present electron spectrometer had a lower resolution than that of Jung *et al* (1982), therefore it was not possible to measure the rotational cross sections in this work. Finally, it should be noted that the differential cross sections of Shyn and Cho (1987) show a sharp rise for scattering angles greater than 120°. As these points lie outside the angular range of both the present work and that of Danjo and Nishimura (1985), it is not possible to say whether this is a true feature of the cross section.

A large number of theoretical studies have been reported for electron scattering from H_2O. Several Born or Born-like calculations have been performed (e.g. Itikawa 1972, Fujita *et al* 1983), but have only had limited success at high energies (>100 eV) and low scattering angles ($\theta < 30°$) and so will not be discussed here.

The theoretical calculations are only partly successful in reproducing the shape and magnitude of the experimental differential cross sections. For incident electron energies between 6 eV and 20 eV the results of Brescansin *et al* (1986) are found to be in good agreement with the experimental results. Their differential cross sections show the minimum around 120° and also a rising cross section for angles greater than 120°, however, it is only at 20 eV that their results approach the magnitude of the large angle cross sections of Shyn and Cho (1987). At all other energies the values of Brescansin *et al* (1986) are lower for scattering angles greater than 120°. The results of Sato *et al* (1988) are also found to be in good agreement with the experimental data, particularly at 20 eV and 50 eV. At the large scattering angles ($\theta > 120°$), the work of Sato *et al* (1988) also shows a rising cross section but not to the same degree as Brescansin *et al* (1986) or Shyn and Cho (1987). Finally, the work of Gianturco and Scialla (1987) compares poorly with the two other theoretical studies. Although, their work agrees well with the experimental results at the smaller scattering angles, the cross sections for angles greater than 70° are significantly lower. Finally, it should be noted that log scales are used on the y axes in figures 1 to 3 to accentuate any differences between the different data sets shown.

Due to the considerable differences in the computational methods used in the theoretical teatments discussed above, it is difficult to evaluate their relative merits except for their ability to match the experimental data. The results of Brescansin *et al* (1986) and Sato *et al* (1988) are seen to be in reasonable agreement with the experiments, but the results of Gianturco and Scialla (1987) are significantly lower for scattering angles in excess of 70°. Sato *et al* (1988) report that the shape and magnitude of their differential cross sections in this angular region were sensitive to the strength of the exchange interaction in their model. Therefore, the discrepancy in the data of Gianturco and Scialla (1987) may be a result of the exchange potential they employed. In addition, no polarization potential was included in their calculation of the differential cross sections and this may also explain the poor quality of their results.

4.2. Integral and momentum transfer cross sections

Integral (σ_t) and momentum transfer (σ_m) cross sections have been derived in the present work by an extrapolation of the measured differential cross section data to the scattering angles 0° and 180°. The errors associated with the values of σ_t and σ_m are larger than those of the differential measurements due to the angular range over which the differential cross section has to be extrapolated. The derived values of σ_t and σ_m are given in table 2 where the percentages represent the contribution to the final values from extrapolated regions of the differential cross sections. The errors in σ_t and σ_m were 20% and 25% respectively.

The total cross sections (σ_T) in figure 4 have been computed by the addition of the total ionization cross sections of Orient and Srivastava (1987) and the integral vibrational excitation cross sections of Shyn *et al* (1988) to the integral cross sections obtained in the present work. No electronic excitation cross sections are available but their contribution is likely to less than 5%. Similarly, the data of Orient and Srivastava (1987) and Shyn *et al* (1988) have also been added to the integral cross sections of

Table 2. Absolute integral and momentum transfer cross sections for electron scattering from H_2O ($\times 10^{-16}$ cm^2). The numbers given as percentages are the contributions from the extrapolated differential cross sections values.

	Incident energy (eV)					
	6	10	15	20	30	50
σ_i	13.76	13.89	13.31	13.66	11.29	7.76
	22%	16%	16%	20%	20%	20%
σ_m	10.63	9.36	8.44	8.90	6.83	5.09
	52%	43%	45%	55%	60%	73%

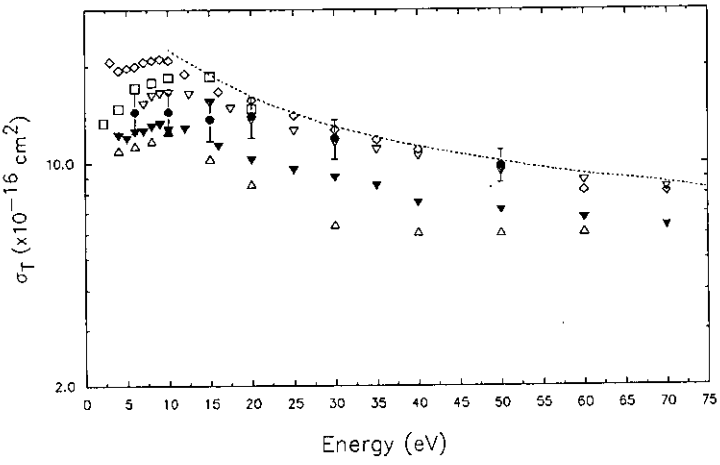

Figure 4. Absolute total cross sections for electron scattering from H_2O ($\times 10^{-16}$ cm^2). Experiment: ●, present work; △, Danjo and Nishimura (1985); □, Shyn and Cho (1988); ▼, Sueoka *et al* (1986); ◇, Szmytkowski *et al* (1987); ▽, Nishimura and Yano (1988). Theory: - - - -, Jain (1988).

Danjo and Nishimura (1985) and Shyn and Cho (1987). The results are shown in figure 4. The data of Danjo and Nishimura (1985) are lower at all energies and this is particularly noticeable above 15 eV. In their differential cross section measurements, it is observed that for incident energies of 15 eV and above, their results are consistently lower than the present work and this explains their smaller values of σ_T. A direct measurement of the total cross section has been reported by Sueoka *et al* (1986), Szmytkowski (1987) and Nishimura and Yano (1988) and their results are also shown in figure 4. In addition to these measurements, Jain (1988) has calculated the total cross section from 10 eV to 1000 eV using a semiclassical complex optical potential and this work follows the shape of the experimental data. There is general qualitative agreement between the experimental measurements, which all indicate a maximum around 10 eV. However, the magnitude of the cross section remains uncertain, particularly below 20 eV.

The momentum transfer cross sections calculated from the present data are shown in figure 5 together with the experimental results of Danjo and Nishimura (1985) and

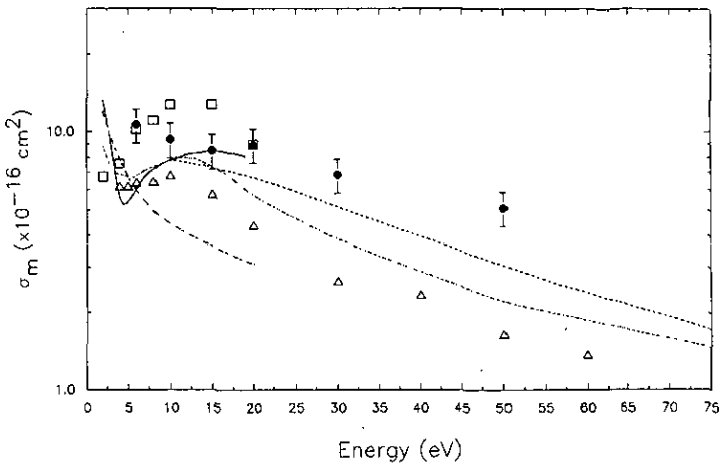

Figure 5. Absolute momentum transfer cross sections for electron scattering from H_2O ($\times 10^{-16}$ cm^2). Experiment: ●, present work; △, Danjo and Nishimura (1985); □, Shyn and Cho (1988). Theory: ——, Brescansin *et al* (1985); — — —, Gianturco and Scialla (1987); — · · —, Sato *et al* (1988); - - - -, Jain (1988).

Shyn and Cho (1987) and the theoretical results of Brescansin *et al* (1986), Gianturco and Scialla (1987), Sato *et al* (1988) and Jain (1988). The data of Shyn and Cho (1987) are generally larger than the other works and this is a result of the increased size of the differential cross sections at large scattering angles ($\theta \geq 120°$). Conversely, the data of Danjo and Nishimura (1985) are notably lower and this is a consequence of their generally lower differential cross section values. At low energies ($E < 20$ eV), the theoretical results of Brescansin *et al* (1986) and Sato *et al* (1988) are in general agreement, showing a minimum at 4 eV and a broad maximum around 15 eV. At the higher energies, the results of Sato *et al* (1988) and Jain (1988) show the same trend, lying between the data from the present work and those of Danjo and Nishimura (1985).

5. Conclusion

In this work absolute vibrationally elastic differential, integral and momentum transfer cross sections have been presented. Using the total ionization cross sections of Orient and Srivastava (1987) total cross sections were also found. For energies between 6 eV and 20 eV good agreement was found between the present work and that of Shyn and Cho (1987). The data of Danjo and Nishimura (1985) are in good agreement with the present work and those of Shyn and Cho (1987) at 6 eV and 10 eV but at higher energies the data of Danjo and Nishimura (1985) can be seen to be significantly lower. This is particularly noticeable at 30 eV and 50 eV where their differential cross sections are up to 50% lower than those of the present work or Sato *et al* (1988). A similar disagreement was reported at higher incident energies (100 eV and above) by Katase *et al* (1986). This would therefore suggest the possibility that the DCS data of Danjo and Nishimura (1985) between 15 eV and 100 eV may be incorrect and that the integral and momentum transfer cross sections at these energies are also in error.

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