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Total Electron Scattering Cross Sections for Ar, N_2, H_2O and D_2O

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Total electron scattering cross sections (σ_t) for Ar, N_2, H_2O and D_2O are determined in the energy range between 7 and 500 eV using a linear electron transmission device. The thermal transpiration effect is considered. Results for Ar and N_2 are in good agreement with those of previous experiments. The present results for H_2O agree well with those of Brüche, Szmytkowski, Szmytkowski *et al.* and Zecca *et al.* However, some difference still remains among the recent results in the energy region below 15 eV. The σ_t for D_2O is much the same as for H_2O within the experimental uncertainty.

§1. Introduction

The behaviour of slow electrons in water vapour is very important for understanding the earth's upper atmosphere, the comet's atmosphere and the gaseous discharge plasma.[1] Reliable experimental results are also needed in the field of the radiation chemistry. However, experimental results have been reported for e–H_2O fragmentarily.[2] The measurement on the σ_t for H_2O was carried out first by Brüche (Ramsauer type, 4.8–46 eV, 1929).[3] Very recently, several authors reported the σ_t for this process. Sokolov and Sokolova (electron-cyclotron resonance, 0.25 –7 eV, 1981),[4] Sueoka *et al.* (time of flight, 1–400 eV, 1986, 1987),[5,6] Zecca *et al.* (Ramsauer type, 81–3000 eV, 1987),[7] Szmytkowski (Transmission type, 0.5–80 eV, 1987)[8] and Szmytkowski *et al.* (Ramsauer type, 60–3000 eV, 1987).[9] In spite of the efforts by these authors, the experimental results still show considerable discrepancy.

Previously, we reported the differential elastic scattering cross sections of electrons for H_2O.[10] As a part of the systematic study for H_2O, the σ_t (7–500 eV) was measured using a simple linear electron transmission apparatus. Measurements were also carried out for its isotopic material D_2O in the same energy range.

For the normalization of the electron energy and the test of this experimental system, the σ_t for Ar and N_2 was measured over the energy range of interest.

Total scattering cross sections have served as a standard value for the normalization of the scattering cross sections for the individual scattering process because they give maximum scattering cross section values and can be determined without any procedures for normalization.

§2. Experimental

2.1 *Apparatus*

A schematic diagram of the apparatus used in this study is shown in Fig. 1. The apparatus consists of an electron source, a scattering cell and a Faraday cup. The electron source is composed of a pierce-type electron gun with hair pin-type tungsten filament and a grounded cylindrical electron drift space with four apertures (A_1–A_4). The apertures A_1, A_2 and A_3 are 1 mm and A_4 is 0.5 mm in diameter, respectively. The scattering cell is terminated by an entrance aperture A_5 (1.4 mm in diameter) and an exit aperture A_6 (2 mm in diameter). Target gas from a needle valve is introduced to the scattering cell through a metal tube. The cell and the sensor head of the manometer are connected with an extra metal tube for monitoring the target gas pressure in the cell. The Fara-

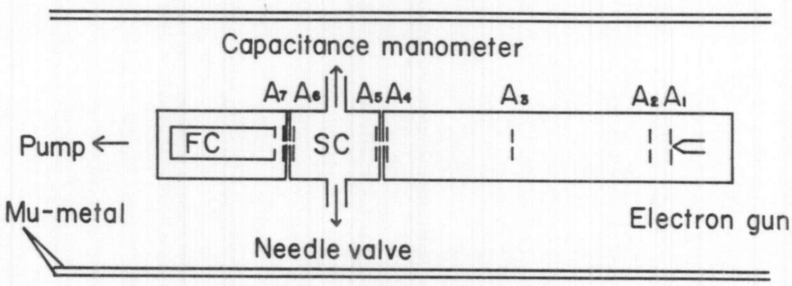

Fig. 1. Schematic diagram of the experimental apparatus.

day cup is enclosed by a cylinder with entrance aperture A_7 (2 mm in diameter). A gap between A_4 and A_5, A_6 and A_7 and the Faraday cup are 2 mm, 1 mm and 4 mm, respectively. The gap between A_4 and A_5 is opened to the vacuum space to minimize gas flow from the scattering cell to the electron drift space. All elements between the filament holder and the Faraday cup are made of nonmagnetic stainless steel (type 310). All apertures are made of molybdenum. The electron source, the electron drift space, the scattering cell and the Faraday cup are arranged and fixed on the axis of a vacuum chamber (150 mm in diameter, 508 mm in length) which is also constructed from nonmagnetic stainless steel (type 304L). The inner wall of the vacuum chamber is covered with a double layered mumetal shielding to eliminate the effect of the earth's magnetic field. The residual magnetic field around the axis of the vacuum chamber was 10 mG or less. The outside of the chamber is enclosed with a nonmagnetic heating coil and the chamber could be warmed up to about 80°C with the heating coil. The cell also could be warmed up to any temperature with an extra Ta wire fixed on the cell. The temperature of the cell is monitored with a calibrated thermistor also fixed on the cell. The vacuum chamber is evacuated to 1×10^{-7} Torr by a four-inch oil diffusion pump with liquid nitrogen trap. The background pressure of the vacuum chamber was kept below 2×10^{-5} Torr during the measurement.

2.2 *Current measurement*

The current intensities to the scattering cell (I_c), the Faraday cup (I_f) and the enclosure of the Faraday cup (I_e) are measured separately. Through the measurements, I_e was negligible compared with I_c and I_f. We assumed

$$I_0=I_{f0}=I_c+I_e+I_f, \tag{1}$$

where I_0 is the incident current and the suffix 0 indicates the current to each section when the scattering cell is evacuated. The relationship among the current intensities I_0, I_f, target gas density, n and the cross section, σ_t is given by

$$I_f=I_0\exp(-\sigma_t n\, l), \tag{2}$$

where l is the scattering length in the cell. According to the theoretical result of Mathur *et al.*,[11] correction factor for the scattering length is close to unity in this geometry. Blaauw *et al.*[12] supported the theory of Mathur *et al.* in their work. Therefore, the inside measure between A_5 and A_6 was chosen as the scattering length $l=25.2$ mm.

In this experimental arrangement, many hours were needed to purge H_2O or D_2O vapour from the cell and the sensor head of the manometer after each exposure. During this time the vapour caused considerable zero point drift in the manometer. Therefore, instead of the zero point adjustment of the manoneter after complete evacuation of the cell at every measurement, the transmitted and the scattered current were measured at the starting pressure P_1 and at the desired pressure P_2, respectively, as discussed by Massey and Burhop.[13]

Let I_{01}, I_{c1}, I_{e1}, and I_{f1} and I_{02}, I_{c2}, I_{e2} and I_{f2} be the current intensities and n_1 and n_2 the target densities at target gas pressures P_1 and P_2, respectively. The relation between the σ_t and these quantities is given as

$$\sigma_t(n_2-n_1)l=\ln(I_{f1}I_{02}/I_{f2}I_{01}). \tag{3}$$

Appropriate bias potential was applied on the Faraday cup to prevent the contribution of inelastically scattered electrons to the transmitted current.

2.3 *Pressure measurement*

The pressure in the scattering cell at the temperature (T°K) was measured by the MKS Baratron capacitance manometer (type 310C). The sensor head of the manometer was kept at a constant temperature (45°C) during the operation. In this study the temperature (T) of the cell varied from about 20°C to 35°C. Taking into account the thermal transpiration effect,[14] reading of the manometer $P(T)$ was corrected by

$$P(T)/P(318.2)=(T/318.2).^{1/2} \tag{4}$$

Throughout this study the thermal transpiration effect on the target gas pressure was taken into account. Ferch *et al.*[15] took care of the temperature. Nickel *et al.*[16] discussed this effect in the error estimation. Zecca *et al.* maintained the temperature of the cell nearly identical to that of the sensor of the manometer.

According to the supplier's statement, the purity of the gases used in this work was 99.95% for Ar and N_2 and 99.5% for D_2O. The water (10 MΩ-cm or higher) used in this study was purified by means of the reverse osmosis with a deionizer.

2.4 *Electron beam and its energy*

In order to make the operation of the electron gun simple over the wide energy range the incident electron beam used in this experiment was not monochromatized. The incident electron beam intensity was kept lower than 10 pA to minimize the space charge effect along the electron path. Typically, the energy width was 1.25 eV at 100 eV incident electron energy.

Usually electron energy calibration is needed for the contact potential between different metals. For this purpose a sharp resonance line profile of suitable atoms or molecules has been detected by the electron spectroscopic method. On the other hand, the electron time of flight (TOF) spectroscopic method is also useful, especially for the determination of both the σ_t and the electron energy scale. The TOF spectroscopic method gives the energy scale directly from the measured data.[17] Since unselected electron beam was used in this study, the energy scale was calibrated for Ar comparing the measured broad peak in σ_t at around 15 eV with that of Ferch *et al.* whose result based on the TOF spectroscopic method. The scale determined for Ar was applied to the measurements for the other molecules through this work.

2.5 *Errors*

At a fixed electron energy, the measurement was carried out by changing the target gas pressure from 0.5 to 5 mTorr by 0.5–1 mTorr steps. Let σ_i be the measured value of the cross section at each gas pressure and $\bar{\sigma}$ the arithmetic average of the σ_i. Then, the standard deviation $\langle SD\rangle$ of the measured cross section is given as

$$\langle SD\rangle=[1/(n-1)]\left[\sum_{i=1}^{n}(\sigma_i-\bar{\sigma})^2\right]^{1/2}. \tag{5}$$

The value $\langle SD\rangle$ was taken as the statistical error of the measured value in this work.

The systematic uncertainties are introduced from the measurement of the currents, the target gas pressure including the temperature correction and the estimation of the scattering length. The overall systematic uncertainty is estimated to be 18%.

§3. Results and Discussion

The present results with the statistical errors are listed in Table I. The systematic error 18% should be added to each statistical error. The measurement was carried out under the single electron scattering condition. The σ_t deter-

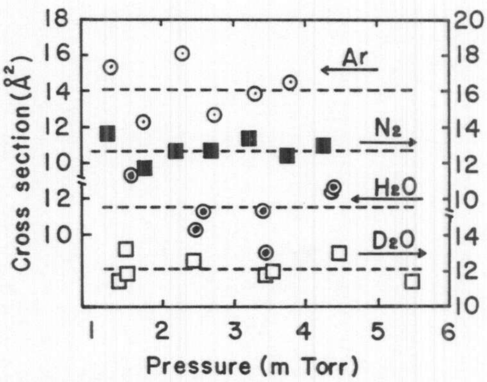

Fig. 2. Dependence of the cross sections on the target gas pressure.

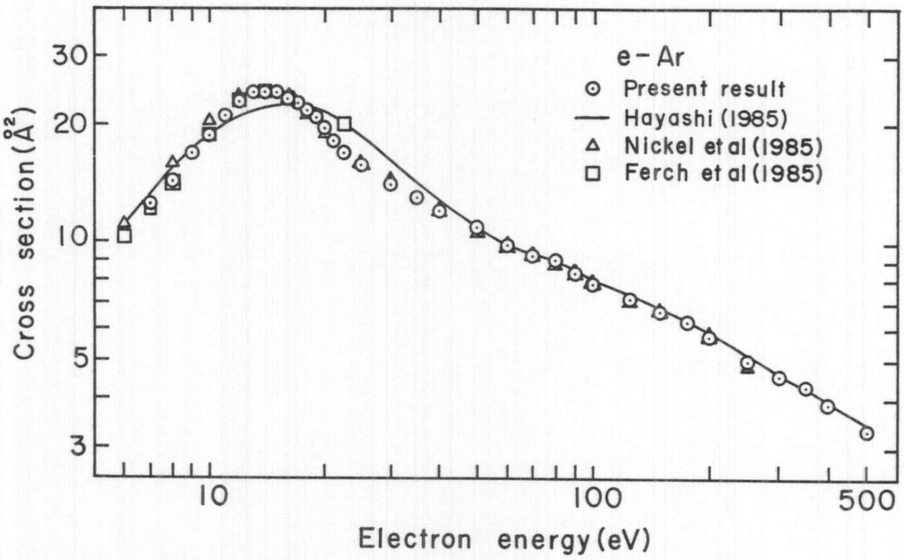

Fig. 3. Total electron scattering cross sections for Ar. Solid curve represents the recommended cross section of Hayashi. The others are the results measured by Ferch *et al.* (time of flight), Nickel *et al.* and the present work (electron transmission).

mined in this work was independent on the target gas pressure as shown in Fig. 2.

As mentioned above, the electron energy scale is calibrated measuring the σ_t for Ar. The present result is compared in Fig. 3 with the experimental results of Ferch *et al.* (1985),[15] Nickel *et al.* (1985)[16] and the recommended value given by Hayashi (1985)[18] which was compiled from the experimental data of electron scattering and the theoretical results prior to 1972. Agreement among the experimental results is very good. A small discrepancy between the experimental and the recommended results may be attributed to the deficiency of reliable results at that time. The elastic cross section for Ar can be replaced by the σ_t in the energy range below 20 eV because contribution of the inelastic scattering cross sections to σ_t is expected to be negligible. The elastic electron scattering cross section σ_{el} determined by Srivastava *et al.* (1981)[19] agrees very well with the present result in the energy range below 10 eV. The experimental σ_{el} given by Williams and Willis (1975)[20] and that of DuBois and Rudd (1976)[21] also show good agreement at 20 eV.

The measured results for N_2 are shown in Fig. 4 with the experimental results of Kennerly (1974)[22] and Sueoka and Mori (1984)[23] and the recommended value given by Hayashi (1985).[18] There is good agreement among the various results.

The present result for H_2O are compared in Fig. 5 with those of Brüche (1929), Sueoka *et al.* (1986, 1987), Zecca *et al.* (1987), Szmytkowski (1987) and Szmytkowski *et al.* (1987). The results agree very well with those of Brüche, Szmytkowski *et al.*, Zecca *et al.* in the energy compared. Agreement between the present results and that of Szmytkowski also very well above 15 eV. Discrepancy between these two results below 15 eV may be due to the difference of the energy width of electrons used in the measurement. Agreement between the present results and that of Sueoka *et al.* is good within the experimental error. While their result is lower about 25% than the present one at lower energy region. The reason for such discrepancy may be attributed to the effect of the magnetic field used in their measurement.

As shown in Table I, there is little difference between the σ_t for D_2O and that for H_2O within the experimental error. This is expected from the fact that these isotopic molecules have the same electronic structure and nearly identical dipole moment (1.94 ± 0.06D for H_2O, 1.87 ± 0.01D for D_2O).[24] Such fairly large dipole moment dominates in the electron scattering processes for these polar molecules,

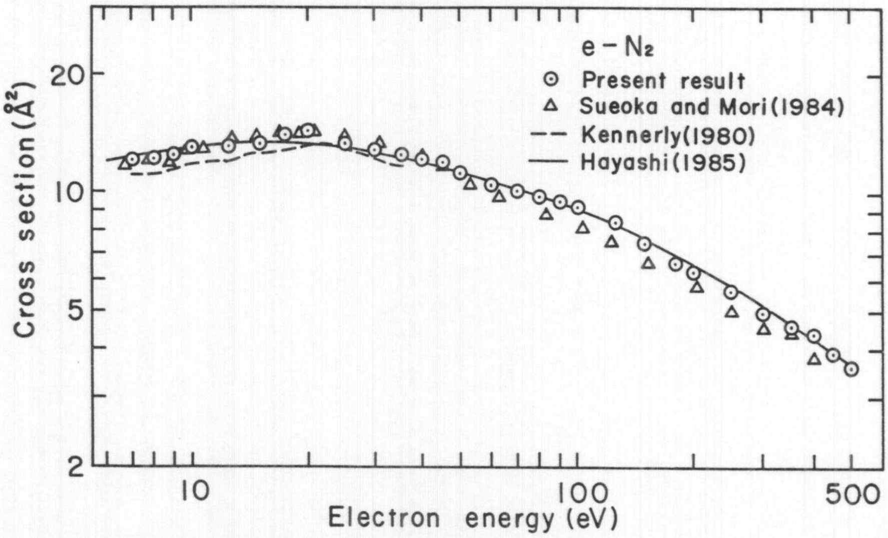

Fig. 4. Total electron scattering cross sections for N_2. Solid curve represents the recommended cross section of Hayashi. The others are the experimental results obtained by Kennerly and Sueoka (time of flight) and the present result (electron transmission).

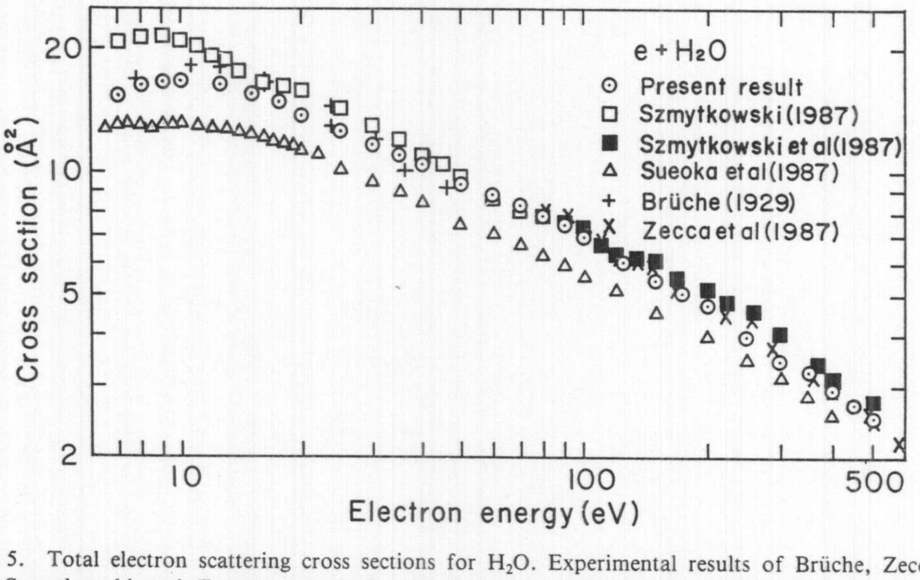

Fig. 5. Total electron scattering cross sections for H_2O. Experimental results of Brüche, Zecca *et al.* and Szmytkowski *et al.* (Ramsauer type), Sueoka *et al.* (time of flight) and Szmytkowski and present result (electron transmission).

whereas considerable differences exist in the vibrational molecular constants between those molecules due to the difference in the masses. The present results for H_2O and D_2O suggest that contribution from the vibrational excitation cross sections to the σ_t for these molecules is very small in the measured low energy range.

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Table I. Total cross sections (Å^2). The numbers in parentheses indicate statistical errors.

E (eV)	Ar	N_2	H_2O	D_2O
7	12.5(1.3)	12.0(1.7)	15.3(1.2)	16.3(2.5)
8	14.3(1.9)	12.2(1.6)	16.2(1.1)	16.5(1.5)
9	17.0(1.3)	12.4(1.2)	16.5(2.3)	16.7(2.1)
10	18.7(1.3)	13.0(2.3)	16.6(1.5)	17.3(1.6)
11	21.0(0.9)			
12	23.0(1.6)			
12.5		13.2(2.5)	16.4(1.2)	16.6(2.7)
13	24.2(1.6)			
14	24.3(1.8)			
15	24.3(1.7)	13.4(1.6)	15.5(0.4)	15.7(0.7)
16	23.4(0.8)			
17	22.9(2.2)			
17.5		13.9(1.0)	14.8(1.3)	15.3(1.4)
18	21.8(0.9)			
19	20.8(0.6)			
20	19.6(0.9)	13.7(1.6)	13.6(1.1)	14.6(1.1)
21	18.1(0.9)			
22.5	17.0(1.2)			
25	15.9(0.8)	13.5(1.3)	12.6(1.7)	13.1(0.7)
30	14.1(1.5)	12.8(0.7)	11.6(1.6)	12.1(0.8)
35	13.1(1.5)	12.4(0.6)	11.0(1.0)	11.7(1.9)
40	12.1(1.8)	12.2(1.4)	10.5(0.9)	11.2(1.0)
45		12.0(0.7)		
50	11.0(1.8)	11.4(1.2)	9.39(1.94)	10.2(0.3)
60	9.84(1.13)	10.6(0.6)	8.78(1.48)	9.87(0.75)
70	9.28(0.96)	10.2(1.3)	8.30(1.13)	8.62(1.22)
80	9.04(1.17)	9.75(1.22)	7.83(0.75)	8.19(1.23)
90	8.39(0.88)	9.51(0.57)	7.43(1.12)	7.69(0.93)
100	7.82(1.15)	9.24(1.28)	6.99(1.13)	7.29(1.20)
125	7.20(0.45)	8.43(0.90)	6.03(1.50)	6.28(0.81)
150	6.68(0.91)	7.45(0.66)	5.45(1.37)	5.59(1.29)
175	6.24(0.99)	6.61(0.57)	5.05(1.29)	5.10(1.31)
200	5.76(0.55)	6.33(0.54)	4.75(1.19)	4.80(1.52)
250	4.99(0.99)	5.65(0.79)	3.96(0.48)	4.08(0.96)
300	4.59(0.82)	4.95(0.49)	3.47(0.74)	3.78(1.23)
350	4.32(0.95)	4.66(0.80)	3.27(0.73)	3.27(0.47)
400	3.87(0.69)	4.40(0.47)	2.93(0.53)	2.97(0.68)
450		3.93(0.74)	2.70(0.23)	2.83(0.65)
500	3.27(0.29)	3.61(0.70)	2.50(0.69)	2.56(0.92)

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