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Measurements of total absolute cross sections for 0·2–100 eV electrons on H_2†

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Abstract. Absolute total electron scattering cross sections for H_2 have been measured from exponential attenuation in the energy range 0·2–100 eV with 0·07 eV FWHM energy resolution. Resonant structures in the 11–15 eV energy range are presented for the first time with their absolute size. The overall absolute error on the cross section was estimated to be from ●1·7 to ±5% in different energy regions. Comparison with previous experimental and theoretical results are given. Our measurements are higher than the previous ones although not further than the combined error. A fair agreement can be found with the calculations of Hara.

1. Introduction

Several fields of physics (astrophysics, plasma physics, high-atmosphere physics) (Massey and Burhop 1969) and of technological sciences (Rhodes and Szöke 1972) require the accurate knowledge of the electron–atom (and molecule) total cross sections at low energies. On the other hand, a number of theoretical papers on the electron–atom (molecule) interaction assess the need for new more precise experimental data (Takayanagi and Itikawa 1970, Golden *et al* 1971, Temkin 1976). The e^-–H_2 cross section is particularly interesting because it deals with the simplest electron–molecule system.

An experimental programme on the measurement of total absolute cross sections is being carried out at our laboratories. Two sets of apparatus are being used in the low and intermediate energy range: 0–100 eV and 100–2000 eV respectively.

This paper reports the results of the measurement of the e^-–H_2 total cross section in the energy range from 0·2 to 100 eV.

2. Experimental apparatus

2.1. Electron spectrometer

The electron spectrometer used in this work consisted of a linear electrostatic monochromator coupled with a gas chamber and a Faraday cup detector. The entire

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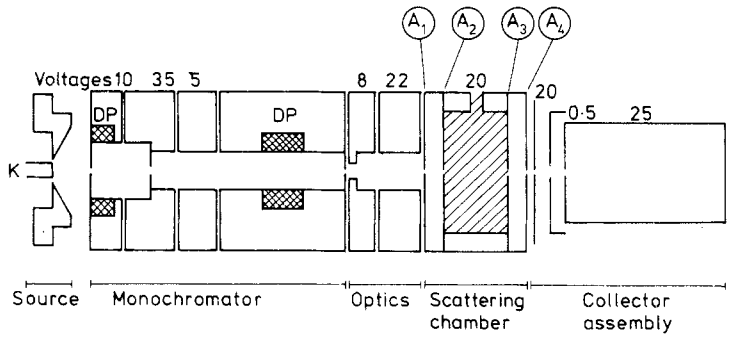

Figure 1. Scale drawing of the spectrometer: K, cathode; DP, deflection plates; a typical set of voltages for 20 eV operation is shown.

spectrometer was operated in a 'zero' magnetic field, the Earth's field being compensated through the use of square coils within ±10 mG.

The electrostatic monochromator worked on a principle originally suggested by Read *et al* (1974). Figure 1 shows a scale drawing of the entire spectrometer. The numbers in the upper part of the figure are typical voltages for 20 eV operation. The energy selection takes place at the last aperture of the part labelled 'monochromator'. It is known (Kuyatt 1969) that the pupil of the anode–cathode stage has a size which is different for electrons emitted with different energies. The monochromator consists of a system of lenses which makes an enlarged image of these pupils onto the exit aperture. The aperture being small, electrons are preferentially transmitted in the low-energy part of the original distribution. The details of this monochromator will be given elsewhere. The monochromator was operated at a typical beam intensity of 10^{-9} A and 0·07–0·08 eV FWHM. The energy resolution and the energy scale calibration were periodically checked on the 19·35 eV helium resonance. The high stability of this monochromator allowed the use of long measurement times.

Two gas cells were used, differing only in their length: 18 ± 0·2 mm and 50 ± 0·2 mm. The first cell was used at low energies (0–10 eV) and the second from 10 to 100 eV. The first one allowed for a higher current and the second one was used to obtain a better angular resolution. Figure 1 shows a scale drawing of the spectrometer. Note the peculiar design of the scattering chamber. The gas was confined by the apertures labelled A_2 and A_3. Aperture A_1 and A_4 were added: (i) to improve the gas pumping, (ii) to eliminate the field penetration from the neighbouring lenses in the gas region and (iii) to improve the scattered electrons angular resolution (this last point was discussed by Dalba *et al* 1979). Apertures A_1 and A_4 were 1·2 mm in diameter. A_2 and A_3 were 1·0 mm diameter. The larger cell was identical to this one except that the distance between A_2 and A_3 was 50 mm instead of 18 mm.

The detector was a shielded Faraday cup. Figure 1 shows, after aperture A_4, the shield aperture, then the retarding aperture and the collector aperture. With the voltages shown, the system builds up a simple retarding analyser for the scattered electrons. The energy resolution of such an analyser was measured to be about 0·5 eV. A positive voltage on the collector cup and a large ratio of total internal area to entrance area, were used to make the efficiency of the Faraday cup to be very nearly one.

The spectrometer was built from a non-magnetic alloy (arcap, grade AP4) and operated in a bakeable vacuum system at a base pressure of 10^{-9} Torr.

2.2. Data reduction formula

The cross section was obtained through an attenuation measurement: the collector current was measured at several gas pressures for each electron energy. The electron current transmitted through the gas at a pressure P (in Torr), at temperature T (K) is given by:

$$I(P, E) = I_0 \exp \frac{-l\sigma(E)}{3{\cdot}68 \times 10^{-3} T} \tag{1}$$

where E is the electron energy, I_0 is the current with no gas, l is the interaction region length (cm) and $\sigma(E)$ is the total cross section (a_0^2).

The interaction region length l was assumed to be the gas chamber length (A_2 to A_3 in figure 1). This was done after the two different cells had been used to measure the cross section around 10 eV. The two measurements gave the same results, within the instrumental reproducibility. This was taken as evidence that the end effects at the entrance and exit apertures of the gas region are small. It is to be noted that the end effects calculated through the theory of Mathur *et al* (1975) for our cells give an error much smaller than 1%, in agreement with our result. A similar test was performed in the intermediate energy apparatus (Dalba *et al* 1979), confirming that the interaction length can be assumed to be very close to the geometrical length of the gas cell.

2.3. Measurement procedure

It was impossible to get a cross section measurement in the full range from 0·2 to 100 eV with a single set of voltages on the spectrometer. Due to electron optical effects the current to the collector was a strongly varying function of the voltage applied to the gas chamber (i.e. of the scattering energy).

The measurements were made in several overlapping energy intervals. The intervals were chosen so that E_{max}/E_{min} was about 5. The spectrometer was tuned in each energy range in such a way that $I(E_{max})/I(E_{min})$ was between 0·95 and 1·00. This condition (flat transmission function), not required for relative measurements, is known to be a necessary condition for obtaining precise absolute measurements (Weingartshofer *et al* 1970).

After the spectrometer tuning, an on-line computer was used to measure the collector current as a function of energy at several pressures (5 to 9). Note that no direct measurement of I_0 is needed as long as I_0 is independent of the gas chamber pressure. This independence is assured by the use of a diverter valve (see § 2.4). The number of energy channels was between 100 and 500, the width being chosen between 0·01 and 0·5 eV. The computer was used to average a number of sweeps on the same energy range. The averaging procedure was used to improve the signal-to-noise ratio; it proved to be particularly useful when measuring the resonance region (figures 2 and 3). For each energy channel, the $\ln I(P, E)$ against P dependence was best fitted with a straight line. The linear correlation coefficient was computed and used as a reliability test of the experimental data. The value of the cross section was then computed from the slope of the best fit line.

The results presented later were obtained from 25 measurement runs with a total measurement time of about 60 hours.

The cross section values obtained in the same energy range with different runs were averaged. The averages obtained in different energy ranges were then joined to give the

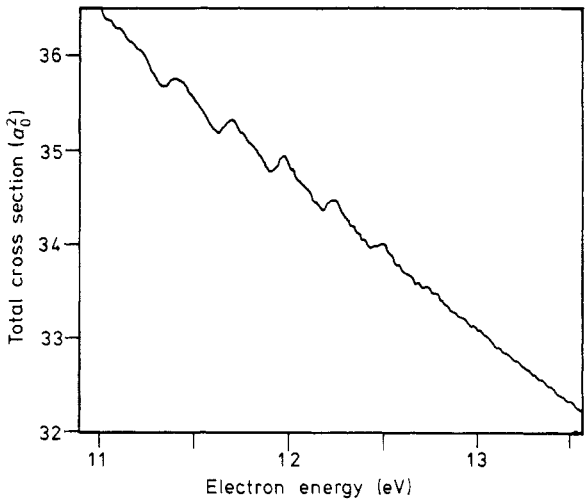

Figure 2. e^-–H_2 total cross section, 11–13 eV. The resonant series 'a' is visible.

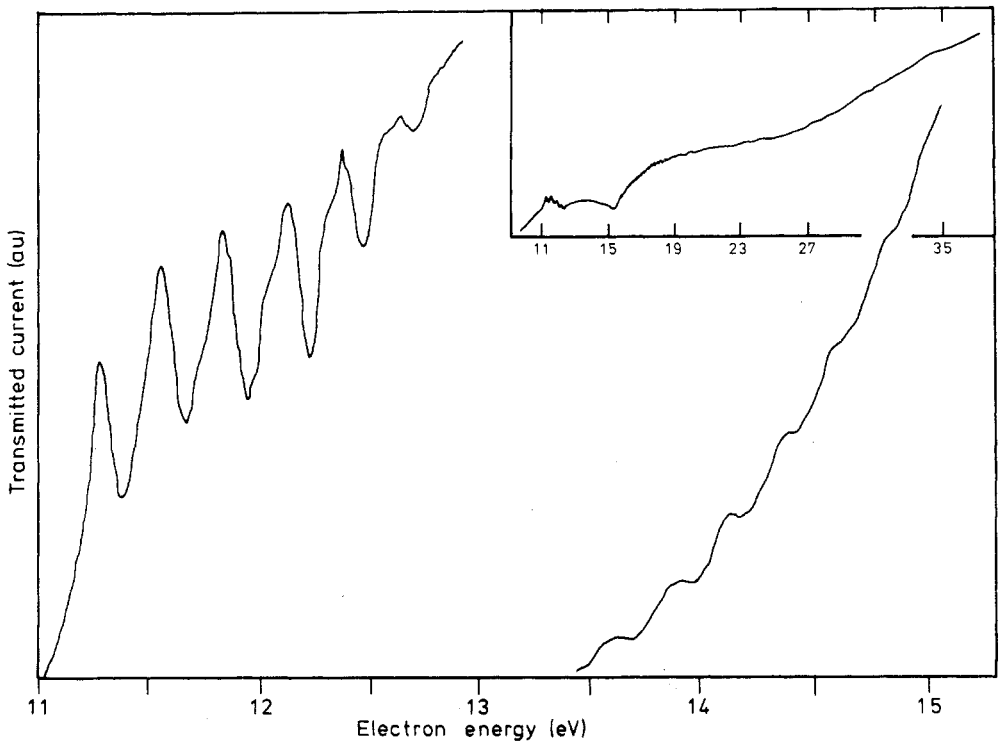

Figure 3. Transmitted current, arbitrary units, 11–15 eV. Series 'a', 'c' and 'f' are visible.

smooth-looking curves of figures 4 and 5. The junction was done by assuming a cross section value equal to the average of the two values for the range of overlap. Neighbouring points in the lower and higher energy range were then smoothed. This procedure was justified by the fact that the cross section discrepancy in the overlapping

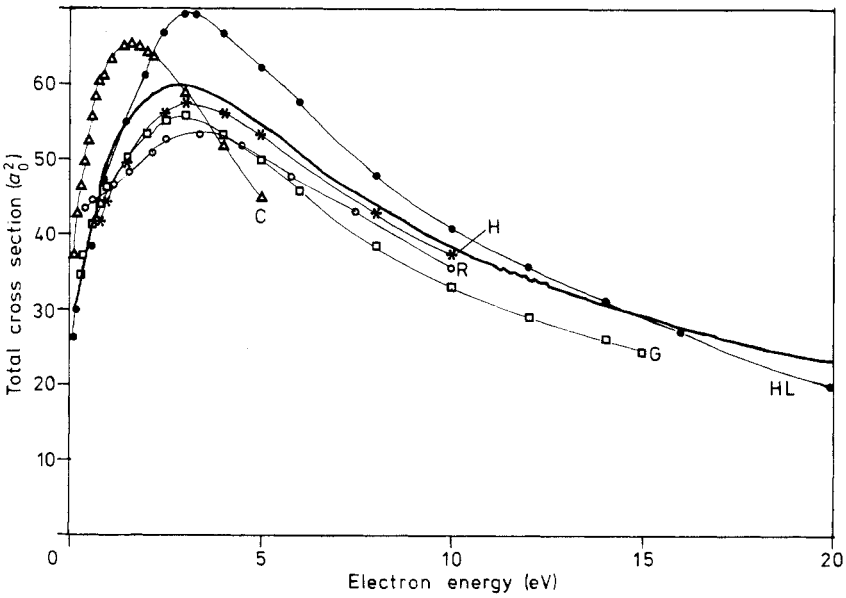

Figure 4. e^-–H_2 total cross section 0–20 eV. Full curve, present measurements; full circles, Henry and Lane (1969); asterisks, Hara (1969); open circles, Ramsauer and Kollath (1930); squares, Golden *et al* (1966); triangles, Crompton *et al* (1969).

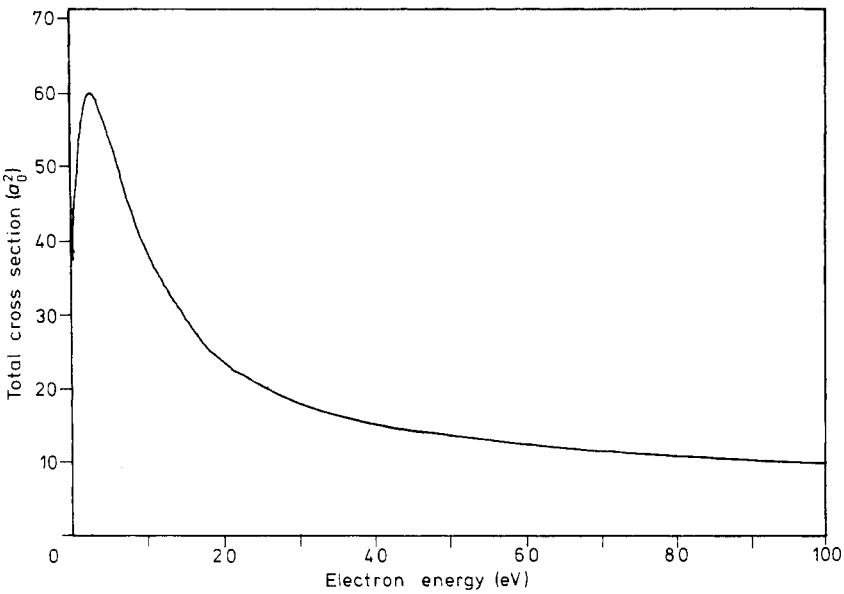

Figure 5. e^-–H_2 total cross section 0–100 eV: present measurements.

ranges was always smaller than the random error. This in turn was smaller than the absolute error, as estimated in § 4.2.6. Figures 2 and 3 are unretouched recordings of the cross section and transmitted current respectively in the 11–13·5 and 11–15 eV range.

2.4. Pressure measurement

The gas chamber pressure was measured with a capacitance manometer (Baratron 94 AH-1 from MKS). The head temperature was made to track the gas chamber temperature within 0·5 °C to avoid thermal transpiration errors. This temperature was higher than the ambient one due to the heat flow from the cathode. Both temperatures were measured with T-type thermocouples. The manometer was used with the MKS temperature compensation unit to allow for its temperature coefficient. The gas temperature was assumed to be the gas chamber temperature although this was questioned by Barbarito *et al* (1979). The reasons for assuming this equality are the following.

(i) The H_2 molecules suffer 600 collisions (average) with the walls before escaping from the gas chamber.

(ii) The accommodation coefficient was measured for noble gases on several surfaces (Kostoff *et al* 1967) and ranges from 0·1 to 1·0.

(iii) We assume that the value of the accommodation coefficient for H_2 on the gas chamber alloy is not very far from those values. As a consequence, a few tens of collisions should be enough to thermalise the gas with the gas chamber walls.

As a further remark we note that the background pressure around the electron spectrometer was held constant (whilst changing the gas chamber pressure) through the use of a diverter valve. The relevance of this precaution on the quality of the measurements was discussed by Dalba *et al* (1979). Here we note that a constant background pressure at the cathode and in the spectrometer is essential to get I_0 constant.

3. Results

3.1. e^-–H_2 cross section: 0 to 20 eV

Figure 4 shows the e^-–H_2 total absolute cross section in the 0–20 eV energy range. The lowest energy was 0·20 eV: below this value the scatter of the measured value increased rapidly. Correspondingly it was not possible to obtain a flat transmission function below 0·2 eV. Starting from this energy the cross section rises steeply. Around 2·9 eV the cross section shows a wide structureless maximum. This was attributed (Bardsley *et al* 1967, Ehrhardt *et al* 1968) to the formation of a shape resonance. The temporary state is the $^2\Sigma_u^+$ of the H_2^- ion and its lifetime is very short. For this reason its energy width is large and therefore no structure can be seen in the cross section maximum.

Above 3 eV the cross section decreases steadily with increasing energy. The resonances above 10 eV ('a' series) are barely visible in figure 4 as small oscillations in the cross section. Figure 2 shows a zero suppressed plot of the cross section as measured in the resonance region (11–13·5 eV). The 'a' series is ascribed to the formation of a Feshback-type core-excited resonance. The resonant state is $(1s\sigma_g)\,(2p\pi_u)^2\,{}^2\Sigma_g^+$ of the H_2^-; the parent state is the $c^3\Pi_u$ of H_2 (Eliezer *et al* 1967). In figure 2 each resonance presents a small shoulder at the higher energy side. These were attributed to the resonance series known as the 'c' series (Joyez *et al* 1973). These resonances are not clearly resolved due to lack of energy resolution. The resonant state was identified recently as $(1s\sigma_g)\,(2p\pi_u)^2\,{}^2\Delta_g$ of H_2^- (Chang 1975). A typical transmitted current plot is shown in figure 3 from 11 to 15 eV. The series 'a' is clearly shown; the shoulders of the 'c' series are more visible here. Above 13·5 eV another series (which was not visible in

figure 2) is shown. This is known as the 'f' (Ehrhardt and Weingartshofer 1967) series and the resonant state is $(1s\sigma_g)\ (3p\pi_u)^2\ ^2\Sigma_g^+$ of H_2^- (Weingartshofer *et al* 1970). The insert in figure 3 shows a transmitted current plot of the region from 11 to 35 eV. The 'a' series is barely resolved. Another wide structure is visible starting from about 15 eV. This structure was not observed in previous work, probably due to its large width. The structure is not instrumental although it does not appear in the total cross section given in figure 4.

3.2. e^-–H_2 cross section: 0 to 100 eV

Figure 5 shows the measured cross section up to 100 eV. The energy channel width used in the higher energy measurements was from 0·2 to 0·5 eV so that no structure can be seen.

4. Discussion

4.1. Comparison with previous results

Figure 4 reports the measurements of Ramsauer and Collath (1930) (open circles), Golden *et al* (1966) (squares) and Crompton *et al* (1968) (triangles). Note that the measurements of Crompton *et al* were performed with a swarm technique and give a momentum transfer cross section. The measurements of Crompton *et al* are believed to be accurate within a few per cent (Bederson and Kieffer 1971) but can be compared directly with total cross sections only in the limit of very low energies. The present data are higher than Ramsauer's and Golden's data throughout the range studied except at energies below 0·5 eV.

The oldest data of Ramsauer and Kollath are believed to be inaccurate by some 10% or more. This inaccuracy can be attributed mostly to the inaccuracy of the pressure measurements.

The measurements of Golden *et al* (1966) were performed with a Ramsauer-type apparatus and were quoted by the authors to be good within 3%. Bederson and Kieffer (1971) give an analysis of their experiment and suggest that an appropriate error limit could be 10%. The differences between the present experimental set-up and the one used by Golden *et al*, are the following. Firstly, we used a capacitance meter (MKS-94 AH-1) which is sold by the manufacturer with a stated accuracy of ±0·15% in our measurement range. Golden *et al* used an ion gauge which was calibrated against a McLeod gauge. Thermal transpiration from the ion gauge to the scattering chamber was not accounted for in the data analysis. The calibration against a McLeod gauge is known to be a difficult task which can be vitiated from several error sources. Secondly, the pumping scheme used in Golden's apparatus does not give good differential pumping. More importantly, the use of a 'pressure-dropping channel' can introduce an indetermination in the interaction length measurement. Thirdly, both the energy and angular resolution are better in the present apparatus.

As a conclusion of the comparison of the present data with previous measurements, it can be stated that both Golden's and Ramsauer's data are not conflicting with the present ones. As will be quoted in § 4.2.6, the absolute error of the present data is probably a factor of 3–5 times smaller than the error in Golden's measurement.

Figure 4 also shows the theoretical data of Henry and Lane (1969) (full circles) and Hara (1969) (asterisks). Henry and Lane used a close-coupling approximation; Hara

used a fixed-nuclei calculation. Our results are a few per cent higher than the results of Hara. The results of Henry and Lane lie more than 10% higher than our measurements in the region of the cross section maximum and they become about 15% lower at 20 eV.

Table 1 shows our measured cross section for several energies compared with Golden *et al* (1966), Henry and Lane (1969) and Hara (1969).

Table 1. e^-–H_2 total scattering cross section (units a_0^2).

Energy	Present	Golden *et al* (1966)	Henry and Lane (1969)	Hara (1969)
0·3	33·1	34·8		
0·6	41·3	41·4	38·5	
1·0	50·0	46·5	46·8	44·1
1·5	54·9	50·3	55·2	49·2
2·0	57·7	53·4	61·0	52·8
2·5	59·5	55·2	66·7	56·1
3·5	59·1		68·8	
4·5	56·4			
6·0	50·8	46·0	57·7	46·6
8·0	44·1	38·7	47·9	42·8
10·0	38·5	33·2	41·0	37·4
15·0	29·3	24·7		
20·0	23·4		20·0	
30·0	17·9			
40·0	15·1			
50·0	13·6			
60·0	12·4			
70·0	11·5			
80·0	10·8			
90·0	10·2			
100·0	9·8			

4.2. Error evaluation

4.2.1. Current measurement. The collector current was measured with a digital electrometer used in a single range. Therefore, only the non-linearity of the electrometer enters into the cross section error. Our electrometer was linear within 0·1%. With the current attenuation used, the error introduced in the cross section was estimated to be less than 0·6%.

4.2.2. Pressure measurement. The accuracy quoted by the manufacturer for the capacitance manometer was ±0·15% at pressures higher than 10^{-3} Torr. A conservative figure of ±1% will be used in the following.

The thermal transpiration error was limited to a temperature differential of ±0·5 °C. This limits the error on the cross section to ±0·2%. The error introduced by the temperature measurement was less than ±0·2%. The hydrogen gas had a certified purity of 99·999%; the contaminants were: $N_2 < 3$ ppm; $O_2 < 2$ ppm; $H_2O < 3$ ppm.

4.2.3. Energy measurement. The energy scale was calibrated by comparing the position of the first four peaks in the 'a' series with the values given by McGowan *et al* (1974). By

taking into account the accuracy claimed in this work, the accuracy of our measurements and the discrepancy of the four determinations for the energy zero, the overall accuracy of our energy scale can be evaluated to be ±0·05 eV. This was about the same accuracy obtainable with the 19·35 He resonance. A 0·05 eV figure reflects on the cross section accuracy as a ±7% at 0·2 eV, less than 3% from 0·6 to 1 eV, less than 1% up to 2 eV and less than 0·3% in the remaining energy interval. The field penetration in the gas chamber was considered negligible, due to the chamber geometry (figure 1).

4.2.4. Angular resolution. The error due to the finite angular resolution was evaluated taking into account the different values for the angular resolution of the two cells. The error was calculated from the available differential elastic cross section data. As a matter of fact the contribution of small-angle inelastic electrons can be neglected because of the use of the retarding field analyser.

The elastic differential data used from 0·2 to 2 eV are those of Linder and Schmidt (1971): they give an error (for our geometry) of less than ±0·15% up to 2 eV. From 3 to 75 eV the differential calculations of Srivastava *et al* (1975) were used giving an error of less than ±0·5% up to 10 eV, less than ±1% up to 60 eV and less than 1·2% up to 75 eV. The error is about 3% at 100 eV, using the results of Huang and Chan (1977).

4.2.5. Interaction chamber length. The percentage error for the 18 mm chamber was ±1%; for the 50 mm chamber, it was ±0·4%.

4.2.6. Overall error. The overall error was calculated as the quadratic sum of the single errors. Taking into account the energy dependence of some source of error, the overall absolute error is a little more than ±7% at the lowest energy, decreasing to 3·5% up to 1 eV; 2% from 1·2 to 2 eV; 1·7% from 2·0 to 15 eV; rising to 1·9% from 15 eV to 75 eV; 3·5% from 75 to 100 eV. Note that the random error is smaller than the absolute one for energies above 0·6 eV. Starting from this energy, the random error increases to ±15% at 0·2 eV.

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