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Elastic differential cross section measurements for electron scattering from Ar and O_2 in the intermediate-energy range

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Abstract. In this paper we report experimental elastic differential, integral and momentum transfer cross sections for electrons scattered by Ar and O_2 in the impact energy range 300-1000 eV and in the angular range 5-140°. In our experiments, the intensities of the elastically scattered electrons were obtained in the crossed-electron-beam-molecular-beam geometry and the absolute values of the cross section were derived through the relative-flow technique using N_2 as the secondary standard. For both gases, the cross sections are compared with the available experimental and theoretical results.

1. Introduction

The interaction between electrons and atoms (and molecules) has been the subject of extensive theoretical and experimental investigations (Bransden and McDowell 1977, 1978, Lane 1980, Trajmar *et al* 1983) in recent years. However, most of the studies reported in the literature concern interactions in the low-energy range and in small systems such as light atoms and diatomic molecules. Considering the recent progress in the theoretical studies of electron-molecule scattering in intermediate- and high-energy ranges (Jain and Thompson 1982, Gianturco *et al* 1985, Lee *et al* 1985), and particularly for polyatomic molecules, the need for experimental absolute cross sections becomes apparent, since the applicability of these theories should be tested against experimental measurements over wide energy and angular ranges.

In this work, we report experimental values of the differential, integral and momentum transfer cross sections derived from the relative-flow technique for electrons elastically scattered from Ar and O_2 at impact energies in the range 300-1000 eV. The relative-flow technique (Srivastava *et al* 1975), which is currently in wide use, has the advantage that it gives absolute values of the elastic and inelastic cross sections without explicit knowledge of the scattering geometry (target density distribution, electron density distribution, scattering volume, etc), which is difficult to determine experimentally. One of the aims of this work is to establish the relative-flow technique in the intermediate-energy range; in this context, Ar is a suitable target since, historically, e^--Ar scattering has been extensively investigated both theoretically and experimentally. Once the applicability of this technique is confirmed, as in our case, the elastic cross sections of a variety of systems in the intermediate-energy range can be determined.

Elastic differential cross sections have been reported by Bromberg (1974b), Williams and Willis (1975), Jansen *et al* (1976) and Dubois and Rudd (1976) for

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electron-Ar scattering at intermediate energies, while theoretical studies of the electron-Ar interaction have been made by Walker (1971) in the relativistic approach and by Khare and Shobha (1974) in the plane-wave approximation. McCarthy *et al* (1977) and Joachain *et al* (1975, 1977) have also calculated cross sections for elastic electron-Ar scattering in the optical model potential approach.

Electron-oxygen scattering is a very important interaction in atmospheric studies, since oxygen is one of the major constituents of the Earth's atmosphere; however, there are relatively few experimental and theoretical investigations in the literature. The overall situation has been reviewed recently by Trajmar *et al* (1983). In the intermediate-energy range, absolute elastic differential cross sections for e^--O_2 scattering have been reported by Bromberg (1974a) at 300-500 eV and over the angular range 2-40°. Normalised cross sections have also been reported by Wakiya (1978) and Daimon *et al* (1982) for impact energies up to 500 eV and scattering angles below 150°. At these intermediate energies, theoretical calculations of e^--O_2 cross sections have been reported (Hayashi and Kuchitsu 1976, 1977, Daimon *et al* 1982) in the modified version of the independent-atom model including the intramolecular multiple-scattering effect.

In this work, values of the differential, integral and momentum transfer cross sections for electrons elastically scattered by Ar and O_2 at incident energies in the range 300-1000 eV and in the angular range 5-140° are reported. Some of these results have not, to our knowledge, appeared previously. Our data are compared with the available theoretical and experimental results. For oxygen, we also report cross sections calculated using the modified independent-atom model including the intramolecular multiple-scattering effect in the same way as Hayashi and Kuchitsu (1976).

2. Experimental details

A schematic representation of the experimental set-up is shown in figure 1. The scattering experiment is carried out inside a vacuum tank with a diameter of 60 cm and a height of 30 cm. A pumping system consisting of a 6 in oil diffusion pump equipped with a liquid nitrogen trap provides an ultimate pressure of 1.0×10^{-6} Torr. The magnetic field in the scattering region is kept at around 15 mG by three sets of Helmholtz coils. A crossed-beam geometry is used for the e^--gas collision. The electron beam is extracted from a hairpin tungsten filament and is accelerated, focused and

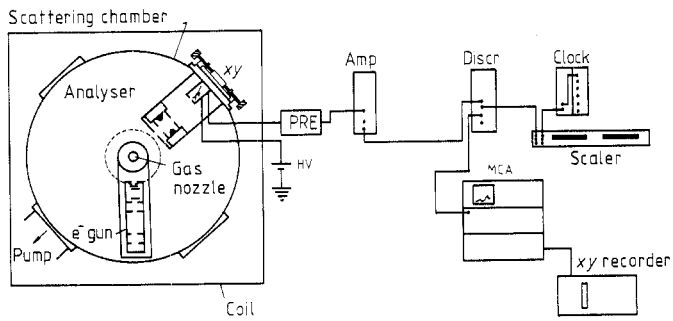

Figure 1. Schematic representation of the apparatus used to measure the elastic scattering intensities.

crossed with the target beam without prior energy selection. The electron gun provides a typical beam current of around 4.0×10^{-6} A and an approximate beam diameter in the scattering region of around 1 mm. The target beam is formed by passing the sample gas through a capillary array nozzle. This nozzle has a diameter of 0.10 cm, with individual capillaries of 50 μm diameter and 5 mm long (aspect ratio $\gamma = 0.01$). In our experimental set-up the electron gun is allowed to rotate around the scattering centre from -130 to $+140°$. The rotation of the gun is facilitated by a high-precision positioning mechanism (ORTEC 3701B) which allows the angle scale to be read to within an accuracy of 0.05°. Nevertheless, the angular resolution, defined by the set of apertures in front of the detection system, is 0.2° in our experiment. This set of apertures also limits the solid angle covered by the detector view cone; in the scattering region the circular base of the cone has a diameter of around 2 mm.

The scattered electrons, after passing through the set of apertures are energy selected by a Möllensted velocity analyser (Peixoto *et al* 1979) and the intensity of the elastically scattered electrons is counted by a channeltron electron multiplier. The energy resolution of this system is around 1.5 eV. This energy resolution allows a complete discrimination of the electrons resulting from the electronic excitation and ionisation processes, although the measured elastic scattering intensities are vibrationally unresolved for the electron-molecule scattering. A typical energy loss spectrum for electron-Ar scattering at 2.0° and 600 eV is shown in figure 2.

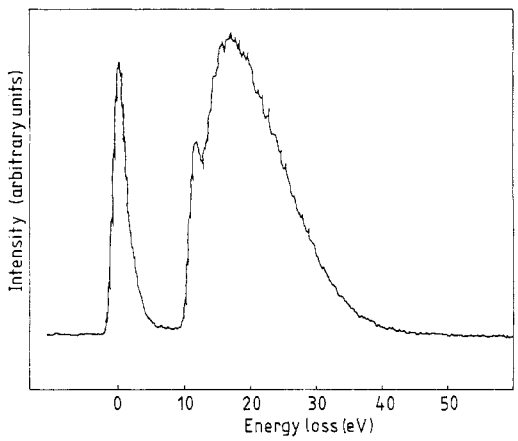

Figure 2. A typical energy loss spectrum for the electron-Ar interaction at 600 eV and 2°.

The absolute differential cross section is related to the measured intensity of the elastically scattered electrons by the expression (Srivastava *et al* 1975)

$$\dot{N}_e = k \int_V \int_\Omega \sigma'|\theta(x, y, z)| F_e(x, y, z) n_b(x, y, z)\, d\Omega\, d\tau \tag{1}$$

where $\dot{N}_e$ is the number of the scattering electrons per second, k is the transmission efficiency of the analyser-detector system, $\sigma'|\theta(x, y, z)|$ is the differential cross section for scattering from the element of volume $d\tau$ located at (x, y, z), $F_e(x, y, z)$ is the flux of electrons crossing unit area located in $d\tau$ and n_b is the density distribution of the target gas in $d\tau$.

The experimental determination of the physical quantities k, F_e and n_b is difficult except for the case of a steady target. These quantities are dependent on the geometry of the experimental set-up and also on the experimental conditions; hence, if the scattering conditions are kept unchanged during the measurements of the scattering intensities from two different gases and if the flow of the target gases through the capillary array is of molecular type ($K_{nD} \geq 1$), one can obtain the following expression for the ratio of the intensities (Srivastava *et al* 1975):

$$(\dot{N}_e)_1/(\dot{N}_e)_2 = (\sigma_1(\theta)/\sigma_2(\theta))(p_2/p_1) \tag{2}$$

where p_1 and p_2 are the absolute pressures measured for both gases in the gas reservoir. If the absolute elastic differential cross section (EDCS) for the electrons scattered by a known specimen (the secondary standard) is known, one can obtain from equation (2) the absolute value of the EDCS for the sample gas under study. In this work, N_2 was chosen as the secondary standard because of the numerous experimental absolute cross sections reported in the literature in this energy range.

In order to fulfil the conditions already discussed, some precautions were taken. The electron gun was turned on at least three hours before the measurement was carried out. During this period, the stability of the beam current was monitored by a Faraday cup. For each impact energy, the measurement of the scattering intensity was performed for one gas, and the measurements were subsequently repeated for another gas under the same experimental conditions. During the experiments, the primary beam current was monitored from time to time by a Faraday cup connected to a Keithley 602 electrometer and the pressure of each gas in the gas reservoir was calibrated using a McLeod gauge. In addition, measurements of the scattering intensity as a function of the gas pressure in the reservoir were performed for both gases at three scattering angles (30, 40 and 50°) and the results showed that the intensity is a linear function of the pressure in the reservoir in the interval 0.5–2.0 Torr. Therefore, in our experiment, the pressure in the gas reservoir was set between 1.0 and 2.0 Torr for all the measurements.

For each pair of gases, this procedure was repeated several times over different periods. The contribution of the background scattering was also determined in all the measurements: here the sample gas was introduced into the system through a side inlet. The measured scattering intensities for each gas, after subtraction of the background contribution, were used to obtain the ratio of the intensities at each scattering angle. These ratios from the various experiments were then used to obtain the absolute cross sections from equation (2). It was found that the primary beam current stabilised at slightly different values from one gas to another during the measurements of the scattering intensities for the pair N_2/O_2. This variation of the beam current was also conveniently corrected for in the calculation of the absolute cross sections.

3. Experimental uncertainties

Besides the statistical uncertainties, the most common sources of systematic and random experimental errors are the fluctuation of the primary electron beam current, pressure fluctuations, uncertainty in the scattering angle, scattering by delocalised gases and background scattering from surfaces inside the vacuum chamber.

In our experiments, the primary beam current was monitored by a Faraday cup attached to a Keithley 602 electrometer. It was verified that fluctuations during the measurement did not exceed 1%. In addition, the scale of the electrometer allows an

uncertainty of 0.5%, so the overall uncertainty in the primary beam current is estimated to be 1.2%; this gives rise to uncertainties in the scattering intensities of the same order. The fluctuations in the sample pressure were verified by a Varian 845 vacuum ionisation gauge meter to be less than 0.5%. However, the main source of the uncertainty in the pressure is the measurement of the absolute gas pressure in the reservoir: in this case, the maximum uncertainty is 2% for each gas, which leads to an error of 3% in the cross sections. The main source of the angular uncertainty is the determination of the zero scattering angle, since the mechanical imperfection of our positioning mechanism does not exceed 0.05°. The zero scattering angle in our case is determined by the symmetry of the scattering in the positive and negative angular regions, with an uncertainty of 0.2°. This angular error can contribute up to 6% of the scattering intensity at small angles. However, for large scattering angles, the contribution of this uncertainty does not exceed 1%. An additional error in the scattering intensity is due to the angular resolution of 0.2° defined by the set of apertures; in this case, it has been estimated to be 1%.

The scattering of the delocalised gases and the background scattering were measured for each gas by introducing the sample gas, at the same working pressure, through a side inlet. In our experiment, this contribution never exceeded 10% of the intensity. Thus we estimate an uncertainty of 2% in the scattering intensity.

There are also uncertainties caused by the residual magnetic field, the fluctuation of the incident energy, etc. However, in the energy range 300–1000 eV these effects are small and are estimated not to exceed 1% of the intensity.

We must now consider the statistical uncertainties, which are taken as the square root of the counting rate and vary with the scattering angle, from up to 6% at large angles to less than 1% at small angles.

The overall uncertainty in the cross sections is obtained from the equation

$$\Delta = \left(\sum_i d_i^2 + D^2 \right)^{1/2} \tag{3}$$

where d_i is the uncertainty in each parameter as discussed above and D is the quoted uncertainty in the absolute cross sections of the secondary standard. In this work, we used as the secondary standard the absolute EDCS of N_2 obtained by Dubois and Rudd (1976) at impact energies up to 800 eV, with the quoted uncertainty of 12%. At 1000 eV, the absolute EDCS of N_2 measured by Jansen (1975) with a 6% uncertainty was used. The overall uncertainty is therefore around 15% for impact energies below 800 eV and 10% at 1000 eV.

4. Results and discussion

The elastic differential, integral and momentum transfer cross sections for e^-–Ar and e^-–O_2 scattering at impact energies in the range 300–1000 eV are shown in tables 1 and 2 respectively. In this work, the integral and momentum transfer cross sections were obtained through a numerical integration using a Simpson quadrature, where contributions of the differential cross sections for angles larger than those determined experimentally were neglected. For the e^-–Ar interaction the integral cross sections of Jansen (1975) and Dubois and Rudd (1976) are also shown. We note that there is agreement between these experimental results except at 500 eV, where our results for the integrated cross section are respectively about 17 and 25% lower than those obtained by Jansen (1975) and by Dubois and Rudd (1976). However, these discrepancies still

Table 1. (*a*) Elastic differential and (*b*) integral and momentum transfer cross sections for e^--Ar scattering (in atomic units). Numbers in parentheses denote powers of ten.

(*a*)

	Energy (eV)			
Angle (deg)	400	500	800	1000
5	3.93 (1)	3.16 (1)	2.69 (1)	3.32 (1)
10	1.57 (1)	1.46 (1)	8.95	1.04 (1)
15	6.62	5.94	3.55	3.45
20	3.22	2.69	1.85	1.42
25	1.99	1.38	9.49 (−1)	8.00 (−1)
30	1.21	8.47 (−1)	5.37 (−1)	5.18 (−1)
35	8.32 (−1)	5.97 (−1)	3.44 (−1)	3.12 (−1)
40	5.99 (−1)	4.23 (−1)	2.52 (−1)	2.26 (−1)
45	5.02 (−1)	2.79 (−1)	1.71 (−1)	1.63 (−1)
50	3.47 (−1)	2.10 (−1)	1.43 (−1)	1.20 (−1)
60	2.00 (−1)	1.37 (−1)	8.53 (−2)	6.35 (−2)
70	1.41 (−1)	9.89 (−2)	6.02 (−2)	4.69 (−2)
80	9.48 (−2)	6.58 (−2)	4.95 (−2)	3.66 (−2)
90	7.10 (−2)	5.43 (−2)	4.01 (−2)	2.93 (−2)
100	6.25 (−2)	5.05 (−2)	4.04 (−2)	2.72 (−2)
110	7.06 (−2)	5.29 (−2)	3.94 (−2)	2.59 (−2)
120	9.58 (−2)	5.88 (−2)	3.91 (−2)	2.55 (−2)
130	—	—	4.35 (−2)	2.64 (−2)
140	—	—	4.62 (−2)	2.66 (−2)

(*b*)

	Energy (eV)			
Source	400	500	800	1000
Integral				
This work	7.61 ± 1.52	6.09 ± 1.22	4.67 ± 0.93	4.81 ± 0.96
Jansen (1975)	8.01	7.12		4.68
Dubois and Rudd (1976)		7.61	4.81	
Momentum transfer				
This work	1.01 ± 0.20	7.24 ± 1.45 (−1)	7.99 ± 1.60 (−1)	6.51 ± 1.30 (−1)

lie within the error bars. For O_2, there are no other experimental integrated cross section results for comparison.

In figure 3 we compare our experimental results for the elastic differential cross sections for the e^--Ar interaction with the experimental values reported by Jansen *et al* (1976) the theoretical results of McCarthy *et al* (1977). At 400 eV the comparison is extended to the experimental results of Williams and Willis (1975) and the theoretical values of Walker (1971), and at 500 eV to the experimental results of Dubois and Rudd (1976). The theoretical results of Joachain *et al* (1975, 1977) are essentially the same as those of McCarthy *et al* (1977), and for this reason they are not included in figure 3.

Our results agree very well with the experimental results of Jansen *et al* (1976) for all these energies; the maximum discrepancy does not exceed 10%. Our experimental values at 1000 eV also agree very well with the theoretical results of McCarthy *et al* (1977) for scattering angles lower than 80°, whereas our results lie approximately 10%

Table 2. (*a*) Elastic differential and (*b*) integral and momentum transfer cross sections for e^--O_2 scattering (in atomic units). Numbers in parentheses denote powers of ten.

(*a*)

	Energy (eV)			
Angle (deg)	300	400	500	1000
5	5.40 (1)	4.52 (1)	3.99 (1)	2.50 (1)
10	2.12 (1)	2.09 (1)	1.56 (1)	7.44
15	8.70	7.12	5.73	2.62
20	4.00	3.46	2.67	1.76
25	2.13	2.30	1.83	9.40 (−1)
30	1.50	1.61	1.27	4.95 (−1)
35	1.09	1.19	7.12 (−1)	3.18 (−1)
40	9.20 (−1)	8.03 (−1)	4.99 (−1)	2.25 (−1)
45	5.95 (−1)	4.81 (−1)	3.17 (−1)	—
50	4.70 (−1)	3.44 (−1)	2.47 (−1)	1.23 (−1)
55	3.68 (−1)	2.78 (−1)	—	—
60	3.03 (−1)	2.45 (−1)	1.95 (−1)	7.40 (−2)
70	2.28 (−1)	1.96 (−1)	1.33 (−1)	4.55 (−2)
80	1.72 (−1)	1.42 (−1)	9.63 (−2)	3.34 (−2)
90	1.42 (−1)	1.09 (−1)	8.20 (−2)	2.52 (−2)
100	1.43 (−1)	1.03 (−1)	7.03 (−2)	1.95 (−2)
110	1.32 (−1)	9.54 (−2)	6.30 (−2)	1.68 (−2)
120	1.20 (−1)	9.24 (−2)	5.18 (−2)	1.56 (−2)

(*b*)

	Energy (eV)			
Source	300	400	500	1000
Integral				
This work	1.03 ± 0.21 (1)	9.08 ± 1.82	7.15 ± 1.41	3.74 ± 0.75
Momentum transfer				
This work	1.60 ± 0.32	1.31 ± 0.26	9.21 ± 1.84 (−1)	3.53 ± 0.71 (−1)

lower for larger angles. At 500 eV our results agree with the experimental data of Dubois and Rudd (1976) within the experimental uncertainties; good agreement is also obtained with the theoretical values of McCarthy *et al* (1977). However, a major discrepancy is observed when our results for 400 eV are compared with the experimental data obtained by Williams and Willis (1975). In this case, although the shape of the two angular distribution curves is similar, the quantitative agreement is poor: at some large scattering angles, the discrepancy is over 50%. Nevertheless, the comparison with theory at 400 eV shows that our data are in fairly good agreement with the results of Walker (1971). The values of McCarthy *et al* (1977) agree with our experimental results only at scattering angles lower than 20°; at larger angles their results are always about 20% lower, although the shape of the curve is well reproduced.

Figure 4 shows our experimental results for elastic differential cross sections for e^--O_2 scattering. At 400 and 500 eV, a comparison is made with the experimental data reported by Bromberg (1974a) and Daimon *et al* (1982). We observe that there is agreement between the experimental results; the major discrepancy between our values and other experimental data does not exceed 10%, which is within the experimental

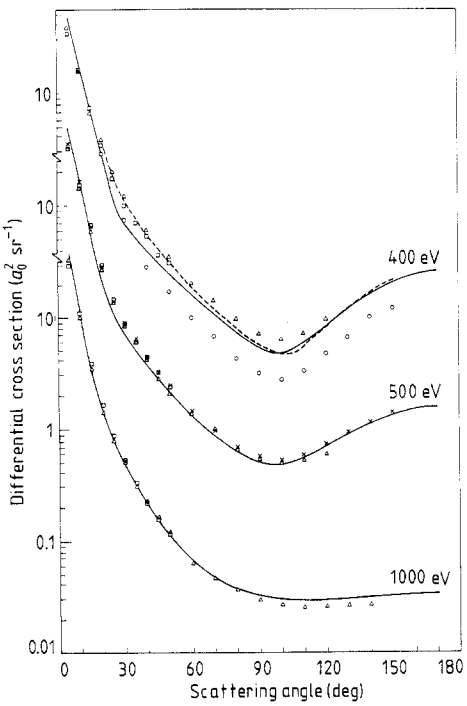

Figure 3. Elastic differential cross sections for e^--Ar scattering at 400, 500 and 1000 eV: △, present work; ○, Williams and Willis (1975); ×, Dubois and Rudd (1976); □, Jansen *et al* (1976); ——, McCarthy *et al* (1977); ---, Walker (1971).

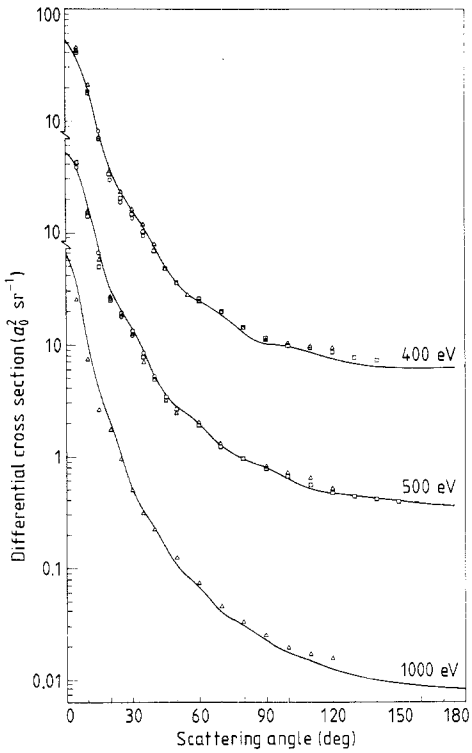

Figure 4. Elastic differential cross sections for e^--O_2 scattering at 400, 500 and 1000 eV: △, present work; ○, Bromberg (1974a); □, Daimon *et al* (1982); ——, independent-atom model calculations including the multiple-scattering effect.

uncertainties. For higher energies there are no experimental results available for comparison.

On the other hand, it is known that the theory for electron-molecule scattering at intermediate energies is not yet well established. One of the simplest theoretical models, the independent-atom model (IAM) is still widely in use. However, it is known that the IAM with the partial-wave method gives values of EDCS which are too large at large scattering angles (Hermann *et al* 1976, Khare and Raj 1979, Choi *et al* 1979). Several authors state that the inclusion of the intramolecular multiple scattering (IMS) can reduce significantly the difference between experiment and theory. In this work, we have calculated the DCS for the elastic e^--O_2 interaction using the modified independent-atom model including the intramolecular multiple scattering (Hayashi and Kuchitsu 1976). In our calculations, only the short-range Yukawa-type potentials (Cox and Bonham 1967) have been considered and the non-local exchange effect and the long-range polarisation potential have been neglected. The multiple-scattering effect has been truncated to include double scattering only. These calculated values are also plotted in figure 4. At 400 and 500 eV, our experimental data agree with the IAM-IMS results within the experimental uncertainties. It is interesting to note that the non-inclusion of the polarisation effect in the calculation seems to be well compensated by the lack of binding effects in the independent-atom model. At 1000 eV, however,

the polarisation effect becomes less important. The IAM results at small angles lie above our experimental data by approximately 25%; for larger angles, the agreement is fair. In addition, our calculation shows that at 300 eV the IMS reduces the differential cross section by up to 12% at larger scattering angles, whereas at 1000 eV the contribution of the IMS to the EDCS oscillates around 2%, never exceeding 5% over the entire angular range.

In contrast to the situation for e^--Ar scattering, both theoretical and experimental curves for the EDCS from the e^--O_2 collision show oscillating features. These are the diffraction patterns of the electron-molecule scattering and are better observed in figure 5, where the reduced scattering intensities at two incident energies are shown. The reduced scattering intensity is defined as

$$I_R = \text{EDCS}/I_{at} \tag{4}$$

where I_{at} is the scattering intensity of two oxygen atoms, which was obtained by the partial-wave method using the Yukawa-type (Cox and Bonham 1967) atomic potential.

It can be seen that the diffraction pattern of the e^--O_2 scattering follows a sine function and the experimental data reproduce the shape fairly well, particularly at small scattering angles.

5. Conclusions

From the comparison of the EDCS obtained by the relative-flow technique in this work with the absolute values reported in the literature for e^--Ar and e^--O_2 scattering, one can conclude that the applicability of this technique in the intermediate-energy range to obtain the absolute EDCS is confirmed. It is important to state, however, that the use of this technique depends on the availability of the absolute EDCS of a secondary

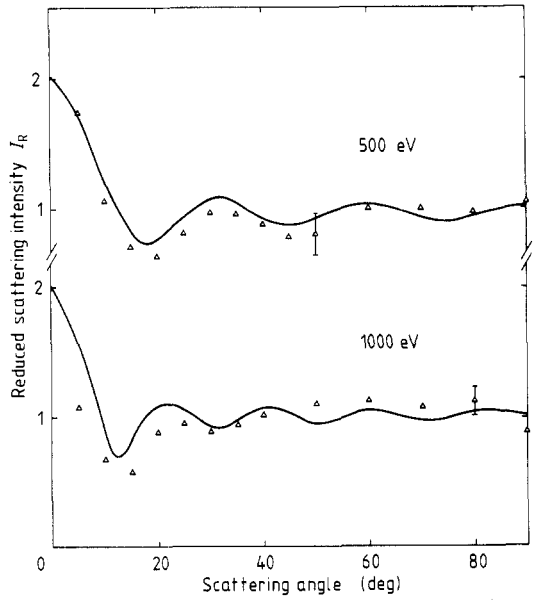

Figure 5. The reduced scattering intensities for the e^--O_2 interaction at 500 and 1000 eV: ——, IAM-IMS results; △, present experimental data.

standard and this fact effectively limits the use of this method to impact energies up to 1000 eV. This is because, to our knowledge, there are few results for the EDCS above 1000 eV reported in the literature, and all of them are limited to scattering angles below 50°.

Despite this restriction, it is important to obtain absolute EDCS for a variety of systems, particularly for polyatomic molecules, because of the recent theoretical interest. Some of these measurements are now in progress.

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