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Low-energy electron scattering from O_2

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Abstract. Absolute differential cross sections for elastic electron scattering from molecular oxygen have been measured at incident electron energies between 1 and 30 eV and over a range of scattering angles between 10 and 130°. These results are compared with other measurements and they highlight a substantial degree of discrepancy between the various experiments at energies below 10 eV. They also indicate some interesting behaviour at low energies and forward angles, which appears to be common to a number of other diatomic molecules. The data are also compared with a recent Schwinger variational calculation, and total elastic and elastic momentum transfer cross sections, derived from the differential measurements, are also presented and compared with a recent R-matrix calculation.

1. Introduction

Electron scattering from molecular oxygen represents an interesting and, given the level of agreement displayed in the literature, apparently difficult challenge for modern experiment and scattering theory; interesting because of its obvious importance in the earth's atmosphere and, from a theoretical point of view, difficult due to its open shell structure. However the number of elastic scattering studies on this target, including those using modern theoretical techniques, are relatively few and, as we have discovered during a survey of available results, suffer from substantial disagreement as to the shape and magnitude of the cross section.

Our initial interest in elastic scattering cross sections for O_2 arose from a proposed study on electron scattering from O_3. As ozone beams of high purity are difficult to produce and maintain, the principal impurity being O_2 which arises from collisional dissociation of the ozone, we required accurate elastic electron–O_2 cross sections in order to correctly account for the contribution from these dissociation product impurities. Examination of the available cross sections at energies below about 10 eV indicated, however, that there were substantial differences amongst the previously measured cross sections (e.g. by as much as 40–50% at 5 eV and a scattering angle of 40°). This work includes the early measurements of Linder and Schmidt (1971), Trajmar *et al* (1971), Dehmel *et al* (1976) and Shyn and Sharp (1982), and the more recent experiments of Doering (1992) and Middleton *et al* (1994). Whilst each of these investigations used different techniques to establish the absolute scale for their measured angular distributions, the discrepancies cannot be due to this alone as there are also substantial differences in the shape of the differential cross section (DCS). The angular distributions of both Linder and Schmidt and Trajmar *et al* were placed on an absolute scale by normalizing to the total cross sections of Sałop and Nakano (1970).

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For the low energy (< 2 eV) results of Linder and Schmidt, the total cross sections of Salop and Nakano had to be extrapolated. We note that the normalization of Trajmar *et al* also took into account contributions from electronic excitation. Dehmel *et al* also used a similar technique but with the total cross sections of both Sunshine *et al* (1967) and Salop and Nakano. Shyn and Sharp relied upon measurements of relative scattering intensity for O_2 and helium, with their scattering chamber flooded with each gas and the relative gas number densities determined from measurements with a calibrated ionization gauge. Doering's experiments were principally aimed at measurements of excitation cross sections for the a $^1\Delta_g$ state which were normalized against the elastic scattering intensity which was, in turn, normalized to the cross section of Trajmar *et al* (1971). Recently, Middleton *et al* (1994) have measured relative angular distributions between 5 and 20 eV, which they normalized to the measurements of Shyn and Sharp. There have been several measurements of total scattering cross sections for O_2, but they are not of direct relevance to this discussion. We also note the recent measurements of absolute backscattering cross sections for O_2 by Randell *et al* (1994), although their energy range (10–175 meV) is well below that of the present measurements. There have also been several measurements of elastic differential cross sections at higher energies (Trajmar *et al* 1972, Bromberg 1974, Wakiya 1978, Daimon *et al* 1982). A good summary and discussion of available cross sections for O_2 is given by Itikawa *et al* (1989) and Itikawa (1994).

Thus the present work is motivated by a need for accurate low-energy cross sections for the ozone studies and also a desire to investigate and hopefully unravel the situation regarding the previous measurements. This measurement also represents the first investigation of electron–oxygen scattering using the well-known relative flow technique. We also note that there are few theoretical calculations for differential elastic electron scattering from O_2, indeed to our knowledge the only available results are recent, as yet unpublished, calculations by Ribeiro *et al* (1994) using the Schwinger variational technique. Noble and Burke (1992) and Higgins *et al* (1994) have calculated the integral elastic cross section between threshold and 15 eV using the *R*-matrix approach and this shows good agreement with the integrated cross sections of Shyn and Sharp (1982) at energies of 5 eV and above. At lower energies (2–5 eV) the theory lies as much as 50% above experiment. Noble and Burke attribute this to the incomplete description of polarization effects in their model, which includes twelve scattering symmetries but only accounts for about 30% of the ground-state polarizability of O_2. The inclusion of polarized pseudostates is thought to be necessary to correctly describe these long-range polarization effects which are of primary importance for low-energy scattering from open shell systems (Noble 1994).

2. Experimental apparatus and techniques

The crossed electron–molecular beam apparatus used for the present measurements has been described in substantial detail in a number of recent publications (e.g. Gulley *et al* (1993) and references therein) and we will not repeat that detail here. The energy resolution for the present experiments was typically 60 meV, which is clearly sufficient to resolve the first vibrational excitation mode (E_{loss} = 0.192 eV) from the elastic channel. The absolute cross sections at each energy and scattering angle were determined by use of the relative flow technique (Srivastava *et al* 1975), with helium as the cross section standard. Relative flow rates of O_2 and helium, through the beam-forming multicapillary array, were measured as a function of capillary driving pressure in a separate series of experiments. The driving pressures for each gas, measured with a capacitance manometer, were adjusted such that their mean-free-paths in the region immediately prior to the capillary array were equivalent.

Based on the molecular diameters for He (2.55 Å, Reid *et al* (1987); 2.65 Å, Weast (1973)) and O_2 (3.47 Å, Reid *et al* (1987)), this required that $P(\mathrm{He}) \approx 1.7 \times P(\mathrm{O_2})$. In addition, the driving pressures were such that for each gas the mean-free-path was greater than twice the diameter of the capillary array tubes, a general protocol that we have established as a result of previous studies (Buckman *et al* 1993). This resulted in driving pressures $P(\mathrm{He}) \leqslant 1.3$ mbar and $P(\mathrm{O_2}) \leqslant 0.75$ mbar. As a check of the sensitivity of the measured cross sections to the ratio of gas driving pressures, we carried out a series of measurements of the cross sections at an energy of 7 eV and a scattering angle of 30° for driving pressure ratios ($P(\mathrm{He})/P(\mathrm{O_2})$) between 0.4 and 3.6. The only significant difference in these measurements came at pressure ratios less than 1.0. For all of the measurements reported here the driving pressure ratio did not vary from 1.7 by more than 5%. As in our other recent applications of the relative flow technique (e.g. Gulley and Buckman 1994), we have operated with both gases present in the spectrometer at all times in order to reduce contact potential variations and to increase the stability of the apparatus. This we feel is particularly useful when the target gas is a reactive species such as O_2.

The incident energy was calibrated by reference to the energy of the He^- $1s2s^2\,^2S$ resonance in elastic electron helium scattering at 19.367 eV (Brunt *et al* 1977) and to the position of the second quasi-vibrational $N_2^-\,^2\Pi_g$ resonance peak in elastic scattering from N_2 at a scattering angle of 60° and energy of 2.198 eV (Rohr 1977). The experiments were conducted by scanning the energy-loss voltage of the analyser across the elastic peak and collecting data in a multichannel analyser. Thus the measurements represent rotationally summed, vibrationally elastic cross sections. Data collection was performed entirely under computer control, with the computer scanning the energy loss and associated analyser voltages, varying the electron scattering angle, monitoring the electron beam current, via a retractable Faraday cup, and the gas driving pressures via an MKS Baratron.

3. Results and discussion

Absolute differential cross sections for elastic electron–molecular oxygen scattering are given in table 1 and are compared graphically at a number of chosen energies with other available experimental determinations in figures 1 and 2. The error estimates in both the table and the figures are a combination, in quadrature, of statistical uncertainties and systematic effects. The latter include uncertainties in the measured relative flow rates, variations in pressure and beam current and the uncertainty in the helium cross sections used for normalization. The overall uncertainties lie typically between 7–10%.

At 1.0 eV (figure 1(*a*)) the DCS is seen to increase with scattering angle between 20 and 130°. The present measurements show good agreement with regard to the shape of the DCS with those of Linder and Schmidt (1971), measured at 0.96 eV, although the present results are a little higher (≈ 20%) in absolute magnitude. Linder and Schmidt obtained their absolute scale by normalizing their integrated cross section to that of Salop and Nakano (1970), which in turn they had extrapolated to energies below 2 eV, the lowest measured energy of Salop and Nakano. Thus the difference in absolute magnitude is perhaps not too disconcerting. A similar situation exists for both 2.0 and 3.0 eV (figures 1(*b*) and (*c*)) where the shape of the present DCS is in good agreement with that of Linder and Schmidt but uniformly 10–30% larger in magnitude, the better agreement being obtained at 3 eV. The agreement with the results of Shyn and Sharp (1982) is not so good, particularly at large angles where the latter exhibits an increase in the cross section which is not evident in the present work or that of Linder and Schmidt. At 4.0 eV (figure 1(*d*)) the level of agreement with Linder and Schmidt's DCS is very good, the two data sets lying within error

Table 1. Absolute differential cross sections (10^{-16} cm^2 sr^{-1}) for elastic electron scattering from O_2. Figures in parentheses represent the percentage uncertainty. The integral cross sections Q_t and Q_m (10^{-16} cm^{-2}) have an estimated uncertainty of ±20%.

	Energy (eV)											
Angle	1.0	2.0	3.0	4.0	5.0	7.0	8.0	9.0	10.0	15	20	30
12	—	—	—	—	—	—	—	—	—	3.03 (8)	—	—
15	—	—	—	0.487 (7)	0.585 (7)	0.877 (7)	1.23 (9)	1.45 (7)	1.85 (7)	2.76 (7)	3.44 (8)	5.08 (8)
20	0.165 (11)	0.303 (9)	0.418 (7)	0.514 (7)	0.603 (7)	0.836 (7)	1.03 (9)	1.30 (7)	1.58 (7)	2.29 (7)	2.73 (8)	3.84 (8)
25	—	—	0.480 (7)	0.576 (7)	0.642 (7)	—	—	1.18 (8)	—	—	—	—
30	0.253 (9)	0.406 (7)	0.552 (7)	0.636 (7)	0.706 (7)	0.853 (7)	0.985 (9)	1.12 (7)	1.30 (7)	1.71 (7)	1.77 (7)	2.02 (7)
35	—	—	0.594 (7)	0.699 (7)	0.765 (7)	—	—	1.07 (8)	—	—	—	—
40	0.329 (8)	0.517 (7)	0.644 (7)	0.758 (7)	0.824 (7)	0.910 (7)	0.991(9)	1.06 (7)	1.15 (7)	1.38 (7)	1.30 (8)	1.11 (11)
45	—	—	0.699 (7)	0.806 (7)	0.858 (7)	—	—	1.03 (8)	—	—	—	—
50	0.398 (8)	0.604 (7)	0.728 (7)	0.838 (7)	0.886 (7)	0.931 (7)	0.959 (7)	0.988 (7)	1.04 (7)	1.11 (7)	0.958 (8)	0.654 (10)
55	—	—	0.752 (7)	0.854 (7)	0.899 (7)	0.912 (7)	—	0.942 (7)	—	—	—	—
60	0.459 (8)	0.650 (7)	0.770 (7)	0.856 (7)	0.914 (7)	0.881 (7)	0.920 (7)	0.872 (7)	0.894 (7)	0.844 (7)	0.639 (8)	0.420 (7)
65	—	—	0.760 (7)	0.828 (7)	—	—	—	—	—	—	—	—
70	0.505 (7)	0.667 (7)	0.743 (7)	0.809 (7)	0.815 (7)	0.780 (7)	0.780 (9)	0.720 (7)	0.714 (7)	0.599 (7)	0.431 (8)	0.270 (8)
75	—	—	0.709 (7)	0.749 (7)	—	—	—	—	—	—	—	—
80	0.518 (7)	0.651 (7)	0.685 (7)	0.698 (7)	0.677 (7)	0.620 (7)	0.594 (8)	0.541 (7)	0.554 (7)	0.423 (7)	0.282 (8)	0.184 (8)
85	—	—	0.651 (7)	0.651 (7)	—	—	—	—	—	—	—	—
90	0.540 (8)	0.625 (7)	0.605 (7)	0.589 (7)	0.565 (7)	0.500 (7)	0.461 (7)	0.430 (7)	0.453 (7)	0.333 (7)	0.218 (8)	0.137 (8)
95	—	—	0.569 (7)	0.537 (7)	—	—	—	—	—	—	—	—
100	0.562 (10)	0.583 (7)	0.536 (7)	0.489 (7)	0.464 (7)	0.401 (7)	0.390 (8)	0.379 (7)	0.390 (8)	0.298 (7)	0.218 (8)	0.130 (10)
105	—	—	0.503 (7)	0.470 (7)	—	—	—	—	—	—	—	—
110	0.567 (8)	0.544 (7)	0.470 (7)	0.426 (7)	0.391 (7)	0.359 (7)	0.359 (9)	0.369 (7)	0.397 (8)	0.304 (7)	0.248 (8)	0.166 (9)
115	—	—	0.448 (7)	0.392 (7)	—	—	—	—	—	—	—	—
120	0.556 (8)	0.509 (7)	0.424 (7)	0.372 (7)	0.343 (7)	0.335 (7)	0.360 (8)	0.375 (7)	0.410 (7)	0.347 (7)	0.303 (9)	0.257 (10)
125	—	—	0.406 (7)	0.346 (7)	—	—	—	—	—	—	—	—
130	0.561 (7)	0.468 (7)	0.386 (7)	0.342 (7)	0.323 (7)	0.338 (7)	0.383 (7)	0.391 (7)	0.431 (7)	0.399 (8)	0.392 (8)	0.425 (10)
Q_t	6.1	6.7	6.9	7.0	7.1	7.3	7.8	7.9	8.6	8.8	8.7	8.8
Q_m	6.7	6.5	6.1	6.0	5.7	5.7	6.2	5.9	6.4	5.9	6.1	6.0

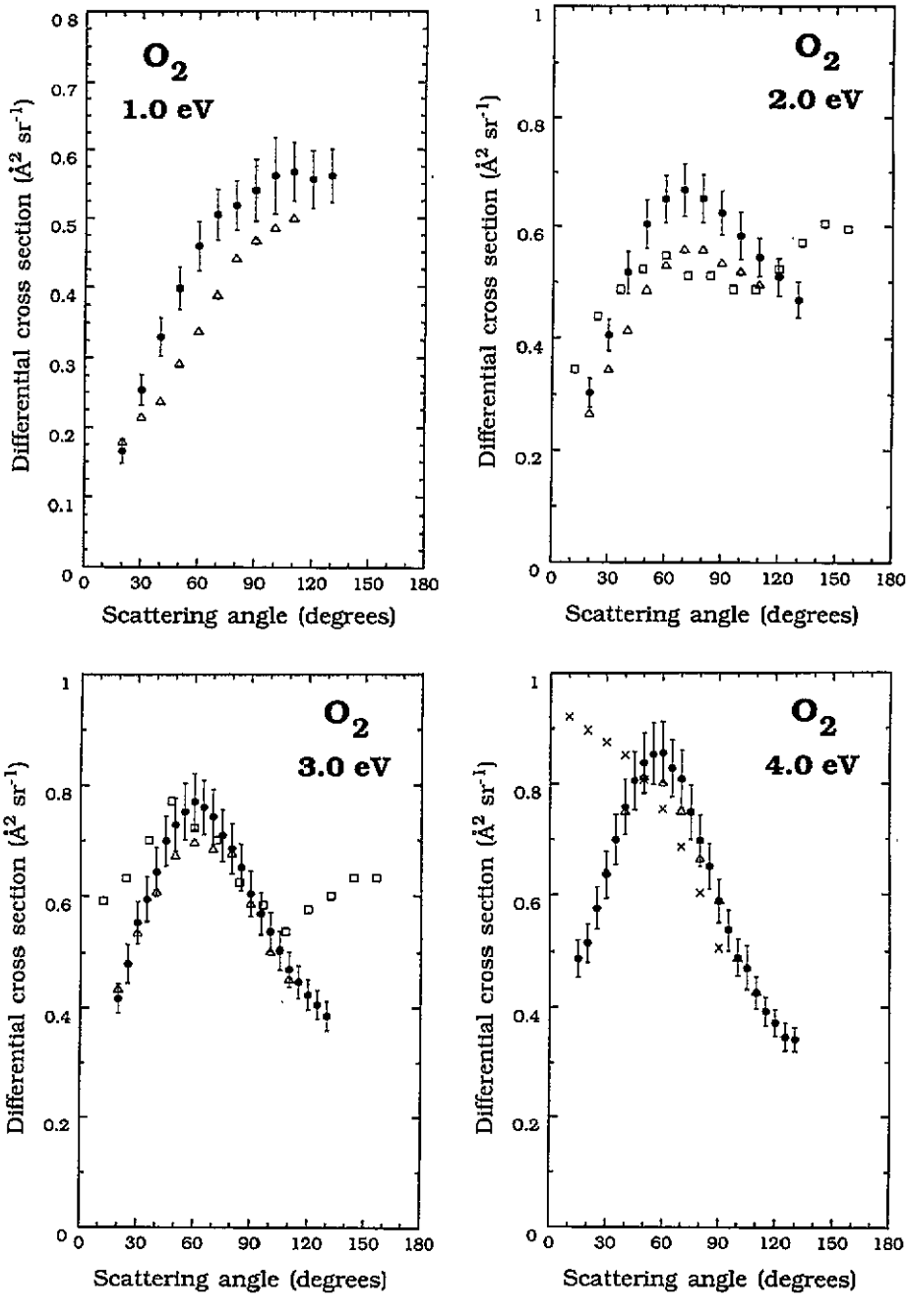

Figure 1. Absolute differential cross sections for elastic electron scattering from O_2 (in units of $Å^2\ sr^{-1}$) at (*a*) 1.0 eV, (*b*) 2.0 eV, (*c*) 3.0 eV and (*d*) 4.0 eV: (●) present results, (×) Trajmar *et al* (1971), (△) Linder and Schmidt (1971), (□) Shyn and Sharp (1982).

bars at all angles. This is also the lowest energy at which comparison can be made with the data of Trajmar *et al* (1971), and we find that for angles below about 60° the two cross sections exhibit entirely different shapes, such that at angles less than 20° the present cross

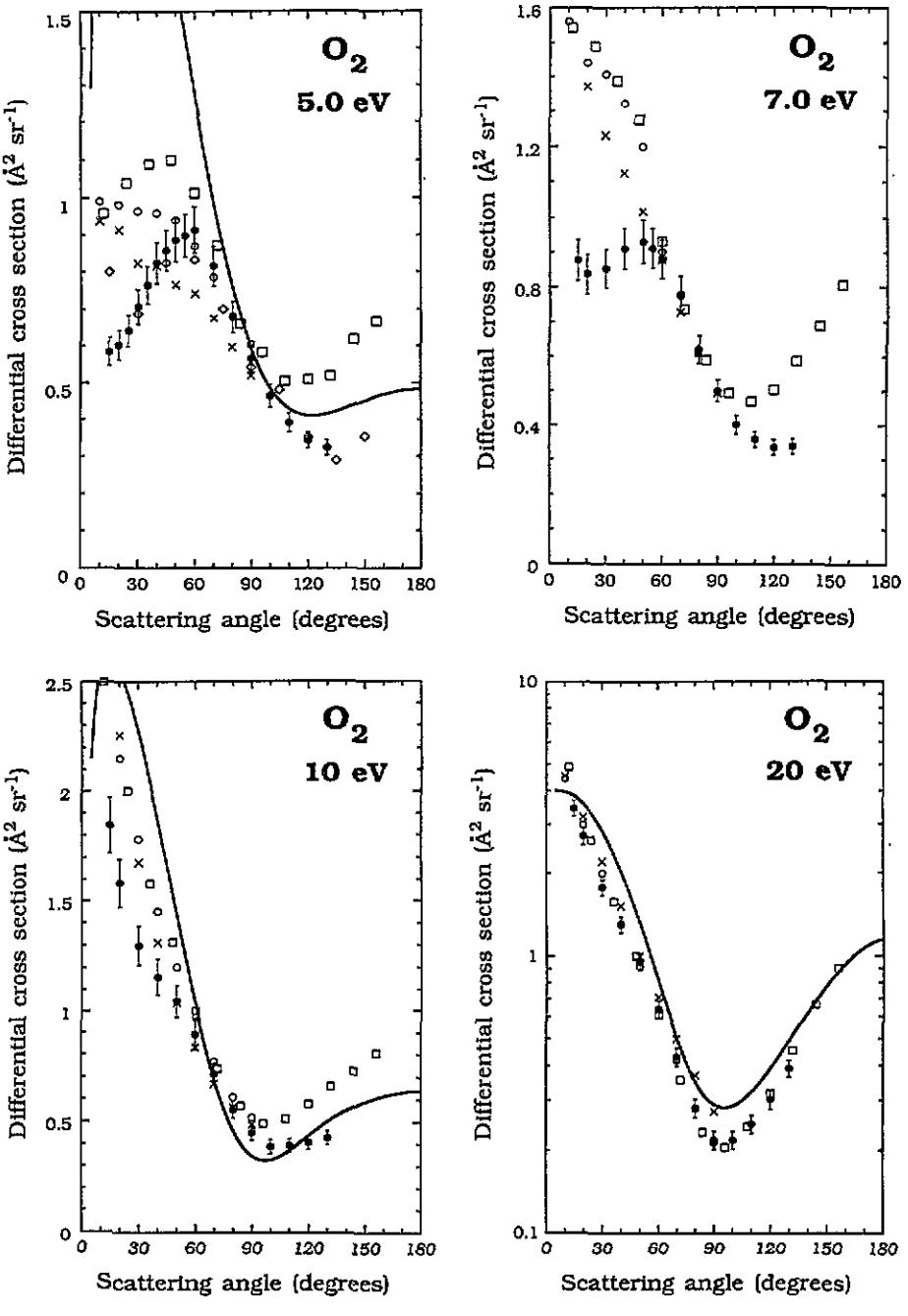

Figure 2. Absolute differential cross sections for elastic electron scattering from O_2 (in units of $Å^2$ sr^{-1}) at (*a*) 5.0 eV, (*b*) 7.0 eV, (*c*) 10.0 eV and (*d*) 20.0 eV: (●) present results, (×) Trajmar *et al* (1971), (□) Shyn and Sharp (1982), (○) Middleton *et al* (1994), (◇) Dehmel *et al* (1976), (——) Ribeiro *et al* (1994).

section is almost a factor of two smaller.

The comparison of all the available DCS data at 5.0 eV (figure 2(*a*)) gives a good

illustration of the discrepancies which provided the major motivation for the present investigations. All four existing data sets (Trajmar *et al* 1971, Dehmel *et al* 1976, Shyn and Sharp 1982, Middleton *et al* 1994) show different shapes and absolute magnitudes, and the only one which resembles the present measurements over the entire angular range is that of Dehmel *et al* (1976), which is in good accord with the present measurements in both shape and absolute magnitude, with the exception of a further rise in the cross section below 30° which is not observed in the present data. We also note, but for reasons of clarity do not show the data, that the relative elastic cross sections of Doering (1992) between 50–135° exhibit similar angular behaviour to that of Dehmel *et al*. In particular, these data predict the decrease of the cross section at scattering angles less than 60° which is observed in the present cross section. The data of Shyn and Sharp also exhibit similar forward angle behaviour, but for angles less than 60° there are substantial (~ 40%) differences in magnitude between their measurements and the present. Once again the data of Shyn and Sharp exhibit a rise in the DCS at angles above 100°. The present data, and certainly the data of Dehmel *et al*, also show this but at slightly higher scattering angles. The DCS of both Middleton *et al* (1994) and Trajmar *et al* (1971), despite showing reasonable agreement at angles between 60–100°, show a steady increase in the cross section at small angles. The Schwinger variational (SV) calculation of Ribeiro *et al* (1994) shows the same overall shape as the present data, including the decrease at forward scattering angles, although it greatly overestimates the cross section between 0 and 60°.

At 7.0 eV (figure 2(*b*)) similar trends are observed in the comparison with the DCS of Trajmar *et al*, Shyn and Sharp and Middleton *et al*, with the present experiment being the only one to indicate a decrease in the magnitude of the cross section at forward scattering angles. At 10 eV (figure 2(*c*)) this discrepancy has largely vanished with all the measured DCS showing the same overall angular behaviour, although the present cross section is still significantly smaller (20–30%) than the others at angles less than 60°, and is also smaller than that of Shyn and Sharp at backward angles. Once again the SV calculation overestimates the cross section at forward angles but is in reasonably good agreement with the present measurements at angles larger than about 60°. Finally, at 20 eV (figure 2(*d*)) there is excellent agreement between the present results and those of Trajmar *et al*, Shyn and Sharp, Doering (not shown) and Middleton *et al*, and good agreement between all of these measurements and the SV calculation. Cross sections at 8.0, 9.0, 15 and 30 eV are not presented graphically but are listed in table 1.

The angular behaviour of the DCS at energies below about 10 eV is rather interesting, in particular the tendency shown in the present measurements for the cross section to flatten out and finally decrease in the forward direction. This behaviour is not unique to O_2 but has been seen in elastic DCS measurements for a variety of diatomic and polyatomic molecules. The fact that this behaviour is not universally observed in all experiments is not of particular relevance to this discussion as it has been independently observed in enough measurements to remove most of the doubt concerning its existence. Unfortunately there are few theoretical calculations for O_2 in this energy range that can be used as a guide in understanding this behaviour, although it does appear that the calculation of Ribeiro *et al* (1994) shows a similar behaviour at 5 and 10 eV, albeit at a somewhat lower scattering angle. Such behaviour has also been observed in other scattering experiments and a few comments may be relevant. The other diatomic systems in which this feature of the DCS has been observed (N_2, NO and CO) have similar dipole polarizabilities to that of O_2, even though many other characteristics of their electronic structure are quite different. One can intuitively relate the forward angle behaviour of the DCS to long range interactions, which are largely mediated by the polarization potential, and it has been demonstrated in N_2 (Sun

et al 1995) that the correct qualitative description of the observed forward angle behaviour of the elastic DCS at energies between 4 and 10 eV requires the inclusion of an enormous number of partial waves in each scattering symmetry. Thus this effect at forward angles appears to be due to subtle interference effects between high angular momentum components of the scattering process. The apparently ubiquitous nature of this effect clearly warrants further detailed theoretical analysis of each of these molecular scattering systems.

The present elastic differential cross sections have been extrapolated to 0 and 180° and integrated to yield total elastic (Q_{el}) and elastic momentum transfer (Q_m) cross sections which can also be compared with values in the literature. In the absence of any theoretical

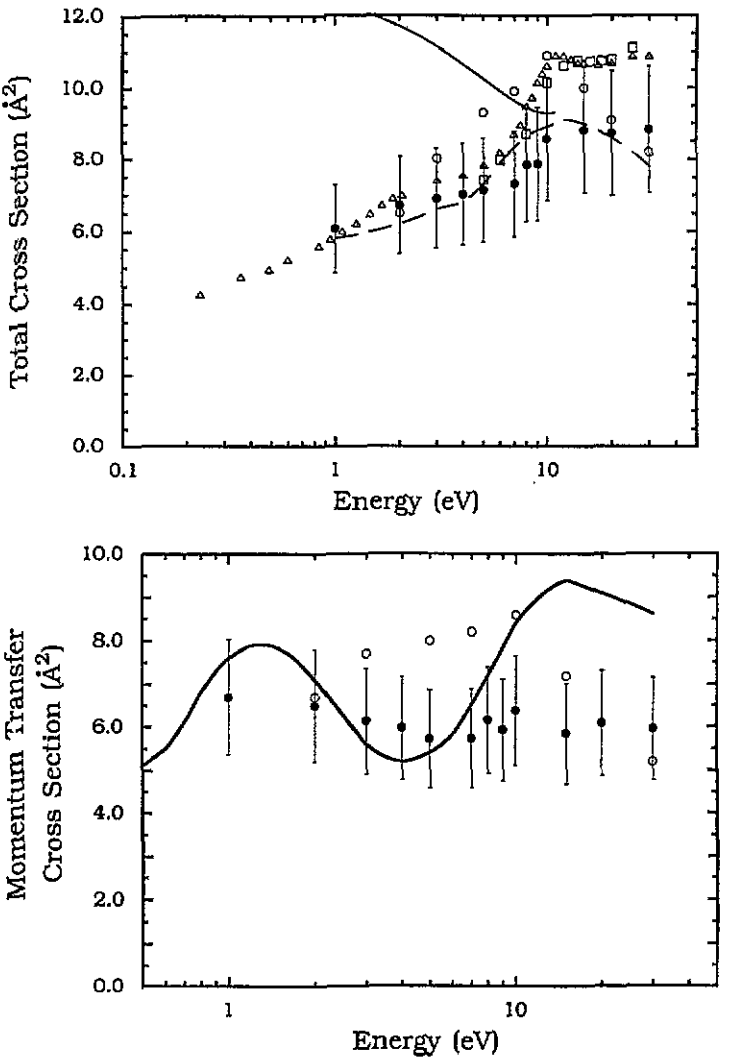

Figure 3. Integral cross sections for O_2 (in units of Å^2): (*a*) total cross section, (●) present total elastic results, (○) Shyn and Sharp (1982) total elastic, (△) Zecca *et al* (1986) grand total, (□) Kanik *et al* (1992) grand total, (– – –) Kanik *et al* (1992) 'recommended' total elastic, (——) Higgins *et al* (1994) total elastic; (*b*) elastic momentum transfer cross section, (●) present results, (○) Shyn and Sharp (1982), (——) Hake and Phelps (1967).

guidance the extrapolation was done by eye, and as a result the derived integral cross sections have an estimated uncertainty of ±20% and are listed in table 1. The total elastic cross sections are compared in figure 3(*a*) with a selection of the most recently measured grand total cross sections (Q_T), with the Q_{el} derived from the DCS measurements of Shyn and Sharp, with a 'recommended' Q_{el} of Kanik *et al* (1993) and with the Q_{el} resulting from the *R*-matrix calculation of Higgins *et al* (1994). In general the agreement is reasonably good, with the present cross section, which does not include contributions from inelastic channels, lying below the grand total cross section measurements of Zecca *et al* (1986) and Kanik *et al* (1992) as expected. It is also in good agreement with the recommended total elastic cross section of Kanik *et al* (1993), but lower than the total elastic cross section of Shyn and Sharp which is *larger* than the measured grand total cross section in the 3–10 eV region. The present results are also lower than the *R*-matrix values (Higgins *et al*) at all common energies. The present measurements appear to show a weak enhancement of the cross section in the 7–10 eV region, where a number of resonances have been observed to enhance vibrational and electronic excitation and dissociative attachment channels (e.g. Burrow 1973, Wong *et al* 1973, Noble and Burke 1992, Middleton *et al* 1994). The momentum transfer cross section is shown in figure 3(*b*) where it is compared with the cross section of Hake and Phelps (1967), which is derived from electron transport measurements, and with the cross section of Shyn and Sharp (1982). The cross section of Hake and Phelps exhibits a substantial amount of structure which is not apparent in the present cross section, the latter being essentially flat over the whole energy range. The present cross section is again lower in magnitude than that of Shyn and Sharp across most of the common energy range.

4. Conclusions

Absolute differential cross sections for elastic electron scattering by O_2 have been measured between 1.0 and 30 eV using the relative flow technique in conjunction with known electron–helium scattering cross sections. These data exhibit a mixed level of agreement with previous measurements, both in shape and absolute magnitude. They agree well with the measurements of Linder and Schmidt (1971), which only extend to 4 eV, and with those of Dehmel *et al* (1976) and Doering (1992) at those energies where comparison is possible between 5 and 20 eV. There are substantial discrepancies between the present data and those of Trajmar *et al* (1971), mainly at forward angles and energies between 4 and 10 eV where the present cross section decreases in the forward direction. Whilst there are some similarities in the overall shape of the present cross sections with those measured by Shyn and Sharp (1982), there are large discrepancies in absolute magnitude at both forward and backward angles for energies below 10 eV, and these are also reflected in the derived total elastic cross sections where the Q_{el} from Shyn and Sharp is larger than the grand total cross section at some energies. There are also substantial discrepancies between the present cross sections and those of Middleton *et al* (1994) at forward angles and energies between 5 and 10 eV. These data are relative measurements and were placed on an absolute scale by normalizing to the cross sections of Shyn and Sharp at middle angles, but this cannot account for the observed discrepancies. At energies above 10 eV there is good agreement with the data of Middleton *et al*. In comparison with the theoretical calculations of Ribeiro *et al* (1994) for the DCS, we see an improving level of agreement as the incident energy is increased from 5 to 20 eV. For the total elastic cross section the values of Higgins *et al* (1994) are uniformly higher than the present measurements at all energies.

Given the differences between experiments outlined above, in particular the behaviour

observed in the present and some earlier measurements at low energies and forward angles, it would clearly be advantageous to have some further theoretical insight to these problems.

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