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Single and double ionization of atomic oxygen by electron impact

W R Thompson, M B Shah and H B Gilbody
Department of Pure and Applied Physics, The Queen's University of Belfast, Belfast, UK

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Abstract. A pulsed crossed beam technique incorporating time-of-flight spectroscopy (previously developed in this laboratory) has been used to study the single and double ionization of ground-state oxygen atoms produced by an iridium tube furnace source. Relative cross sections σ_1 and σ_2 for single and double ionization have been determined in the respective energy ranges 14.1–2000 eV and 90–2000 eV which are wider than any previous measurements. These values have been normalized by reference to the absolute values of σ_1 previously measured by Brook *et al* at energies (50–997 eV) where the possible influence of an unknown admixture of long-lived excited atoms in their experiment should be small. However at energies below about 25 eV our values of σ_1 become much smaller than the values of Brook *et al*. Above 40 eV, the present ratios σ_2/σ_1 are in satisfactory agreement with other measurements which extend to 400 eV. Our values of σ_1 have been compared with selected theoretical predictions. Some evidence of structure in the energy dependence of σ_1 in the region of the cross section maximum is consistent with predicted contributions from inner-shell 2s ionization and from autoionization in addition to outer 2p electron removal.

1. Introduction

Accurate cross sections for the electron-impact ionization of atomic oxygen over a wide energy range are important for a detailed understanding of a number of inter-related collision processes in the earth's upper atmosphere where O atoms are a major constituent (see the review by Itikawa 1994). Application is found also in fusion energy research in the context of the modelling and diagnostics of impurities in magnetically confined plasmas (Janev and Drawin 1993).

There have been a number of previous measurements of cross sections for ionization of O atoms. In early measurements based on the modulated crossed beam technique, Fite and Brackmann (1959) and Rothe *et al* (1962) obtained cross sections σ_1 for single ionization of O atoms in the respective energy ranges 30–450 eV and 100–500 eV. These measurements employed a thermal energy partially dissociated oxygen beam from an rf discharge source. Cross sections σ_1 in O were inferred from measurements of the O^+/O_2^+ product ratio and previously measured cross sections by Tate and Smith (1932) for the ionization of O_2. The accuracy of these measurements based on the modulated crossed beam approach is limited by the poor signal/background ratios inherent in this approach. Uncertainties also arise from the possible influence of an unknown fraction of long-lived excited O atoms in the beam from the rf discharge source. The metastable $O(1s^22s^22p^4)^1S$ and ^1D levels are of greatest concern in this regard.

In subsequent work, Brook *et al* (1978) used a fast crossed beam technique to make absolute measurements of σ_1 in O at energies ranging from threshold to 997 eV. A small calibration correction of +2.4% in these values of σ_1 has since been identified by Montague *et al* (1984). The measurements of Brook *et al* (1978) employed a keV O atom beam prepared by charge transfer neutralization of O^+ in a gas target. The fast O^+ products of electron-impact ionization can then be separated by magnetic analysis and recorded with high efficiency. This fast crossed beam technique with an excellent product ion collection efficiency and a good signal/background ratio is inherently capable of high absolute accuracy. However, some uncertainties still remain since the fast O atom beams prepared by charge transfer could not be guaranteed to be free from metastable atoms. Careful studies by Brook *et al* (1978) of the near-threshold yields of O^+ could not fully resolve this issue.

Ziegler *et al* (1982) also used a fast intersecting beam approach to obtain cross sections σ_2 for the double ionization of O from near threshold to 400 eV. The ratios σ_2/σ_1 of the cross sections for double to single ionization were measured and then values of σ_2 were inferred by normalization to previously measured values of σ_1 including the results of Brook *et al* (1978).

Measurements by Zipf (1985) employed a crossed beam configuration using a thermal energy beam of partially dissociated oxygen derived from a high density atomic oxygen source which was claimed to provide mainly $O(1s^22s^22p^4)^3P$ ground-state atoms. The experiment provided ratios σ_2/σ_1 of the cross sections for double to single ionization in the energy range 40–300 eV which could then be normalized to the previously measured values of σ_1.

In the present work, we have used a pulsed crossed beam technique incorporating time-of-flight spectroscopy to study single ionization from near threshold to 2000 eV and double ionization from 90–2000 eV. The technique was originally developed in this laboratory (Shah *et al* 1987) to carry out accurate measurements of the electron-impact ionization of H atoms over a wide energy range. It has also been applied with advantage to study the ionization of He (Shah *et al* 1988) and other stable targets using thermal energy beams of ground-state atoms. Short duration pulses of electrons are passed through the target atom beam in a high vacuum region. Immediately after the transit of each electron pulse through the beam, the slow ionic products of ionization are swept out of the beam intersection region by a pulsed electric field and selectively identified (in the presence of background gas product ions) by their characteristic times of flight to a particle multiplier. In this technique the collection efficiency of the slow product ions is very high. In addition the ionization takes place in the absence of electric and magnetic fields (unlike most previous measurements) thereby avoiding some of the uncertainties present in other methods. In the present work with oxygen, an iridium tube furnace has been used to provide thermal energy beams of partially dissociated oxygen. The O atoms in these beams are entirely in the ground state and we therefore avoid the uncertainties of previous experiments where the possible influence of metastables was difficult to assess. However, unlike the fast crossed beam approach of Brook *et al* (1978), absolute calibration is difficult since thermal energy atom beam densities are not easy to determine accurately. For this reason our measurements of relative cross sections σ_1 and σ_2 have been normalized with respect to the values of σ_1 obtained by Brook *et al* (1978) at high energies where the possible influence of metastables in their measurements is believed to be very small.

2. Experimental approach

2.1. General description

The basic apparatus and measuring procedure was similar to that used in our previous studies of the electron-impact ionization of H (Shah *et al* 1987) except that an iridium tube furnace rather than a tungsten tube furnace was used to provide the dissociated oxygen beams in the present work. Since a detailed description has been given previously, only the main features of the present apparatus need to be summarized here.

An electron gun was triggered by a pulse generator to provide electron pulses of 100 ns duration at 10^5 pulses s^{-1}. The electrons intersected (at right angles) the thermal energy beam of dissociated oxygen from the iridium tube furnace which was located in a differentially pumped region. The crossed beam intersection region was maintained at a base pressure of about 2×10^{-7} Torr.

Slow ions resulting from ionizing collisions were swept out of the crossed beam region by a pulsed electric field applied immediately after the transit of each pulse of electrons through the oxygen beam. The extraction field was applied between a pair of high-transparency grids located on either side of the crossed beam region. The extracted ions were accelerated through a potential difference (PD) of about 4 kV and then recorded as individual counts by a particle multiplier. The extracted O^+ and O^{2+} ions were identified and distinguished from O_2^+ and various background gas ions by their different times of flight to the multiplier in accordance with their charge to mass ratios. As in our previous work, great care was taken to ensure a high and equal extraction and detection efficiency irrespective of the primary-electron energy. The extraction pulse generator was triggered by the main pulse generator (which operated the electron gun) via a variable delay which could be adjusted according to the transit time of the trailing edge of the electron pulse through the oxygen beam.

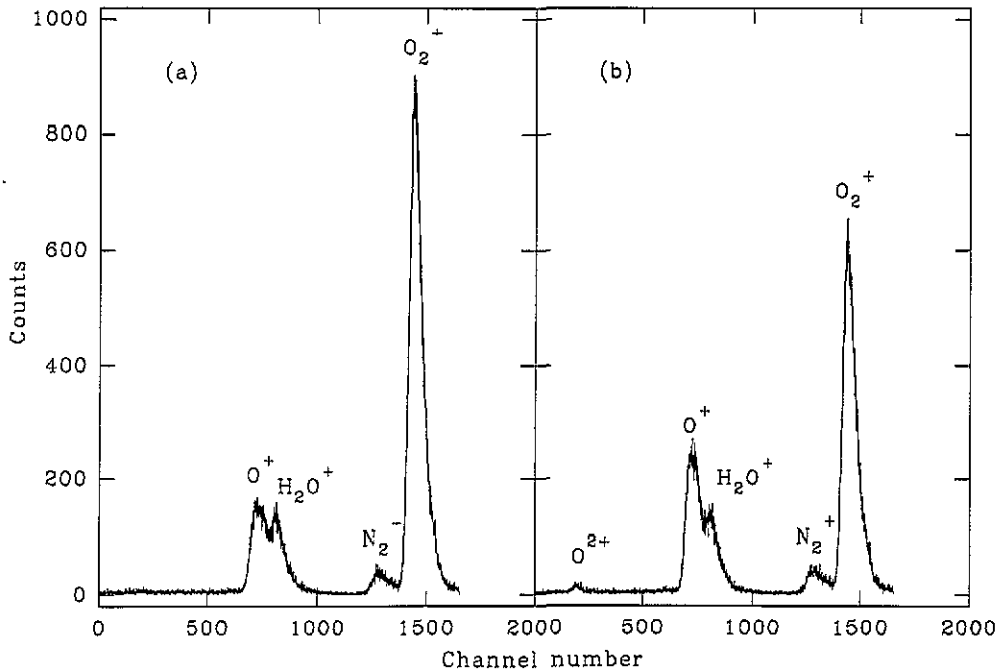

Figure 1. Time-of-flight spectra obtained for the ionization of oxygen by 147 eV electrons using an oxygen beam from an iridium furnace (a) at 1400 K when the oxygen was entirely molecular and (b) at 2300 K when the oxygen was partially dissociated.

A time-to-amplitude converter operated with start pulses from the extraction pulse generator and with stop pulses from the particle multiplier provided time-of-flight spectra on a multichannel analyser. Figure 1 shows the relevant portions of typical time of flight spectra obtained with the iridium furnace at low temperature when the beam was entirely O_2 and at 2300 K when the beam was partially dissociated. Peaks corresponding to O^+, O^{2+} and O_2^+ production can be discerned together with peaks corresponding to background gas ions.

The mounting of the iridium tube furnace was somewhat different from the arrangement described previously (Shah *et al* 1987). The tube (fabricated by rolling and seam welding 0.25 mm thick iridium foil) was 50 mm long and 6 mm inside diameter. It was clamped between two water-cooled copper terminals and heated by passing an alternating current of up to 260 A through it. Oxygen gas was introduced into one end of the tube at a constant rate while the dissociated oxygen beam emerged through a 2 mm diameter aperture at the midpoint of the hot central region. The temperature was determined by an optical pyrometer. The dissociation fraction of the oxygen depended upon the pressure of gas within the tube. Although dissociation fractions of up to about 0.90 could be obtained at low gas flow rates, in order to obtain acceptable signal/background ratios, higher flow rates were necessary which resulted in substantially reduced dissociation fractions. Great care was taken to screen (by means of steel and mu-metal shielding) the crossed beam region from the stray magnetic field associated with the furnace heating current.

The seven-element electron gun (Shah *et al* 1987) utilized a V-shaped thoriated tungsten filament. It provided pulsed beams of electrons equivalent to about 20 nA in the continuous mode with very small angular divergence. The electron beam intensity was recorded by means of a screened Faraday cup. The final four elements in the gun assembly were used to adjust the beam energy within the desired range up to 2 keV while the intensity remained essentially unchanged. In the interaction region, the electron beam had a diameter of less than 2 mm while the diameter of the oxygen beam through which it passed was 4 mm.

As in our previous work the energy of the electron beam could be expressed as $E = V_f - d$ where V_f is the acceleration voltage applied to the mid-point of the V-shaped filament of the gun and d is a correction parameter which allows for filament misalignment and the effect of contact potentials. A value of $d = 0.3 \pm 0.1$ V was obtained by linear extrapolation of the measured O^+ ion yield at low impact energies to the first ionization threshold of 13.61 eV.

2.2. *Cross section measurements and normalization*

In the analysis of the time-of-flight spectra of the type shown in figure 1, it was necessary to determine the O^+ and O^{2+} yields arising from single and double ionization of O atoms (while making accurate allowance for the presence of background gas contributions) and from the dissociative ionization of O_2 molecules. Apart from the undissociated O_2 molecules in the beam, a small increase in the background gas pressure, which necessarily took place when gas was fed into the furnace, led to an extra contribution from O_2 molecules. However, it should be noted that the velocity of formation of the O^+ products of dissociative ionization is such that some of them with initial directions other than towards the multiplier will escape from the extraction region before the extraction field pulse is applied. The contribution of O^{2+} ions from dissociative ionization is believed to be negligible (Massey 1969). Accurate allowance for the background and dissociative ionization contributions to the measured signals was made in the manner which has been described in detail in our H^+–H studies of ionization (Shah and Gilbody 1981).

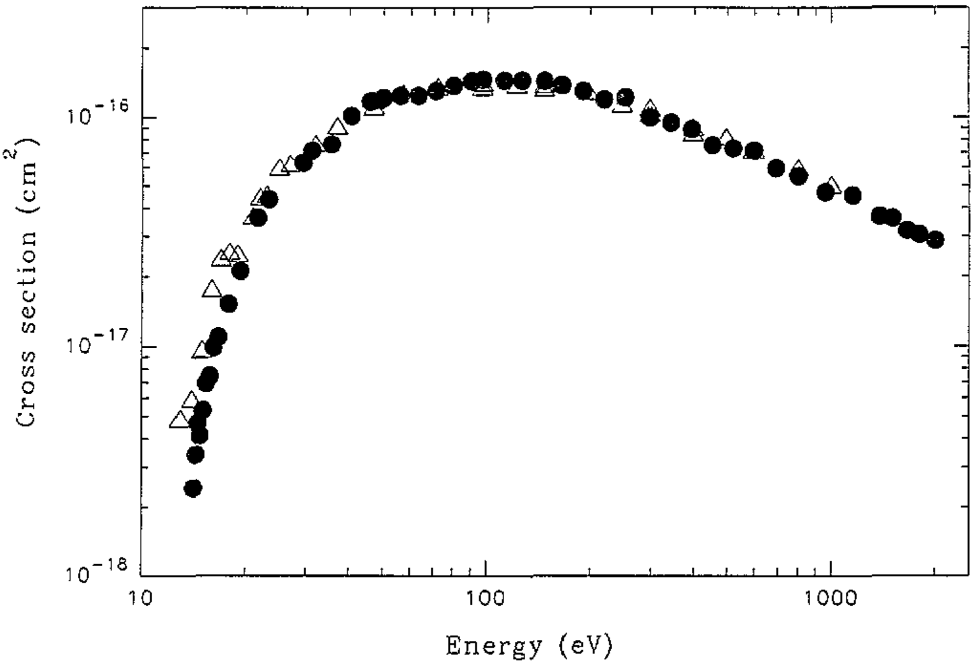

Figure 2. Present relative cross sections σ_1 (shown ●) for single ionization of O shown fitted to the 50–997 eV absolute values of Brook *et al* (1978) (shown △) where the possible influence of an admixture of long-lived excited atoms is believed to be small. The values of Brook *et al* (1978) shown have been increased by the 2.4% recommended by Montague *et al* (1984).

Cross sections σ_n for the n times ionization of O atoms (where $n = 1$ or 2) were obtained from the expression

$$\sigma_n = S_n / k\mu \tag{1}$$

where S_n is the O^{n+} yield per unit electron beam intensity and μ is the effective thickness of the O atom beam, a quantity related to the background pressure recorded in the crossed beam vacuum chamber. The constant k is the overall detection efficiency of the oxygen ions. As stated earlier, accurate determination of μ in the present thermal energy oxygen beam experiment is a difficult problem. We have therefore chosen to effectively determine the product $k\mu$ in equation (1) by normalization of our relative cross sections σ_1 to the absolute values obtained by Brook *et al* (1978) (including the +2.4% correction advocated by Montague *et al* (1984)) at high electron-impact energies where any influence of a possible admixture of metastable O atoms in their measurements is likely to be negligible. Figure 2 shows our relative cross sections σ_1 which extend from near threshold to 2000 eV fitted to the highest energy (50–1000 eV) values of Brook *et al* (1978). Although the fit can be seen to be very satisfactory at energies above about 40 eV, our values of σ_1 fall substantially below those of Brook *et al* (1978) at lower impact energies.

In support of our normalization procedure, it is important to note that our previous measurements of the corresponding cross sections σ_1 and σ_2 for electron-impact ionization of helium (Shah *et al* 1988) using our pulsed crossed beam technique were normalized by reference to known equivelocity proton impact cross sectons. Values of σ_1 for helium obtained in this way were in excellent agreement with the absolute values of σ_1 obtained by

Montague *et al* (1984) using the fast crossed beam approach even down to near threshold energies. Indeed, in measurements with helium in the present apparatus we have again demonstrated this good agreement in figure 3 where our relative values of σ_1 are shown fitted to previous low energy absolute values of σ_1 obtained by Montague *et al* (1984).

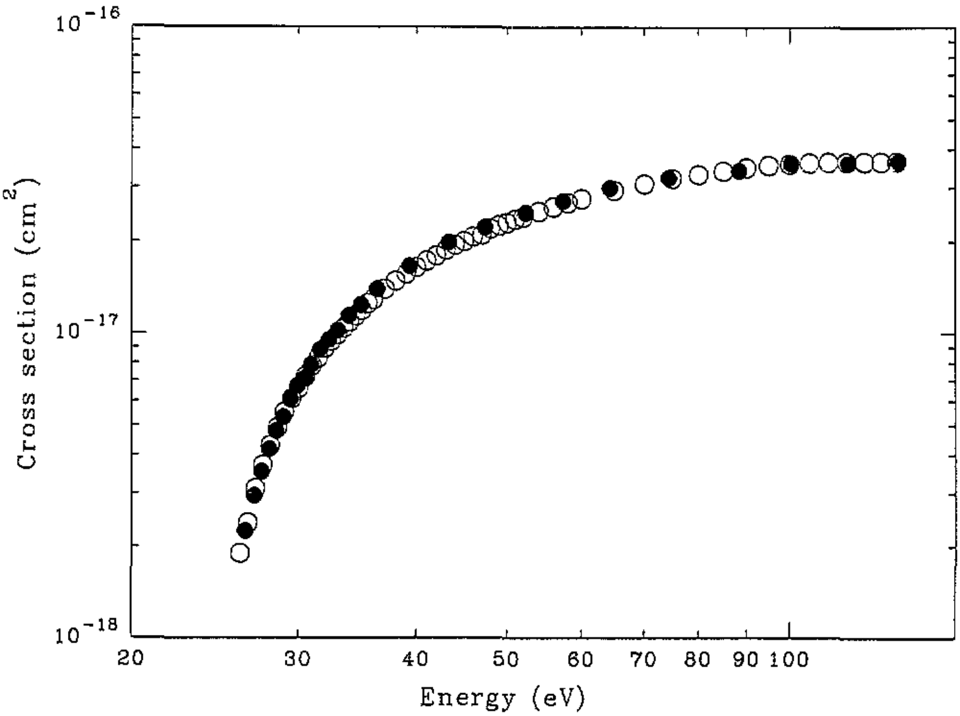

Figure 3. Relative cross sections (shown ●) for single ionization of helium obtained with the present apparatus fitted to the corresponding absolute cross sections (shown ○) measured by Montague *et al* (1984) using the fast intersecting beam technique.

3. Results and discussion

Table 1 shows our measured values of σ_1 and σ_2 in the respective energy ranges 14.1–2000 eV and 90–2000 eV. Uncertainties associated with individual cross sections are assessed at the 67% confidence level and reflect the degree of reproducibility of the values in terms of the various experimental parameters and statistical fluctuations. Additional estimated uncertainties of ±7% in the values of σ_1 and σ_2 respectively are associated with our normalization procedure.

In figure 4 we show the present cross sections σ_1 and σ_2 compared with the more recent previous experimental data. As already noted, although the present values of σ_1 are normalized to the higher energy values of Brook *et al* (1978) as corrected by Montague *et al* (1984), there are serious discrepancies between the two sets of data at energies below about 25 eV. For example at energies of 14 eV and 22 eV our values of σ_1 are about 42% and 80% respectively of the values measured by Brook *et al* (1978). It is possible that part of this large discrepancy between the two sets of data near threshold, might be explained by

Table 1. Cross sections σ_1 and σ_2 for single and double ionization of O atoms by electron impact. Random uncertainties associated with each cross section are shown. In addition all cross sections are subject to an estimated uncertainty of ±7% arising from the normalization procedure.

Energy (eV)	σ_1 (10^{-17} cm^2)	Energy (eV)	σ_1 (10^{-17} cm^2)	σ_2 (10^{-18} cm^2)
—	—	90.0	14.4 ± 0.4	1.89 ± 0.55
14.1	0.24 ± 0.01	97.0	14.6 ± 0.4	2.34 ± 0.63
14.3	0.34 ± 0.01	112.0	14.5 ± 0.4	3.66 ± 0.59
14.5	0.47 ± 0.02	126.0	14.5 ± 0.4	4.62 ± 0.67
14.7	0.41 ± 0.02	147.0	14.3 ± 0.4	4.63 ± 0.61
15.0	0.53 ± 0.02	165.0	13.7 ± 0.4	5.01 ± 0.55
15.3	0.69 ± 0.03	190.0	13.0 ± 0.4	5.10 ± 0.59
15.7	0.75 ± 0.03	219.0	11.9 ± 0.3	4.78 ± 0.55
16.1	0.99 ± 0.04	252.0	12.2 ± 0.3	4.87 ± 0.55
16.6	1.10 ± 0.05	297.0	9.9 ± 0.3	3.83 ± 0.60
17.8	1.52 ± 0.06	342.0	9.5 ± 0.3	3.51 ± 0.57
19.2	2.11 ± 0.08	393.0	8.8 ± 0.3	3.04 ± 0.51
21.6	3.61 ± 0.12	452.0	7.5 ± 0.2	2.48 ± 0.41
23.2	4.36 ± 0.16	520.0	7.3 ± 0.2	2.12 ± 0.47
29.2	6.27 ± 0.21	597.0	7.1 ± 0.2	2.38 ± 0.47
31.2	7.12 ± 0.24	687.0	5.93 ± 0.19	1.77 ± 0.40
35.5	7.61 ± 0.26	797.0	5.48 ± 0.18	1.66 ± 0.36
40.5	10.1 ± 0.3	957.0	4.66 ± 0.17	1.28 ± 0.33
46.0	11.8 ± 0.3	1148.0	4.52 ± 0.16	1.12 ± 0.31
50.0	12.2 ± 0.4	1378.0	3.67 ± 0.14	0.97 ± 0.29
56.0	12.4 ± 0.4	1540.0	3.59 ± 0.12	—
63.0	12.5 ± 0.4	1654.0	3.16 ± 0.12	0.76 ± 0.26
71.0	13.0 ± 0.4	1800.0	3.04 ± 0.11	—
80.0	13.1 ± 0.4	2000.0	2.87 ± 0.10	0.68 ± 0.24

the presence of unknown fractions of ^{1}S and ^{1}D metastable O atoms in the beams employed by Brook *et al* (1978). They considered the possible influence of metastable atoms on their measurements in terms of both outer- and inner-electron removal from these states and carried out careful studies of O^+ formation at energies below and near the ground-state ionization threshold. While these measurements are not conclusive, they do suggest that the influence of metastables on their measurements is not large enough to be the single cause of the large near threshold discrepancy between the two sets of data.

The measurements of Zeigler *et al* (1982) and Zipf (1985) provide ratios σ_2/σ_1 of the double to single ionization cross sections in the respective ranges 45–400 eV and 40–300 eV which are in satisfactory accord (within the combined experimental uncertainties) with the present data. The values of σ_1 and σ_2 due to Zipf (1985) in figure 4 were obtained by normalization to previous values of σ_1 including the values of Brook *et al* (1978). These values can be seen to exhibit an energy dependence which is in excellent agreement with the present values. The values of σ_2 in the range 40–400 eV obtained by Zeigler *et al* (1982) by normalization of measured ratios σ_2/σ_1 to the σ_1 values of Brook *et al* (1978) extend to lower energies than our data but in the energy range of overlap, the agreement between the two sets of data is satisfactory and within the combined uncertainties.

Cross sections σ_1 for single ionization can be seen to peak at about 100 eV while cross sections σ_2 for double ionization peak at about 180 eV where the ratio σ_2/σ_1 is only about 0.04. At 2000 eV, the highest impact energy considered, the ratio σ_2/σ_1 is only about 0.02.

There have been a number of calculations of the cross section σ_1 for the ionization

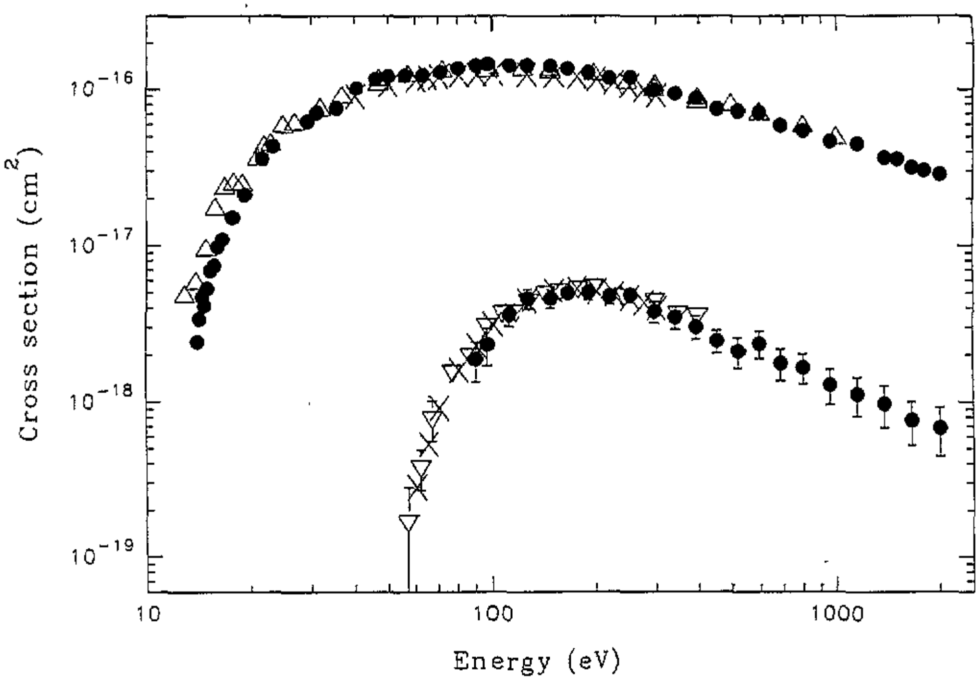

Figure 4. Cross sections σ_1 and σ_2 for single and double ionization of oxygen by electrons. ●, σ_1 and σ_2, present values; △, σ_1, Brook *et al* (1978); ×, σ_1 and σ_2, Zipf (1985); ▽, σ_2, Zeigler *et al* (1982).

of ground-state O atoms based on different approximations; these have been discussed in detail in the recent paper by Chung *et al* (1993). Some of these theoretical predictions have been selected for comparison with our values of σ_1 in figure 5. The calculations by Chung *et al* (1993) of cross sections for ionization of $(2s^22p^4)^3P$ ground-state atoms to form O^+ ions in the $(2p^3)^4S$, ^2D and ^2P states have been carried out by using the Born–Ochkur (BO) approximation and by using the exchange-distorted-wave method with Ochkur's approximation (EDWO) for energies up to 300 eV. They have also carried out similar calculations with no electron exchange based on the Born (B) and distorted-wave (DW) approximations. Previous calculations based on these same approximations by a number of different authors provide slightly different results for the reasons discussed by Chung *et al* (1993). The Born–Ochkur approximation has also been used by Chung *et al* (1993) to calculate cross sections for the inner-shell ionization processes leading to $O^+(2s2p^4)^4P$, ^2P formation. These cross sections, which amount to as much as about 10% of the total ionization, are much smaller than the earlier values calculated by Peach (1970, 1971) who used hydrogenic Coulomb functions rather than the Hartree–Fock continuum state functions used by Chung *et al* (1993). Any comparison of theory with experiment must also take account of autoionization contributions. In particular, the observations of Dehmer *et al* (1977) indicate that autoionization via the $O[2s^22p^3(^2P)3s''^3P]$ and $O[2s2p^5(^3P^0)]$ states should make a significant contribution to the measured ionization cross sections. These contributions have been estimated by Chung *et al* (1993) and amount to as much as about 15% of the total ionization. Chung *et al* (1993) have also noted that, at energies above 37 eV, ionization may take place through channels in which the O^+ ion is initially formed in excited states such as $2p^23s$. However, these contributions were considered to be relatively

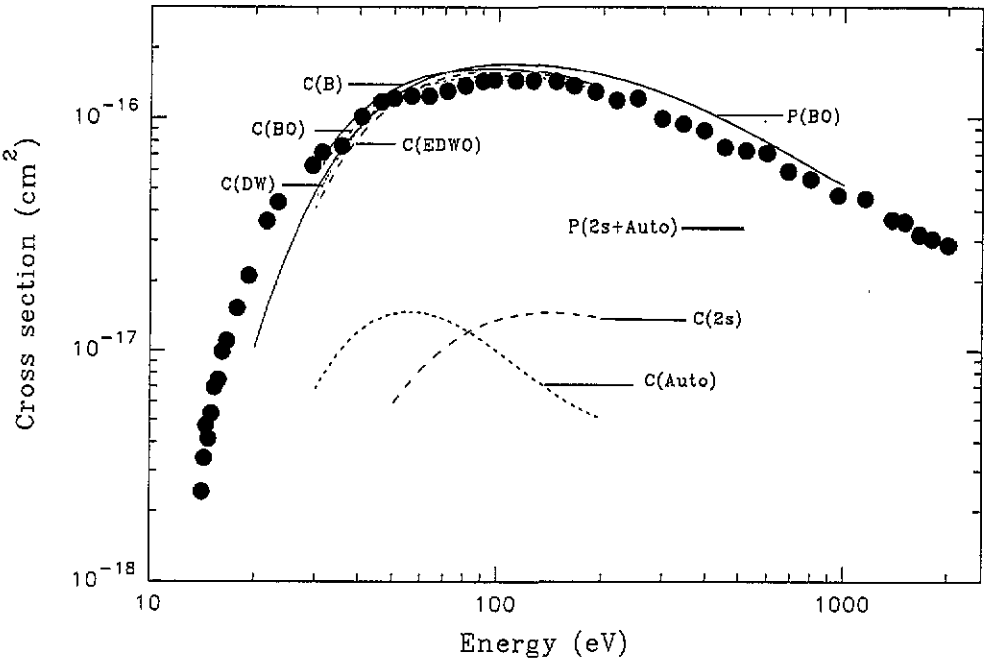

Figure 5. Present cross sections σ_1 (shown ●) for single ionization of O atoms compared with a number of theoretical predictions. Curves C, Chung *et al* (1993); C(2s), 2s inner-shell ionization only; C(Auto), estimated autoionization contribution (see text); C(B), 2p electron removal (Born without exchange)+C(2s)+C(Auto); C(DW), 2p electron removal (distorted waves without exchange)+C(2s)+C(Auto); C(EDWO), 2p electron removal (exchange distorted waves with Ochkur approximation)+C(2s)+C(Auto); C(BO), 2p electron removal (Born–Ochkur approximation)+C(2s)+C(Auto); Curves P, Peach (1970, 1971); P(2s+Auto), 2s inner-shell plus autoionization only; P(BO), 2p electron–electron removal (Born–Ochkur approximation)+P(2s+Auto).

small and therefore not included in their calculations.

In figure 5, while the separate contributions calculated by Chung *et al* (1993) for 2s inner-shell ionization and autoionization are shown, these have also been added to their estimates of the outer 2p electron ionization cross section based on the BO, EDWO, B and DW approximations for direct comparison with our experimental data in the range 30–200 eV. In the region between about 40 eV and 200 eV, their predicted cross sections are all in good general accord with experiment with the BO calculations providing the best agreement. It is also interesting to note, that in the range 30–200 eV, our experimental data points show some evidence of structure which is consistent with the energy dependence of the contributions predicted by Chung *et al* (1993) from both 2s inner-shell ionization and autoionization. The earlier calculations of Peach (1970, 1971) for 2p electron removal based on the Born–Ochkur approximation plus estimates of 2s ionization and autoionization can be seen to provide cross sections which exceed the experimental values by a factor of up to 1.3 at energies between 50 and 1000 eV. Below 30 eV these cross sections fall increasingly below our measured values.

4. Conclusions

Single and double ionization of ground-state O atoms by electron impact has been studied using a pulsed crossed beam technique which has been previously used very successfully in this laboratory for accurate measurements on both stable and unstable target species. The results of some previous experimental studies of atomic oxygen may be influenced by unknown admixture of long-lived excited O atoms in the beams employed. Relative cross sections σ_1 and σ_2 for single and double ionization obtained over wider energy ranges than considered in previous work have been normalized by reference to the highest energy (between 50 eV and 997 eV) absolute values of σ_1 obtained by Brook *et al* (1978) using a fast intersecting beam approach. At these relatively high energies, the possible influence of any admixture of long-lived excited atoms on the measurements of Brook *et al* (1978) is considered to be negligible. Our values of σ_1 when normalized in this way, fall increasingly below the values of Brook *et al* (1978) below about 25 eV. These large discrepancies, which are up to a factor of 2.5 near threshold, seem unlikely to be explainable solely in terms of the possible influence of long-lived excited atoms. Our measured cross section ratios σ_2/σ_1 are in satisfactory accord with the corresponding ratios measured by Zeigler *et al* (1982) and by Zipf (1985) in the respective energy ranges 45–400 eV and 40–300 eV.

Our values of σ_1 have been compared with selected theoretical predictions based on several different approximations. In particular, the calculations of Chung *et al* (1993) based on the Born–Ochkur approximation are in good agreement with experiment in the range 40–200 eV when, in addition to outer 2p electron removal, inner 2s shell ionization and autoionization contributions are included. Our measured values of σ_1 in the region of the cross section maximum exhibit some evidence of structure which is qualitatively consistent with the predicted additional 2s inner-shell ionization and autoionization contributions.

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