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DOUBLE IONIZATION OF ATOMIC OXYGEN BY ELECTRON IMPACT

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Abstract—Laboratory measurements of the cross-sections for double ionization of atomic oxygen by electrons are presented for energies from threshold to ~400 eV. A maximum cross-section of about 5.6×10^{-18} cm^2 is observed at an electron energy of approx. 200 eV. Absolute cross-sections are obtained from measurements of the ratio of the cross-sections for double and single ionization, coupled with absolute cross-sections for single ionization previously measured by other investigators. The possible effects of excited oxygen reactants are examined, and the reported cross-sections are considered to be characteristic of ground state oxygen atoms.

1. INTRODUCTION

In 1966 small concentrations of doubly ionized oxygen were recorded in the Earth's upper ionosphere by the Explorer 31 satellite (Hoffman, 1967). More recently O^{2+} concentrations of ~0.3 particles cm^{-3} have been measured by the GEOS 1 satellite in the Earth's plasmasphere (Young *et al.*, 1977; Geiss *et al.*, 1978). At times the GEOS data show local O^{2+} concentrations 30% as large as those of O^+. Voyager data indicate that O^{2+} concentrations (100–500 cm^{-3}) may actually exceed the O^+ concentrations in the warm, outer portion of the Io torus around Jupiter (Bagenal and Sullivan, 1981; Shemansky and Smith, 1981). Cross-sections for O^{2+} production are required for modeling these regions, and the present experiment is the first quantitative determination of cross-sections for double ionization of oxygen atoms by electron impact.

2. APPARATUS

The apparatus is shown schematically in Fig. 1 and, with the exception of the electron gun, is described in detail by Rundel *et al.* (1979). An ion beam extracted from a magnetically confined electron arc ion source containing O_2 is accelerated to 1500 eV. After momentum analysis the O^+ beam is partially neutralized by charge transfer with N_2. The resulting neutral beam contains both excited and ground state atoms. Highly excited oxygen atoms with principal quantum number $n \geq 20$ are ionized by a 7800 V cm^{-1} field (cf. Damburg and Kolosov, 1979) which deflects the resulting ions out of the beam, together with ions that were not neutralized in the charge transfer cell. Typical neutral fluxes in the interaction chamber are on the order 10^{10} atoms s^{-1}.

The electron gun, shown schematically in Fig. 2, contains an oxide coated cathode from a 6L6

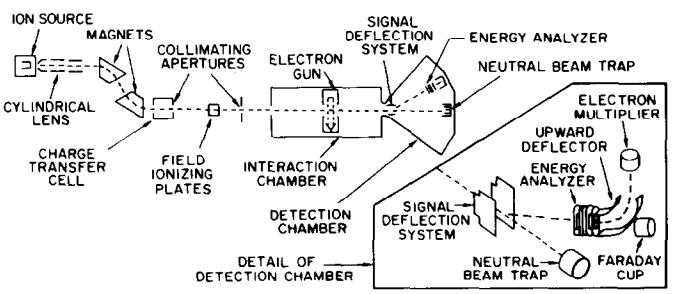

FIG. 1. SCHEMATIC DIAGRAM OF APPARATUS.

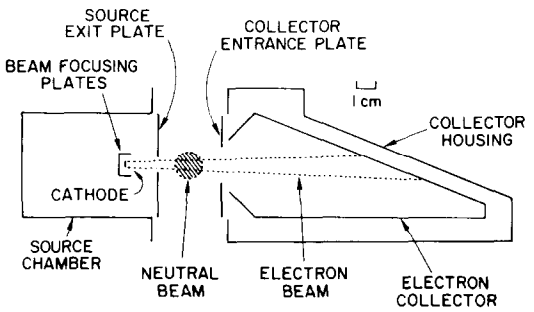

FIG. 2. SCHEMATIC DIAGRAM OF ELECTRON GUN.

electron tube mounted between a set of beam focusing plates. During data acquisition the cathode is biased negatively with respect to ground so that electrons accelerate toward the grounded source exit plate. The beam focusing plates are biased more negatively (usually by about 30 V) to focus the beam through the apertures in the source exit plate and collector entrance plate and then into the electron collector. Normally all the other structures in the gun are at ground potential during data acquisition. Typically 93% of the current leaving the cathode is collected in the electron collector, and to within experimental uncertainty the remaining current goes approximately in equal parts to the source exit plate and collector entrance plate. Electron currents used in these measurements range from 0.15 to 1.5 mA.

At the intersection of the electron and neutral beams product ions are formed by the reactions

$$e + O \rightarrow O^+ + 2e \quad (1)$$

$$e + O \rightarrow O^{2+} + 3e. \quad (2)$$

Since the momentum of the oxygen atoms is large compared to that of the electrons, the product ions are not appreciably scattered and they thus continue along the neutral beam axis. Immediately following entry into the detection region either the O^+ or O^{2+} ions are electrostatically deflected into a five grid retarding potential energy analyzer. Ions transmitted through the energy analyzer are then detected by a Johnston Laboratories MM1 electron multiplier (see Fig. 1). Typical count rates are ~ 50 cps for O^+ and ~ 2 cps for O^{2+}.

The primary purpose of the retarding potential energy analyzer is to reject ions formed in stripping collisions with the background gas. Even though the background gas pressure in the interaction and detection chambers is about 3×10^{-9} Torr the process

$$O + X \rightarrow O^+ + X + e \quad (3)$$

where X represents the background gas, produces typically about ten times as much O^+ as does reaction (1). It is important to observe, however, that the ionization energy in reaction (3) is provided by the kinetic energy of the oxygen atom, and the resulting ions therefore have at least 13.6 eV less kinetic energy than those produced by electron impact. The energy analyzer can thus be set to transmit the signal ions while rejecting ions formed by stripping reactions. Similarly, O^{2+} ions produced in the reaction

$$O + X \rightarrow O^{2+} + X + 2e \quad (4)$$

have at least 48.7 eV less energy than the O^{2+} signal ions, and therefore, they can also be rejected by the energy analyzer during acquisition of O^{2+} signal.

3. DATA ACQUISITION AND INTERPRETATION

The ratio of the cross-sections for double and single ionization of atomic oxygen by electron impact can be expressed as

$$(\sigma_{++})/(\sigma_+) = (R_{++}/R_+)(D_+/D_{++}) \quad (5)$$

where: R_+ = measured rate for production of O^+ by electron impact; R_{++} = measured rate for production of O^{2+} by electron impact; D_+ = detection efficiency for O^+ ions and D_{++} = detection efficiency for O^{2+} ions.

The beam intensities and geometries need not be known since they are the same for a measurement of R_+ and R_{++}.

(a) *Measured quantities*

During data acquisition the beam intensities and the count rates are constant to within about 1%, and the rates R_{++} and R_+ are measured alternately, so that small effects due to beam drifts cancel out in the measurement of the ratio. Typically 10,000 counts are recorded for O^+ production, and 400 counts for O^{2+} production, giving statistical uncertainties of ~ 1 and $\sim 5\%$ respectively for these measurements. It is important to note that when alternating between measurements of R_{++} and R_+ only the signal collection optics in the detection region are changed; nothing is changed in the

interaction region where the product ions are formed.

The ion detection efficiencies for the energy analyzer electron multiplier system are determined using a method very similar to that employed by Rundel *et al.* (1979) and are found to be $(33 \pm 2)\%$ for O^+ and $(36 \pm 3)\%$ for O^{2+}. In that the transmission coefficient of the energy analyzer is observed to be about 50%, these results imply a multiplier detection efficiency of about 70% (cf. Stickel *et al.*, 1980).

(b) *Backgrounds and beam modulation scheme*

During O^+ signal acquisition about 5% of the observed count rate is due to processes other than electron impact. The majority of this background is caused by secondary particles that are produced when stripping ions, formed in process (3), strike the energy analyzer. Also some of this background is caused by particles ejected from the neutral beam trap, by the impact of the neutral beam. About 20% of the counts observed during a measurement of O^{2+} signal are background counts, which in this case, are caused almost entirely by particles ejected from the neutral beam trap. Electron beam backgrounds contribute negligibly to the measured count rates.

In order to separate the signal from these backgrounds a beam modulation scheme, similar to the one used by Rundel *et al.* (1979), is employed. In this scheme both the electron and neutral beams are square wave modulated at 1 kHz, 90° out of phase with each other. Two scalers are gated so that one measures the signal plus background count rates when both beams are present, and the other measures the background count rates due to each beam alone. By subtracting the count rates measured by one scaler from the other one can obtain the true signal count rate.

(c) *Consistency tests*

Possible sources of spurious signal in this experiment include background modulation, charge transfer of fast oxygen atoms with thermal ions in the interaction region, and ionization of oxygen atoms by secondary electrons produced in the gun.

The present beam modulation scheme rests on the assumption that the background produced when both beams are present is equal to the sum of the backgrounds produced by each beam alone. A spurious contribution to the measured signal would occur if one beam modulated the background of the other, in particular if the electron beam modulated the background produced by stripping ions. Measurements made at an electron energy of 177 eV however show that R_{++}/R_+ is constant for electron beam currents from 0.15 to 1.5 mA, indicating that background modulation is insignificant.

Thermal ions may be produced in the electron gun by electron impact collisions with surfaces, and with the background gas. Simple calculations, however show that under the present experimental conditions only a relatively small number of ions are produced by these processes. Furthermore, a computation of the potentials in the gun shows that these ions are not significantly trapped in the region near the neutral beam. Nevertheless two tests show directly that charge transfer between fast oxygen atoms and these background ions does not contribute significantly to the measured signal. In one test the ratio of the count rates R_{++}/R_+ is observed to remain constant when the background pressure in the interaction region is increased by an order of magnitude. Furthermore, measurements of R_+ and R_{++}, performed with the electron collector biased at -10 V to collect ions formed from the background gas or surfaces in the collector, show that, to within experimental uncertainty, the ratio R_{++}/R_+ is independent of this bias voltage.

Two measurements demonstrate that the effects of secondary electrons are insignificant. First the ratio of single to double ionization has been measured, at an electron energy of 177 eV, with the beam focusing plates set at the same potential as the cathode. In this arrangement the current to the source exit plate and collector entrance plate is more than 60% as large as the current to the collector. The cross-section ratio, obtained in this manner, nevertheless falls within 1% of the value obtained with the typical operating conditions as described earlier. Also, all the ratios have been measured using a modified electron gun in which the electron beam is collected on a plane electrode located at the position of the collector entrance plate. The ratios measured in this manner agree with the primary data to within about 5% at all collision energies, despite the fact that this arrangement clearly leads to a much larger number of secondary electrons in the interaction region.

4. NEUTRAL BEAM STATE ANALYSIS

The oxygen ion beam is neutralized in the charge transfer cell by the reaction

$$O^+ + N_2 \rightarrow O + N_2^+ + \Delta E \qquad (6)$$

where ΔE is the energy imbalance in the collision. Neutralization of the beam in this manner may lead to excited oxygen neutrals, and so it is important to know by which channels this reaction is likely to proceed and thus be able to deduce in what state the neutrals are formed.

As a first step, the excited state content of the ion beam has been determined using a gas cell attenuation technique similar to the one employed by Turner *et al.* (1968). Only two ionic states are observed when the ion beam is attenuated in either N_2 or Ar, and the proportion of the state with the higher cross-section for attenuation increases as the electron arc voltage is increased. These results are consistent with the findings of Turner *et al.* (1968), and we conclude that the two ionic states observed are the same as those observed by them, namely the ^4S ground state and the ^2D metastable state.

Further, it is observed that the oxygen neutral flux produced from this O^+ beam by charge exchange with N_2 is directly proportional to the flux of ions in what is identified as the ^2D state, and is independent of the $O^+(^4S)$ flux. From this it is inferred that the energy resonant and near resonant reactions

$$\begin{aligned} O^+(^2D) + N_2(X^1\Sigma_g^+,\ v=0) &\\ &\rightarrow O(^3P) + N_2^+(A^2\Pi_u,\ v=0) + 0.4\text{ eV} \\ &\rightarrow O(^3P) + N_2^+(A^2\Pi_u,\ v=1) + 0.1\text{ eV} \\ &\rightarrow O(^3P) + N_2^+(A^2\Pi_u,\ v=2) - 0.1\text{ eV} \\ &\rightarrow O(^3P) + N_2^+(A^2\Pi_u,\ v=3) - 0.3\text{ eV} \end{aligned} \quad (7)$$

constitute the major source of neutral oxygen atoms, which are thus in the ground state. Contributions from the near resonant reaction

$$\begin{aligned} O^+(^2D) + N_2(X^1\Sigma_g^+,\ v=0) &\\ &\rightarrow O(^1D) + N_2^+(X^2\Sigma_g^+,\ v=0) - 0.6\text{ eV} \end{aligned} \quad (8)$$

which results in excited $O(^1D)$ atoms cannot be entirely excluded however. The channels for the production of the low lying ^1S state of oxygen are significantly out of resonance.

The channels for the production of the more highly excited states of neutral oxygen are at least 7.8 eV endothermic, and thus very few neutrals should be formed in these states. Also it is important to observe that the energy required for these reactions must come from the kinetic energy of the projectile atom. Therefore any signal due to atoms in high lying states would have at least 7.8 eV less kinetic energy than the primary beam, and would be rejected by the energy analyzer.

Cross-section ratios measured using Ar, O_2, CO_2 and Kr for neutralization in place of N_2, and ratios measured with the electron arc potential in the source reduced from 40 to 28 eV, are plotted along with the primary data in Fig. 3. The observed agreement among all these data indicates either that excited atoms are not present, or that they do not affect the values of the measured ratios.

5. RESULTS AND DISCUSSION

The ratios of the cross-sections for double and single ionization of atomic oxygen by electron impact are shown in Fig. 3. The energy scale is established by threshold measurements of the single ionization of helium by electrons. The "contact potential" determined in this manner is about 3 V, and thus, 3 V is subtracted from the cathode potential to obtain the electron energy. Absolute cross-sections for double ionization are obtained by multiplying these ratios by absolute cross sections for single ionization of oxygen by electrons. These single ionization cross-sections can be obtained from Fite and Brackmann(1959), Rothe *et al.* (1962), and Brook *et al.* (1978). Cross-sections have also been measured by Boksenberg (1961), however they are larger than the others by about a factor of two, and there is evidence to indicate that they are in error (cf. Kieffer and Dunn, 1966). The first two measurements were relative and were normalized to the cross-sections for total ionization of O_2 of Tate and Smith (1932). The measurements of Brook *et al.* (1978) were absolute and were made using fast beams. In each case the experimenters took steps to show that their reported cross-sections were for ground state oxygen atoms.

If the measurements of Fite and Brackmann, and Rothe *et al.* are normalized using the more recent cross-sections for total ionization of O_2 of Rapp and Englander-Golden (1965) very good agreement is observed with the data of Brook *et al.* (1978). In fact, if the cross-sections of Rothe *et al.* are reduced by 5% to account for contributions from multiple ionization, all three measurements

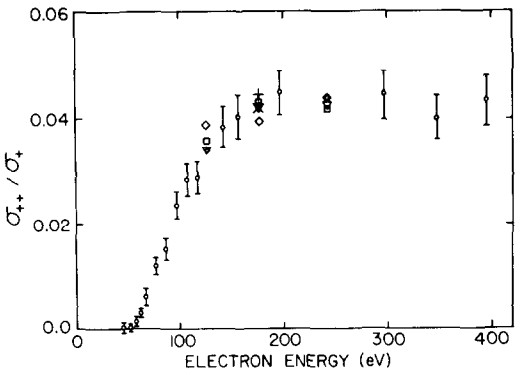

FIG. 3. RATIO OF THE CROSS-SECTIONS FOR DOUBLE AND SINGLE IONIZATION OF ATOMIC OXYGEN BY ELECTRON IMPACT.

Source electron arc potential 40 V and oxygen atom beam formed by charge transfer with N_2–O, CO_2–×, O_2–+, Ar–▽, Kr–□. Source electron arc potential 28 V and oxygen atom beam formed by charge transfer with Kr–◇. Where not shown error bars are approximately equal to those of nearby points.

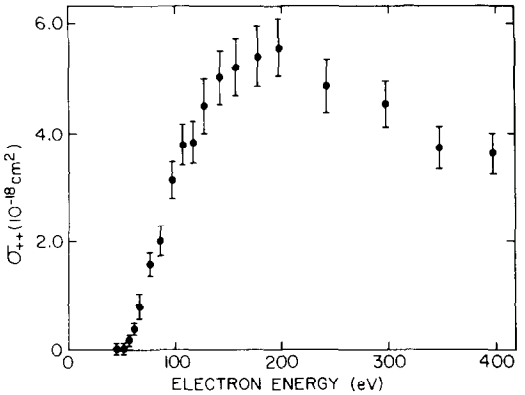

FIG. 4. CROSS-SECTIONS FOR DOUBLE IONIZATION OF ATOMIC OXYGEN BY ELECTRON IMPACT.

agree to within about 5%. The fact that this very good agreement is observed, despite the different measurement techniques, indicates that these data are an accurate measure of the cross sections for single ionization of ground state atomic oxygen by electron impact.

We determine σ_{++} from the present ratio measurements and the data of Brook *et al.* (1978). A linear interpolation procedure is employed to obtain the values of the single ionization cross-sections for the particular collision energies at which the ratios were measured. The error introduced by this procedure along with the experimental uncertainties of the single ionization data is estimated to be about 5%, and therefore it is not considered in computing the error bars for the double ionization cross-sections. These absolute double ionization cross-sections are plotted in Fig. 4. The measured ratios and the absolute cross-sections obtained using oxygen beams neutralized with N_2 are tabulated in Table 1. Apparently, as yet, no quantitative theoretical cross-sections have been calculated for direct double ionization of atomic oxygen by electrons.

The cross-sections presented here will help identify the important sources of O^{2+} in the Earth's plasmasphere and in the Io torus. In fact, these cross-sections, coupled with the analysis of Geiss *et al.* (1978) indicate that direct double ionization of atomic oxygen is not an important source of O^{2+} in the Earth's plasmasphere.

TABLE 1. RATIO OF THE CROSS-SECTIONS FOR DOUBLE AND SINGLE IONIZATION OF ATOMIC OXYGEN BY ELECTRON IMPACT AND ABSOLUTE DOUBLE IONIZATION CROSS-SECTIONS

Electron energy (eV)	$\frac{\sigma_{++}}{\sigma_{+}}$	σ_{++} (10^{-18} cm^2)
45	0.0002(11)	0.02(12)
52	0.0002(08)	0.02(09)
57	0.0014(09)	0.17(11)
62	0.0030(09)	0.38(11)
67	0.0062(18)	0.79(23)
77	0.0120(16)	1.57(21)
87	0.0152(21)	2.00(28)
97	0.0235(26)	3.12(35)
107	0.0284(30)	3.78(39)
117	0.0288(30)	3.84(40)
127	0.0339(37)	4.50(49)
142	0.0384(39)	5.03(51)
157	0.0403(41)	5.20(53)
177	0.0427(44)	5.40(56)
197	0.0449(43)	5.56(53)
242	0.0438(45)	4.85(50)
297	0.0443(46)	4.53(47)
347	0.0400(43)	3.73(40)
397	0.0432(47)	3.63(40)

The numbers in parentheses give the experimental uncertainties in the last two places of the numbers they follow.

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