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MEASUREMENTS OF SELECTED CHARGE TRANSFER PROCESSES AT LOW ENERG--ETC(U)

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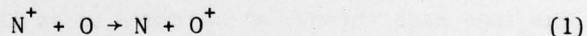
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1. INTRODUCTION

This report summarizes results obtained for the Defense Nuclear Agency under Contract No. DNA001-77-C-0202.

During the artificial disturbance of the atmosphere by a nuclear blast, the free-electron density will be greatly increased, thereby causing a deterioration of the performance of critical electronic systems. It is, therefore, important to know the rate at which the atmosphere returns to its quiescent state. A calculation of this rate of atmospheric deionization requires that the rates of various specific ionic reactions be known. The objective of the research discussed here is to obtain information on the specific ion-neutral reaction



which has been shown to have a significant influence on the deionization rate of the atmosphere.

During the seven months that the program has been active, an intense study of reaction (1) has been conducted. The objective of the study is to obtain charge transfer cross section data for the process at the lowest possible energies obtainable in our experiment. In a previous study of this reaction (Ref. 1), we were only able to determine one upper limit for the cross section. The major emphasis here has, therefore, been to increase both the size of the signal we obtain for the reaction and to lower the detection level of our apparatus.

The following section of this report discusses the experimental approach taken and outlines the changes made to the apparatus (since the previous study) to lower the detection limit. The next section details the approach taken to increase the signal size for reaction (1). The final section gives the problems encountered, the steps taken to overcome these problems, and the preliminary data obtained for the size of the cross section in the low energy regime.

2. THE PRIMARY AND SECONDARY ION SYSTEMS

The crossed-ion modulated-neutral beam apparatus used in this study has been described in the literature (Refs. 2-4). As a consequence, only an abbreviated description of the instrument is given here. The method used to form the neutral beam containing O atoms is described in Section 3.

Figure 1 shows a schematic of the experimental apparatus. The primary ion source shown in the figure is constructed in such a manner that the energy of the electrons used to form the primary ions can be controlled with sufficient precision to allow different concentrations of metastable states in the ion beam to be selected. The primary ions are extracted from this source and mass-analyzed at an energy of 75 eV in a 180° magnetic-mass spectrometer. After analysis, the ions pass through an aperture in an iron plate that shields the magnetic field of the mass analyzer from the succeeding regions of the apparatus. The ions are then retarded or accelerated to the desired collision energy, and pass through a field-free region before intersecting the neutral beam. Collimating apertures ensure that, from purely geometrical considerations, all primary ions pass through the modulated neutral beam. The beam is modulated at 100 Hz by mechanical chopping.

Secondary ions resulting from collisions between the primary ions and neutral species are extracted along the direction of the primary ion beam by an electric field of approximately 2 V/cm. The ions then enter an electric field in which their energy is increased to 1650 eV. Penetration of this accelerating field into the interaction region is reduced by the use of a double-grid structure.

After acceleration, the ions pass through an electrostatic quadrupole lens (Ref. 5) that forms the entrance slit for a 60° -sector magnetic-mass spectrometer. The mass-selected ions impinge on the first dynode of a 14-stage CuBe electron multiplier. The output from the multiplier passes successively through a pre-amplifier, a 100-Hz narrow-band amplifier, and a phase-sensitive detector, and is then integrated. The output is presented on a chart recorder.

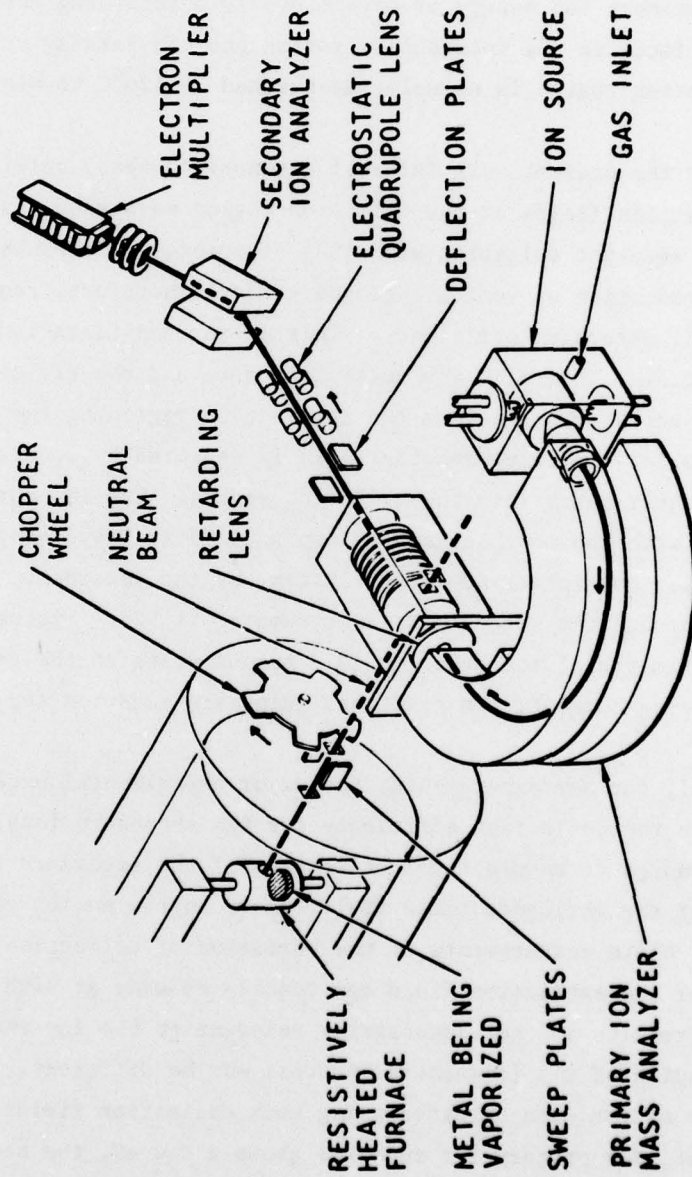


Figure 1. Crossed-ion and neutral-beam apparatus

The primary ion beam intensity is measured at the interaction region with an alkadag coated Faraday cup, which can be moved into the collision region when desired. The primary ion energy is determined from retarding potential measurements. All surfaces in the interaction region and the Faraday cup are gold plated, and the interaction region is normally maintained at 120°C to minimize surface-charging.

Because in the present work interest extended to small-collision energies, only weak extraction fields at the collision region were used. As a result, the secondary ions were not collected with 100% efficiency. Obtaining absolute cross sections for production of various secondary ions, therefore, required determination of their overall detection efficiency. This latter consideration was governed by a number of factors, including the multiplier gain and the efficiency of transmission of the secondary ions from the interaction region to the multiplier.

The multiplier-amplifier-recorder gain is measured by modulating the primary ions prior to their entry into the collision region. The ion current signal is first measured with the movable Faraday cup and, after traversing the secondary mass spectrometer multiplier-amplifier system, by the recorder. Primary ion transmission through the second mass spectrometer is 92%. Typical gains for the entire system are of the order of 10^{15} output volts on the recorder per ampere of incoming current. In practice, gains are measured for each product ion.

In general, for our experiments, the major experimental uncertainty is associated with the collection efficiency for the secondary ions. Collection fields large enough to ensure total collection of the secondary ions cannot be used because of the influence these fields exert on the motion of low-energy primary ions. While measurements of the variation of collection efficiency with the strength of the extraction field can readily be made at high primary ion energies, the results are not necessarily relevant to the low-energy regime, where the dynamics of the ion-neutral process may be different. In most cases, interpretation of the data obtained using weak collection fields is based on the assumption that, for primary ion energies above a few eV, the secondary ions are collected with nearly equal efficiency, and that, as a result, the collection efficiency is independent of both the nature and the energy of the primary ions. This assumption implies that the energy defect in the reaction is not large,

since energy not expended in excitation of the products must appear as kinetic energy and, therefore, would influence the collection efficiency.

For the particular study being conducted here, another method was used to circumvent the problem of determining the collection efficiently. Here, the signals obtained for reaction (1) were placed on an absolute scale by comparison of the signal size for that reaction with the signal size for the reaction



This accidentally resonant reaction has a large cross section which has been measured previously by ourselves (Ref. 1) and other workers (Refs. 6,7). All the previous measurements are in good agreement and this reaction, therefore, serves well as a calibration standard.

The apparatus described above is basically the same as that used for our previous measurements (Ref. 1), but some improvements have been made which lower the detection limit for cross section measurements. The major change made was an upgrade of the magnet on the primary mass spectrometer in the system. The upgrade involved replacement of the original electromagnet by a larger coil with a smaller gap. This improvement enables us to get larger primary ion currents since higher ion energies can be used to mass select the primary ions. This improvement has allowed us to lower our upper limit for detectable signals by a factor of approximately two.

Other improvements in the system have included remounting of the collision chamber and lens systems to ensure more accurate alignment.

3. FORMATION OF THE OXYGEN ATOM BEAM

Beams of neutral particles in our apparatus are made by effusion of a gas or vapor from a small hole in a tube or furnace (see Figure 1). The beam is collimated by two slits in the wall of the separate vacuum chambers employed. Electrically charged plates between the two walls sweep out any ions formed in the furnace. After passing into the experimental chamber, the beam is modulated at 100 Hz by a mechanical chopper.

In our usual mode of operation, the beam density is determined using the cosine law of molecular effusion where the temperature and pressure of the source, the area of the aperture in the tube, and the distance from the source to the interaction region are known factors. A more extensive discussion on the determination of the beam density is given in reference 2.

Three sources of O atoms have been employed during the course of our studies of reaction (1). These sources are: thermal dissociation of O₂ in an iridium furnace, rf discharge of mixtures containing principally O₂ molecules and microwave discharge in similar mixtures. The techniques are described below.

3.1 THERMAL DISSOCIATION

Neutral atomic oxygen was formed by thermal dissociation of pure molecular oxygen in an iridium tube furnace. This technique is similar to that first reported by Fite and Irving (Ref. 8). Thermal dissociation has an advantage over other methods for formation of O atoms such as an rf discharge because problems due to metastable O and O₂ particles in the beam are eliminated.

The major difficulty encountered in the thermal dissociation of the oxygen beam is the low beam density obtained. To get a significant dissociation fraction, it was necessary to use low molecular oxygen pressure in the tube furnace. Pressures of the order of 0.06 torr were employed. At these pressures, dissociation fractions of up to 20% were obtained when the furnace was heated to 2100°K. (The furnace temperature was measured using an optical pyrometer.) The resultant atomic atom-beam density in the collision region was approximately

10^9 atoms/cm³. When calculating this beam density, all necessary corrections were made for the atom velocity and the decrease in the O₂ beam density.

3.2 THE rf DISCHARGE

For studies involving discharges, the normal tube or furnace shown as the beam source in Figure 1 is replaced by a discharge tube. For the rf dissociation studies, two different discharge tube geometries were employed. The first was a 20 mm U-shaped pyrex glass tube. The neutral beam is formed by effusion from a small hole in the outside of the bottom of the U-bend. The rf power was supplied by a 50 MHz supply and the energy was coupled in a gas mixture through a copper band fastened around each leg of the discharge tube. The gas mixture was primarily molecular oxygen with trace quantities of water vapor and molecular hydrogen. This arrangement gave a diffuse discharge which produced atomic oxygen but in rather low quantities.

The second discharge tube employed for the rf studies was a 7 mm quartz tube sealed at one end. A small hole was sandblasted through the sealed end to serve as a port of egress for the gas mixture. The rf power was supplied by the same source as used in the case of the U-tube. A unique feature of this discharge tube was that it possessed a bend near the sealed end. This bend allowed the discharge to be run in a region where no high energy photons produced could exit from the source and enter the collision region without first striking a wall. This precaution was necessary in performing our experiments since the high energy photons had sufficient energy to ionize gas in the neutral beam and produce a coherent noise signal which caused an increase in the lower limit for detection of a signal.

Using a phosphoric acid coat on the tube and the gas mixture described above, it was possible to obtain dissociation fractions of up to 40% when the pressure in the source was 0.3 torr. Careful adjustment of the gas mixture allowed the degree of dissociation to be maximized. The amount of O present in the beam was peaked by observing the signal for reaction (2). A dissociation fraction of 40% corresponds to an atomic oxygen beam density of approximately 2×10^{10} atoms/cm³ which is a factor of 20 better than with thermal dissociation. When the rf discharge was being used, it was found that care had to be taken to eliminate rf pickup in the sensitive detection system used for measurement of the secondary ions. Spurious signals were recorded if this care was not exercised.

3.3 THE MICROWAVE DISCHARGE

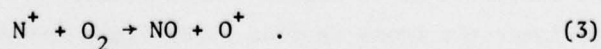
The studies using a microwave discharge as a source of O atoms were performed using the bend quartz tube described above in the rf studies section. The microwave power was supplied by a 100W Raytheon Microtherm unit operating at 2450 MHz. The microwave power was coupled through the vacuum wall using type N connectors. Inside the vacuum system the power was conducted to the discharge region using semi-rigid coaxial cable. The discharge occurred inside an Evenson type cavity through which the quartz tube passed. This cavity was specially constructed to fit into the restricted volume of our neutral beam source chamber. The microwave discharge operated in much the same manner as the rf case and suffered from the same problems such as high energy photon products. There was some indication that the number of metastable states produced by the microwave discharge was greater than the number produced in the rf case. As a consequence, most of the work performed to date using a discharge has been performed using the rf power source.

4. RESULTS TO DATE AND DISCUSSION

In our earlier work on reaction (1) (Ref. 2), we used the thermal dissociation of O_2 molecules in a heated iridium tube to form our O atom beam. Here there are few complications in that the only species emitted from the source are molecular oxygen (with some vibrational excitation), atomic oxygen, and low energy photons. As such, this would seem to be the ideal source for study of atomic oxygen reactions. The problem that was encountered here was the low dissociation fraction which was obtained and the low O_2 pressure which had to be used to achieve that dissociation fraction.

The low O atom density obtained from the thermal source lead us to use a discharge source where higher pressures and dissociation fractions can be realized. The major problem one encounters with a discharge is that a variety of species can be formed and will be emitted from the source. These species include unknown quantities of high energy photons, ions, metastable molecules and atoms. This mixture leads to two problems. The first is the problem of determining the density of the O atoms issuing from the source, and the second is determining the effect these states will have on the measured cross section. As mentioned above, the first of these problems was overcome by using the accidentally resonant charge transfer process between H^+ and O [reaction (2)] as a calibrating reaction to determine the O atom density. The use of this reaction, the cross section of which has been measured by several workers (Ref. 1,6,7), eliminates both the problem of determining the O atom density and establishing the collection efficiency for O^+ .

The second problem is more difficult to circumvent and it has been the attempted resolution of this problem which has been the major undertaking this year. The problem can be illustrated using the following example. Even without the discharge on, a competing reaction is



This reaction is exothermic and even though its cross section is small, it leads to the formation of O^+ if there is some O_2 in the beam. Using the thermal dissociation source, the effect of this reaction could be accounted for by measuring the cross section for reaction (3) with the source cooled to the point where no dissociation takes place and then heating the source, determining the amount of dissociation occurring and subtracting the appropriate amount from the O^+ signal registered.

With a discharge source the problem is not this simple. Here you have the possibility of forming a substantial amount of the $O_2(^1\Delta)$ metastable in the discharge. If the cross section for reaction (3) is different when O_2 is in the ground state or the metastable state, which it may well be, the method of correction described above is no longer valid and different techniques must be employed.

There are two ways to approach this problem. The first is to find a gas to flow through the discharge tube which will produce O atoms but has no other component which leads to O^+ ions in the reaction region. In our studies we have attempted to form O atoms by discharges in CO_2 . The results with the CO_2 discharges are very similar to those obtained with O_2 which are described below.

The second possible way around the problem is to attempt to determine the effect of $O_2(^1\Delta)$ present in the beam formed by a discharge in O_2 . We have attempted to do this by running a discharge in pure O_2 in a clean quartz tube which is not coated with phosphoric acid. Such a system gives very little dissociation but appears to give up to 20% $O_2(^1\Delta)$ (Ref. 9). The result of this experiment is that the amount of O^+ formed in the beam by reaction (3) with the discharge off is very slightly less than that formed with the discharge on. The cross section for formation of O^+ by reaction (3) is therefore relatively independent of the state of the O_2 and this effect will not have a large influence on the size of the measured cross section.

In our earlier studies we determined that the upper limit for the cross section of reaction (1) was $5 \times 10^{-17} \text{ cm}^2$ at 5 eV ion energy. Improvements in our apparatus have allowed us to gain about a factor of two in our detection limit. Further, the increased O atom density obtained using a discharge has given an additional factor of 30 to our detection limit. Theoretically we should therefore be able to lower the cross section upper limit for reaction (1) to approximately $8 \times 10^{-19} \text{ cm}^2$. In practice, however, noise problems inherent in the detection

electronics and associated with the increased beam densities have limited our detection limit to a value of approximately $3 \times 10^{-18} \text{ cm}^2$.

During the course of the experimental program to date, some data have been obtained for reaction (1) in the center-of-mass energy range from 0.5 eV to 5 eV. These data consistently indicate that the cross section for reaction (1) lies between 5×10^{-18} and $2 \times 10^{-17} \text{ cm}^2$ in this energy range. To date there has not been time to obtain all the data required or to fully interpret the data which has been obtained. As a consequence, the cross sections given above must be considered preliminary.

5. SUMMARY

During the seven months the program has been active, considerable work has been done in developing and understanding the operation of various O atom sources. In addition, some modifications have been made to the apparatus to improve the detection limit for charge transfer cross sections. Also, a considerable amount of data have been accumulated using different gases in the discharge tube. Use of this data has allowed us to make a preliminary evaluation of the cross section for reaction (1). More data and interpretation are required to arrive at a final value.

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