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# Absolute differential and total cross sections for charge transfer of $O^+$ ground and mixed states ions in $N_2$

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**Abstract**

We report measurements of the total and absolute differential cross sections for charge transfer of ground- and excited-states $O^+$ ions at 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5 and 5 keV in collisions with $N_2$ at scattering angles $-5.2^\circ \leqslant \Theta \leqslant +5.2^\circ$ in the laboratory frame. Total cross sections for ground- and mixed state ions are compared with previous measurements. The behaviour of the absolute differential cross sections display an expected decreasing behaviour with increasing angle. The mixed state ions cross sections are considerably higher than those measured for the ground state ions.

## Introduction

The importance of charge transfer of $O^+$ in $N_2$ has been established from the fundamental atomic physics point of view. One important reason for this is the fact that the cross section is strongly dependent on the initial electronic state of the $O^+$ ions [1–5], and in this case the two low-lying excited states are $^2D$ and $^2P$ with approximate lifetimes of 3.6 h and 5 s, respectively [6]. The $O^+$ ($^2D$) metastable state is in resonance with the $N_2(A^2\Pi_u)$ state and the cross section is expected to be large, while there is 2 eV between the $O^+$ ($^4S$) and the nearest $N_2^+(X^2\Sigma_g^+)$ state, resulting in smaller cross section. Equally charge transfer of $O^+$ with $N_2$ is of importance in the field of aeronomy [7, 8] since the $O^+$ is the dominant ion in the F region of the atmosphere and both metastable species of $O^+$ ($^2D$) and $O^+$ ($^2P$) have been detected. Furthermore one of the major magnetospheric ions near the orbit of Titan are $O^+$ ions [9]. Titan occasionally orbits outside of Saturn's magnetosphere and is in direct contact with the solar wind. This occurs when the solar wind pressure is high enough to significantly compress Saturn's magnetosphere. The solar wind may penetrate into Titan's upper atmosphere. Charge exchange measurements are important since the $O^+$ ions interact with neutrals in the extended regions of the atmosphere called the atmospheric corona. The charge exchange produces fast

  

neutrals that are unaffected by the local fields and can directly penetrate into the atmosphere making collisions with the atmospheric neutrals causing heating, collisional ejection of atoms and molecules and expansion of the corona.

Clearly the $O^+$ reactions with $N_2$ have received a considerable amount of attention and several experimental studies [1, 4, 5, 10, 11] and more recently [12] have been conducted, yet within the keV region of interest to this investigation, there still exists a large amount of doubt to the magnitude of the total charge transfer cross section. Uncertainties quoted by early groups of investigators are rather high and there is lack of agreement among them with the exception of the apparent agreement for the total cross section measurements of Lindsay *et al* [12] and Moran and Wilcox [3], although they seriously disagree with respect to the ground state cross sections. Lindsay *et al* [12] conducted an investigation into the relationship of the differential cross sections (DCSs) and the initial electronic state of the $O^+$ ions; they found strong dependence between the DCSs and the projectile degree of excitation, and reported differential and integral cross sections in the low keV energy range for both ground state and metastable $O^+$. Their finding showed that the metastable state exhibits a significantly higher cross section compared to the ground state ions in the lower scale of their energy, while the two cross sections tend to be of the same magnitude as the collisional energy reaches 5 keV. Although Lindsay *et al* [12] measured the differential cross sections up to 3.1° for the state selective $O^+$ ions, to the best of our knowledge, there has been no previous measurements of the total differential cross section for the process investigated here.

This paper reports measurements of the differential and total charge exchange cross sections. The presently measured total charge exchange cross sections are compared to those available in the literature.

## Experimental apparatus

The experimental apparatus and technique needed to generate the fast ion beam were recently reported [13]. Briefly, the $O^+$ ions formed in an arc discharge source containing $O_2$ gas (99.99% purity) at ion source pressures of 0.02–10 mT, accelerated to the desired energy and selected by a Wien velocity filter. The $O^+$ ions were then allowed to pass through a series of collimators before entering the gas target cell, which was a cylinder of 2.5 cm in length and diameter, with a 1 mm entrance aperture, and a 2 mm wide, 6 mm long exit aperture. The target cell was located at the centre of a rotatable, computer-controlled vacuum chamber that moved the whole detector assembly which was located 47 cm away from the target cell. A precision stepping motor ensured a high repeatability in the positioning of the chamber over a large series of measurements. The detector assembly consisted of a Harrower-type parallel plate analyser and two channel electron multipliers (CEMs) attached to its exit ends. The O atoms passed straight through the analyser. Separation of neutral and charged particles occurred inside the analyser, which was set to remove the charged fragments and detect the neutral atoms ($I_f(\theta)$) per unit solid angle per second at a laboratory angle $\theta$ with respect to the incident beam direction with the on-line CEM. A retractable Faraday Cup was located 33 cm away from the target cell, allowing the measurement of the incoming $O^+$ ion–beam current ($I_0$ is the number of $O^+$ ions incident per second on the target). Under thin target conditions used in this experiment, the differential cross sections for the O formation were evaluated from the measured quantities by the expression

$$\frac{\mathrm{d}\sigma(\theta)}{\mathrm{d}(\Omega)} = \frac{I_f(\theta)}{I_0 n l} \tag{1}$$

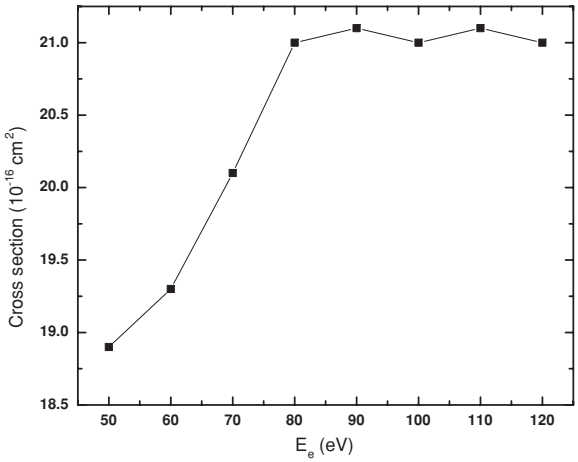


**Figure 1.** Charge transfer cross section as a function of the ionizing electrons.

where $n$ is the number of target atoms per unit volume (typically $1.2 \times 10^{13}$ atoms cm$^{-3}$) and $l$ is the length of the scattering chamber ($l = 2.5$ cm). The total cross section $\sigma_{10}$ for the production of the O particles was obtained by the numerical integration of $d\sigma/d\Omega$ over all angles measured. For $\theta \geqslant \theta_m$ the differential cross sections vanish ($\theta_m$ is the maximum angle). Care was taken when the absolute differential cross section was measured. The reported value of the angular distribution was obtained by measuring it with and without gas in the target cell with the same steady beam in order to eliminate the counting rate due to background effects. The estimated rms error is 15%, while the total cross sections were reproducible to within 15% from day to day. Several sources of systematic errors are present and have been discussed in a previous paper [13]. The absolute error of the reported cross sections is believed to be less than 15%. This estimate accounts for both random and systematic errors.

## Results and discussion

The evaluation of the exact metastable/ground state $O^+$ fraction in the ion beam and its effect on the measured cross section was determined by investigating the influence of the electron energy (Colutron ion source) on the $O^+$ ions produced in the ion source as well as the ion source pressure effect on the metastable/ground state $O^+$ fraction. Cross sections were measured for $O^+$ ions at electron energies within the range of (50–120) eV. The results are presented in figure 1. The total cross section shows a sharp increase for electron energies between 50 and 80 eV. At $80 \leqslant E_e \leqslant 120$ eV, the cross section remains constant indicating constant metastable/ground state $O^+$ fraction. Electron energy ($E_e$) $\geqslant 80$ eV was chosen and maintained during the course of the present experimental work in order to have quantitative assessment of the metastable/ground state $O^+$ fraction as well as to obtain an accurately measured cross sections for ground- and mixed state ions in collisions with $N_2$.

One method of removal of metastable species from an ion beam is the collisional quenching of the excited species. Once the electron energy of $\geqslant$80 eV was chosen, the cross section at 3 keV accelerating energy was measured for source pressures between 0.02 and 10.0 mT and the results are plotted in figure 2. The cross section remains constant within source pressures of 0.02–0.1 mT. At pressures $\geqslant$0.1 mT, the cross section decreased rapidly from $21.00 \times 10^{-16}$ cm$^2$ at 0.1 mT to $6.00 \times 10^{-16}$ cm$^2$ at 2 mT. Further increases in the ion source

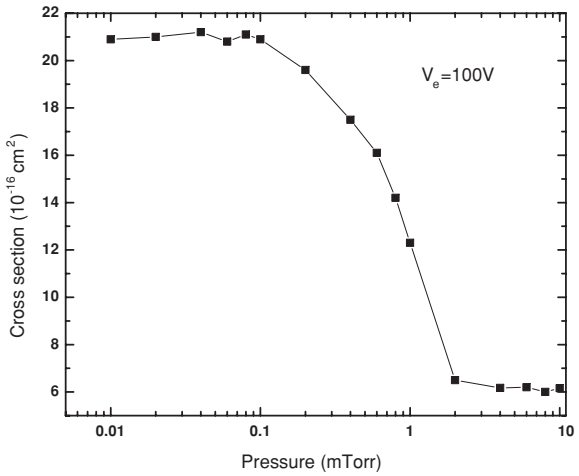


**Figure 2.** Charge transfer cross section as a function of ion source pressure.

pressure up to 10 mT showed no changes in the measured cross section. The sharp decline in the cross section between 0.1 and 1.0 mT can be explained in terms of quenching and depleting the excited metastable species within the ion beam. As a consequence, it is safe to assume the cross section at source pressures $\geqslant$2 mT are those pertaining only to the ground state $O^+$ ions or to a mix of ground state and excited-state ions provided that they have the same cross sections, while the cross section at source pressures below 0.1 mT pertains to an ion beam that is composed of a mix of ground- and excited-state ions. The exact metastable/ground state $O^+$ fraction was determined by measuring the ion–beam intensity at 0.04 mT and 6 mT at the exit point of the Wien velocity filter. Direct measurements yielded 30% and 70% ground state and excited-state ions, respectively. On the other hand minor fluctuations in ion–beam intensities prevented accurate determination of an attenuation curve of beam intensity verses target cell pressure in order to confirm the above-mentioned fraction. As a result, it is possible to assume the decrease of about 71.5% (see figure 2) in the measured cross section as the source pressure was increased above 2 mT, points to cross sections of metastable species that are higher than those of ground state cross section. This fact has been verified (within the energy range of the present investigation) by several groups of investigators [1, 11–12].

Absolute DCSs data for charge transfer of $O^+$ ions impinging on $N_2$ target have been measured at laboratory angles $-5.2^\circ \leqslant \Theta \leqslant 5.2^\circ$ and collisional energies $1.0 \leqslant E \leqslant 5.0$ keV. Measurements were carried at source pressures of 0.02 mT and 6 mT and the results are plotted in figures 3 and 4. Our measured DCSs for all collisional energies show a decrease with increasing angle. The detected number of scattered particles at 5.2° is about four orders of magnitude smaller than those detected at zero scattering angle. Evidently this points out to about total collection of the scattered particles. Smaller cross section and background effect prevented any meaningful measurements at scattering angles higher than 5.2° particularly at the lower collisional energies.

The differential cross sections were integrated to yield the total cross sections for ground- and mixed state ions. The results are plotted in figure 5, together with the data of Lindsay *et al* [12] for $O^+(^2D$ and $^2P)$ and $O^+(^4S)$; Moran and Wilcox [1] for ground and metastable cross sections; Li *et al* for total and ground state cross sections; Rutherford and Vroom [2] for $O^+(^2D)$; Hoffman *et al* [5] for ground- and mixed state ion cross sections

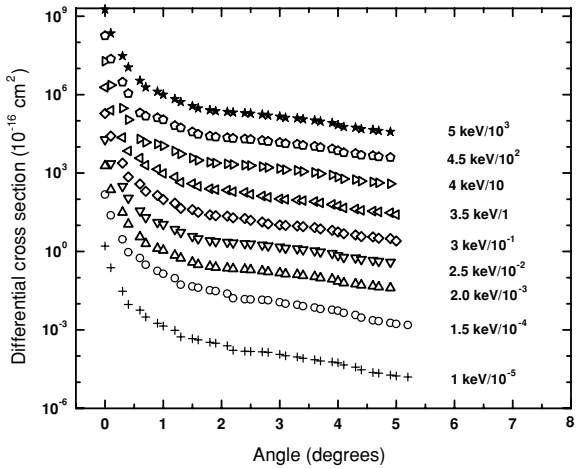


**Figure 3.** Charge transfer differential cross sections for a mix of ground- and excited-state ions.

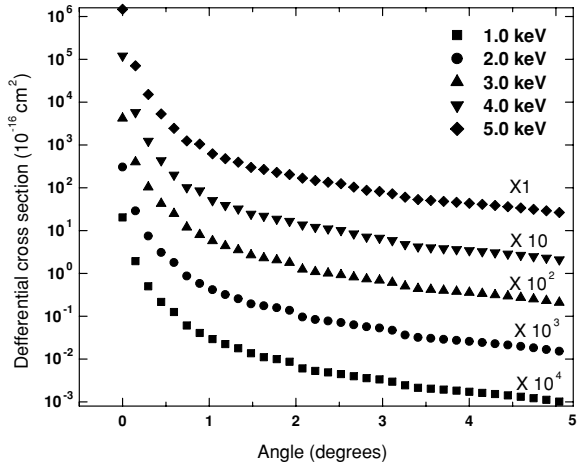


**Figure 4.** Charge transfer differential cross sections for ground state ions.

and Flesch and Ng [10] for ground state ions for comparison. Error bars of $\pm 15\%$ are given to indicate the maximum reproducibility of the data in the present investigation.

Figure 5 manifests the overall general shape of the curve of charge transfer cross sections for the $O^+$–$N_2$ system over a wide range of energies (0.001–20 keV). Since full analysis of the differential rate equations for tenuous target thicknesses need to be corrected to a second order, we have performed the following analysis in order to estimate the nonlinear contribution (curvature) of the growth curve. We calculated the cross section taking into account the possibility of multiple collisions. In considering a three-component system, we have (McDaniel *et al* 1993) [15]

$$F_j = \sigma_{10}\, nl + \tfrac{1}{2}[\sigma_{1-1}\sigma_{-10} - \sigma_{10}(\sigma_{10} + \sigma_{1-1} + \sigma_{01} + \sigma_{0-1})]\, n^2 l^2 \qquad (2)$$

where $F_j$ is the fraction of the beam in the charge state $j$ which is evaluated from the measured beam intensities with and without gas in the target cell. As the two charge transfer

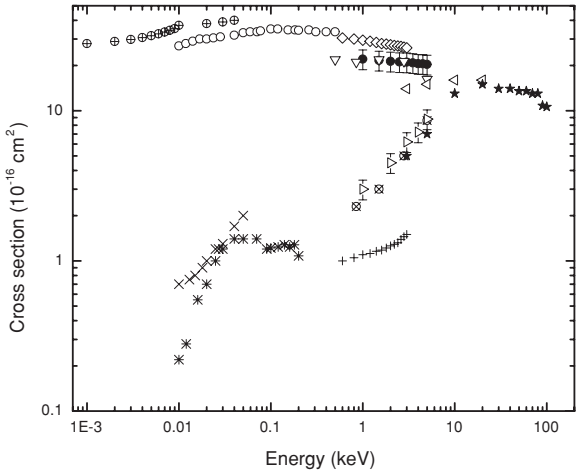


**Figure 5.** Cross sections for charge exchange of $O^+$ ions in $N_2$ molecular target measured by various groups including the present results: ● present results for mixed state ions, ⊕ from Li *et al* [11] for $O^+(^2D)$, ⊗ from Lindsay *et al* [12] for ground state ions, ▽ from Lindsay *et al* [12] for excited-state ions, ○ from Rutherford and Vroom [2] for excited-state ions, ▷ from Hoffman *et al* [5] for mixed state ions, ▷ present results for ground state ions, ◇ from Moran and Wilcox [3] for excited-state ions, + from Moran and Wilcox [3] for ground state ions, ⋆ from Hoffman *et al* [5] for ground state ions, × from Li *et al* [11] for ground state ions, and ∗ from Flesch and Ng for ground state ions.

**Table 1.** Measured and corrected cross sections taking into account second-order corrections.

| $E$ (keV) | $F_j$ ($10^{-16}$ cm$^2$) Fraction | $\sigma_{01}$ ($10^{-16}$ cm$^2$) Measured | $\sigma_{10}$ ($10^{-16}$ cm$^2$) Corrected |
|---|---|---|---|
| 1.0 | 6.63 | 22.1 | 22.9 |
| 2.0 | 6.39 | 21.3 | 22.1 |
| 3.0 | 6.3 | 21.0 | 21.7 |
| 4.0 | 6.18 | 20.6 | 21.3 |
| 5.0 | 6.09 | 20.3 | 21.0 |

cross sections are much smaller than those of one electron processes, equation (2) can be approximated yielding

$$F_j = \sigma_{10}\, nl + \tfrac{1}{2}[\sigma_{10}(\sigma_{10} + \sigma_{01} + \sigma_{0-1})]\, n^2 l^2. \tag{3}$$

Taking the values of $\sigma_{10}$ and $\sigma_{01}$ from Lindsay *et al* [12] for 3 keV $O^+ - N_2$ system, together with the fractions measured in this work at $nl$ value of $3.3 \times 10^{13}$ atoms cm$^{-2}$, we calculated the value of $\sigma_{10}$ using equation (3) for all energies studied in this work and the results are presented in table 1. The value obtained for $\sigma_{10}$ has a maximum error of 3.6% compared to the value that we have obtained by integrating the angular distribution. The data in figures 3, 4 and 5 for differential and total cross sections are corrected to account for the second-order correction.

The data in figure 5 are basically of two categories: the first that involve measurements of slow product ions and the second are those in which the fast neutrals are detected. Both methods lead to the same result. Rutherford and Vroom [2], Flesch and Ng [10] and Li *et al* [11] observed the slow product ions. Moran and Wilcox [3], Hoffman *et al* [5] and Lindsay *et al* [12]

all detected the fast neutrals. The various above-mentioned studies employed different techniques in order to obtain total and state specific cross sections. Generally, the ground state cross section is expected to be considerably smaller than that of the excited-state ions at low collisional energies. This is due to the fact that charge exchange with the ground state is relatively unlikely at low impact energies, because the approximately 2 eV between the $O^+(^4S)$ and that of the nearest $N_2^+$ state $\left(X^2\Sigma_g^+\right)$ that is required for the ground state reaction to proceed must come from the kinetic energy of the incident ion. As the kinetic energy is increased, this 2 eV becomes a smaller fraction of the impacting ions kinetic energy and thus more easily available. As a result, we assume here at energies below 3 keV, the contribution of the metastable species to the total cross section is dominant. Moran and Wilcox [3], Hoffman *et al* [5] employed controlled electron impact ion source to produce $O^+(^4S)$ ground state beam and mixed beam. They operated their ion source at $10^{-5}$ mT pressure in order to maximize the excited-state fraction by minimizing the occurrence of ion–molecule reactions in the source. However, Hoffman *et al* [5] found that there were no measurable differences in the cross section when their ion source pressure was varied from about 5 to 50 mT similar to our finding above 2 mT as shown in figure 2. Li *et al* [11] coupled dissociative charge transfer and rf octopole ion trap for beam separation, while Lindsay *et al* [12] filtered out the metastable state ions. As for the mixed state work, Moran and Wilcox [3], and Lindsay *et al* [12] measured the cross sections for the mixed state ion beam. Rutherford and Vroom produced $O^+$ ions by dissociative charge exchange and determined the abundance of $O^+$ ($^2D$) ions and based their observations on the small cross section for the $NO^+$ formation.

In order to compare our present data for the total charge transfer of $O^+$ in $N_2$, it is imperative that uncertainties of the data reported by various groups are available. Rutherford and Vroom do not explicitly state the accuracy of their data, yet as their experimental apparatus is the same as that of employed by Stebbings *et al* [1] and stated to be good to better than a factor of 2. Moran and Wilcox [3] identified the absolute error associated with their measured cross sections for ground and metastable states to be approximately $\pm 7\%$ resulting from the absolute measurements of the target gas concentration and the assessment of the neutral beam flux. Moran and Wilcox [3] derived their metastable fractions from the data of Turner *et al* [14], whom they state that metastable fractions apply strictly to their experimental configuration which it seems there should be a considerable degree of uncertainties associated with the procedure used by Moran and Wilcox [3]. Lindsay *et al* [12] stated their uncertainties for ground and metastable states to be about $\pm 16\%$. These errors were used in evaluating their total cross sections for the $O^+$ in $N_2$ which are plotted in figure 5. The data of Li *et al* [11] for total cross sections are considerably higher than all other groups. That may be due to the fact that their ion beam was composed of higher fraction of metastable ions (89% $O^+(^2P)$ and 5% $O^+(^2D)$) compared to other groups. Our present ground state cross sections are in excellent agreement with those of Lidsay *et al* [12] and Hoffman *et al* [5]. Taking into account that the ground state cross section is considerably smaller than that of metastable ions, it can be seen from figure 5 that our total cross section would overlap well with that of Lindsay *et al* [12] and to some extent with those of Hoffman *et al* [5]. This agreement with the results of Lindsay *et al* [12] can be explained by the fact that fractions of ground and excited ions within our ion beam is similar to those stated by Lindsay *et al* [12]. The agreement between our present results and those of Hoffman *et al* [5] can be understood due to the similarities in conditions employed in obtaining the ion beams. Hoffman *et al* [5] determined the reactant ion electronic state population by the electron kinetic energy in the ion source. A reactant ion beam with a mixed population distribution was produced by increasing the electron energy above the threshold for excited-state production. They used electron kinetic energy of about 105 eV and stated that the abundance of the metastable ions was dependent upon the pressure in the ion

source as well as the electron energy. Their estimated uncertainty was given to be 10%. Our ion source operating conditions (source pressure of 0.04–0.07 mT) for the mixed state ion beam are similar to that used by Hoffman *et al* [5] with the exception that it cannot be operated at lower electron energies. Such operating conditions close to those stated by Hoffman *et al* [5] are expected to produce a mixed state beam of similar metastable abundance. The fact that our measured total cross section is about 30% higher than that of Hoffman *et al* [5] do confirm that expectation to a certain extent. The ground state cross sections of Moran and Wilcox [3] lie considerably lower than our present data for the ground state. It is worth mentioning that recent measurements of the charge transfer cross section for $O^+(^4S)$ in $H_2$ by other investigators [15–16] also lie higher than those of Moran and Wilcox [3] for the relevant cross sections. It is most likely that all the direct total cross section measurements of mixed ions of the various groups are valid within the experimental parameters of each of the methods employed. It is these initial conditions of the ion source with respect to electron energy and pressure that are expected to influence the total cross sections. The weakness of the attenuation technique in evaluating the metastable fractions accurately can seriously influence the evaluation of the state selective cross sections, yet it has no baring on the total cross section. Higher fractions of metastable species would certainly result in larger total cross sections particularly at energies below 3 keV. This fact is confirmed by Li *et al* [11] whom they employed a state selective method in a combination of dissociative charge transfer and rf octopole ion trap in obtaining their ion beam. The higher fraction of metastable species within their ion beam is clearly reflected in the magnitude of the total cross section measured by this group for the main product channel of $N_2^+ + O$.

## Conclusion

Total and absolute differential cross sections for charge transfer of 1.0–5 keV $O^+$ ions in their mixed and ground state in collisions by $N_2$ at scattering angles $-5.2^\circ \leqslant \Theta \leqslant +5.2^\circ$ in the laboratory frame have been determined. The excited-state cross section is very much higher than that for the ground state. Reliable previous measurements for total cross section measurements are in good agreement with those reported here for mixed state and ground state ions. It is our opinion that differences in the measured total cross sections are mainly due to different metastable fractions within the ion beams reported by various groups. These results are of fundamental interest and have potential applications in fields of aeronomy and interplanetary physics.

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