Absolute differential and total cross sections for charge transfer of O^+ with H_2

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Absolute differential cross sections (DCS's) are reported for charge-transfer scattering of 0.5-, 0.85-, 1.5-, 2.8-, and 5.0-keV O^+ ions by H_2 molecules at angles between 0.04° and 5.4° in the laboratory frame. Cross sections for $O^+(^4S)$ ground state and $O^+(^2D,^2P)$ metastable ions are presented. The ground-state differential cross section is much less forward peaked than is the metastable differential cross section. Over most of the range of impact energies studied, the $O^+(^4S)$ total cross section is found to be approximately six times smaller than that for $O^+(^2D,^2P)$. Based on the DCS data reported here, a possible explanation for the large discrepancies in the published ground-state total cross sections is given. The present data confirm the existence of a minimum in the ground-state total cross section at about 2 keV, which has been observed by some other workers. [S1050-2947(99)00205-X]

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INTRODUCTION

Despite the increasing sophistication of experimental techniques, the accurate determination of state-selected charge-transfer cross sections has remained a challenging task. The charge-transfer reaction of O^+ with H_2 is similar to that of O^+ with N_2 [1] in that the ground state O^+ ion has only nonresonant channels available, whereas reactions with the metastable ion are essentially resonant. Charge transfer of $O^+(^4S)$ ground-state ions with H_2 is approximately 1.8 eV endothermic and the charge-transfer cross section for this process would therefore be expected to be smaller than that for $O^+(^2D,^2P)$ metastable ions and to fall significantly with decreasing projectile energy. While the available experimental data confirm these general trends, a literature survey of the published cross sections for the reaction of O^+ with H_2 shows marked disagreements both in the absolute values and in the energy dependences of the measured ground-state cross sections. The primary purpose of the present work is to attempt to resolve some of these ambiguities.

Charge-transfer reactions have long been recognized as playing key roles in many diverse environments. Atomic oxygen is an abundant impurity in tokamak edge plasmas, second in importance only to carbon [2]. In these hydrogen plasmas the temperature is low enough, and the particle densities are high enough, to permit significant rates of charge-transfer collisions between oxygen ions and hydrogen molecules which lead to undesirable plasma cooling. Knowledge of the relevant cross sections is therefore necessary in order to understand the detailed behavior of these plasma devices. Charge transfer of O^+ with H_2 is also important to the aeronomy of Jupiter. Evidence for energetic particle precipitation into Jupiter's upper atmosphere was initially provided by the Voyager 1 and Voyager 2 spacecraft [3,4]. Subsequent observations have indicated that the precipitating auroral particles may be oxygen and sulfur ions from the Io plasma torus ([5, 6], and references therein). One of the most important processes by which these ions interact with the neutral atmosphere is charge transfer with H and H_2 [5], and in order to model the Jovian atmosphere effectively accurate cross-section data are needed.

Differential cross-section (DCS) and total cross-section measurements are reported here for charge-transfer scattering of ground-state $O^+(^4S)$ and metastable $O^+(^2D,^2P)$ oxygen ions with H_2. As was the case for the reaction of O^+ with N_2 [1], simple theoretical considerations indicate that the DCS's should have a strong dependence on the projectiles' state of excitation. The total cross sections reported here are for production of fast neutral O atoms. Neither the ultimate state of the reactants, nor whether the slow product ions are H_2^+ or H^+, is assessed. It should be noted that both metastable states of O^+ may be produced within the ion source used in the present study. Since it is not possible, using the techniques employed here, to differentiate between them, the metastable cross sections reported here pertain to an unspecified mixture of $O^+(^2D)$ and $O^+(^2P)$.

APPARATUS AND EXPERIMENTAL METHOD

The apparatus and the experimental method have both been described in detail previously [1,7]. The apparatus is shown in Fig. 1. Either O_2 or CO is admit-ted to a magnetically confined plasma ion source.[1] Ions are extracted from the source through a small aperture, accelerated, and focused to form a beam of the desired energy. Two confocal 60° sector magnets are used to select ions of the desired mass to charge ratio. Ions passing through a pair of laser drilled apertures, separated by 23 cm, form a beam with an angular divergence of approximately 0.03°. The first aperture, 100 μm in diameter, forms the exit of the filter cell. The second aperture, 20 μm in diameter, forms the entrance to the target cell whose length is approximately 1 mm. A 200 μm aperture forms the exit of the target cell. A position-sensitive

[1] O_2 was originally used as a source gas, but to avoid the possibility of producing O_2^{2+} ions the source gas was changed to CO. The measured cross sections were found to be independent of source gas used.

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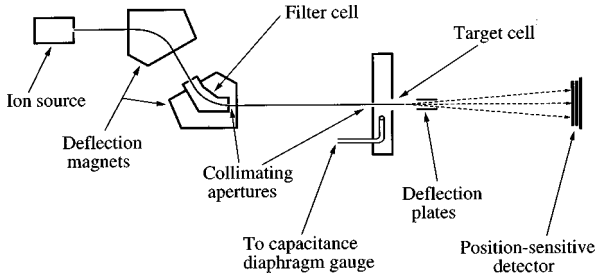

FIG. 1. Schematic of the apparatus.

detector (PSD) [8] normally located 26 cm beyond the target cell serves to measure the flux of ions passing through the target cell and to measure the flux and positions of impact of product neutral O atoms. A pair of deflection plates located between the target cell and the PSD is used to deflect the ion beam when required.

In order to measure the differential charge-transfer cross section, H_2 is admitted to the target cell and the angles of scatter of the neutral O atoms, formed by charge transfer of the primary O^+ ions, are determined from their positions of impact on the PSD. Unscattered primary O^+ ions are normally deflected from the PSD but are allowed to impact the PSD periodically to assess the primary beam flux. These measurements, together with the target number density and target length, are sufficient to determine the DCS. The O^+ beam, as it emerges from the ion source, consists of both ground-state and metastable-state components. Cross sections for both components of the ion beam are obtained by utilizing the fact that $O^+(^4S)$ ground-state ions have a much lower charge-transfer cross section with N_2 than $O^+(^2D,^2P)$ metastable ions. During the measurement of the ground-state cross section, the filter cell is filled with several mtorr of N_2. The emerging O^+ beam then consists essentially of only ground-state ions as practically all of the incident $O^+(^2D,^2P)$ ions are converted to neutrals [1]. The $O^+(^2D,^2P)$ cross section is determined by evacuating the filter cell, measuring the effective cross section for the mixed composition beam, determining the fraction of ions in the ground state, and subtracting the contribution these ground state ions have made to the total scattering signal. The effective cross-section measurement and two fraction measurements are generally made within a period of less than 1 h. The fraction of ions in the ground and excited states is measured using a modified version of the attenuation technique originally developed by Stebbings, Turner, and Rutherford [9]. As noted in a previous publication, only two components are readily identifiable from these attenuation curves, a ground-state component and a metastable excited-state component.

With the PSD located 26 cm from the target cell, it is only possible to measure cross sections for scattering out to approximately 3°. For many of the processes previously studied, this was not a significant limitation as almost all the neutrals were scattered into angles less than this and the integral cross sections obtained were thus, to a very good approximation, equal to the total cross sections. Indeed, from inspection of the metastable state DCS's shown in Fig. 2 it is apparent that only a small fraction of the scattered neutrals, estimated to be less than 8%, fail to impact the PSD. Hence, within the uncertainties reported, the metastable state inte-

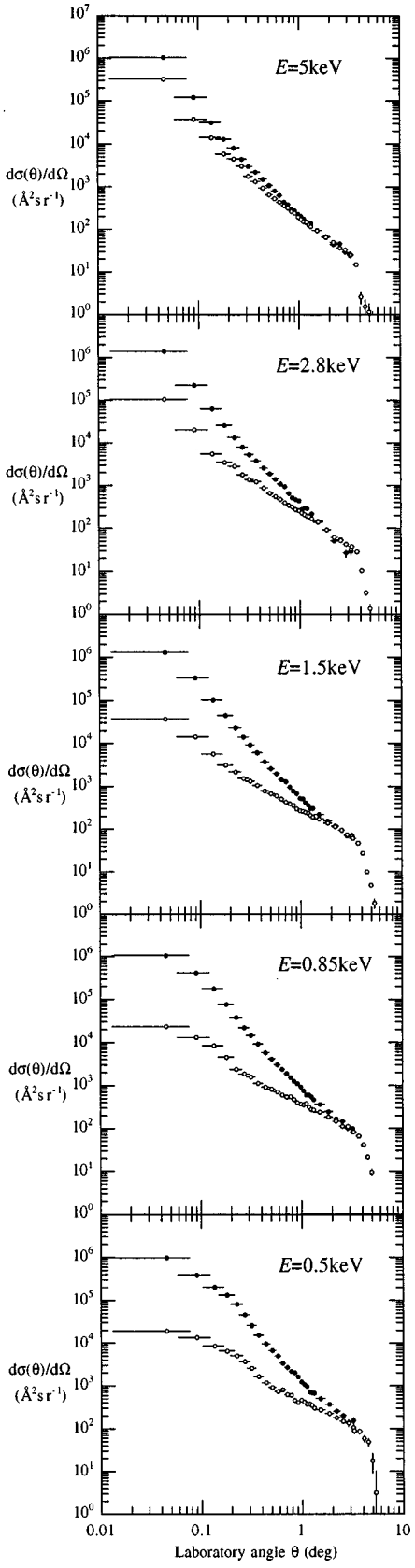

FIG. 2. Absolute differential cross sections for charge-transfer scattering of $O^+(^4S)$ by H_2 (hollow circles) and $O^+(^2D,^2P)$ by H_2 (filled circles) at the projectile energies indicated.

TABLE I. Laboratory frame differential charge-transfer cross sections for $O^+(^4S)$-H_2 collisions, where E is the projectile energy and the numbers in square brackets represent powers of ten.

Laboratory angle θ (deg)	$\frac{d\sigma(\theta)}{d\Omega}$ (Å^2 sr^{-1})				
	$E=0.5$ keV	$E=0.85$ keV	$E=1.5$ keV	$E=2.8$ keV	$E=5$ keV
0.045	1.92[4]	2.33[4]	3.73[4]	1.06[5]	3.26[5]
0.090	1.35[4]	1.31[4]	1.42[4]	2.05[4]	3.77[4]
0.135	8.52[3]	8.42[3]	5.67[3]	5.50[3]	1.40[4]
0.180	6.53[3]	4.53[3]	3.13[3]	3.49[3]	5.87[3]
0.225	5.03[3]	2.38[3]	2.18[3]	2.79[3]	4.46[3]
0.270	3.70[3]	1.86[3]	1.53[3]	1.77[3]	3.03[3]
0.315	2.55[3]	1.59[3]	1.33[3]	1.37[3]	1.78[3]
0.371	1.64[3]	1.13[3]	1.05[3]	1.22[3]	1.34[3]
0.438	1.17[3]	9.16[2]	7.83[2]	8.72[2]	9.39[2]
0.506	9.13[2]	8.24[2]	6.77[2]	6.51[2]	6.45[2]
0.573	7.31[2]	7.11[2]	6.00[2]	5.46[2]	5.29[2]
0.641	8.07[2]	5.99[2]	5.09[2]	4.68[2]	4.36[2]
0.708	6.21[2]	5.39[2]	4.32[2]	3.89[2]	3.62[2]
0.776	6.00[2]	5.46[2]	3.94[2]	3.41[2]	3.15[2]
0.843	4.59[2]	4.68[2]	3.61[2]	3.13[2]	2.62[2]
0.911	4.11[2]	3.98[2]	2.96[2]	2.73[2]	2.39[2]
0.978	4.63[2]	3.69[2]	2.69[2]	2.56[2]	1.95[2]
1.05	4.20[2]	3.50[2]	2.58[2]	2.29[2]	1.72[2]
1.11	3.74[2]	3.80[2]	2.48[2]	2.08[2]	1.53[2]
1.18	3.73[2]	3.10[2]	2.32[2]	1.92[2]	1.47[2]
1.25	3.46[2]	2.75[2]	2.11[2]	1.86[2]	1.27[2]
1.32	3.03[2]	2.67[2]	1.91[2]	1.66[2]	1.15[2]
1.52	2.75[2]	2.41[2]	1.73[2]	1.38[2]	9.37[1]
1.85	2.25[2]	1.85[2]	1.38[2]	9.17[1]	6.53[1]
2.19	1.81[2]	1.48[2]	1.14[2]	6.19[1]	4.98[1]
2.53	1.49[2]	1.12[2]	9.43[1]	5.22[1]	3.67[1]
2.87	1.34[2]	1.10[2]	7.40[1]	4.29[1]	3.32[1]
3.20	1.07[2]	8.24[1]	6.83[1]	3.28[1]	2.67[1]
3.27	8.87[1]	8.22[1]	6.10[1]	3.78[1]	2.52[1]
3.68	8.71[1]	6.61[1]	4.69[1]	2.84[1]	1.49[1]
4.10	5.91[1]	4.16[1]	2.69[1]	1.04[1]	2.63[0]
4.52	4.91[1]	2.16[1]	9.88[0]	3.19[0]	1.56[0]
4.94	1.76[1]	9.49[0]	4.85[0]	1.36[0]	1.17[0]
5.35	3.16[0]		1.81[0]	0.51[1]	0.68[1]

gral cross sections presented here are equivalent to total cross sections. This is not the case for the ground-state integral cross section because the ground-state DCS's are much less forward peaked and a larger fraction of the neutrals are scattered at angles greater than 3°. However, because the O atoms are eight times more massive than the H_2 target molecules, the maximum possible laboratory frame scattering angle is only 7.2°. Therefore, to measure ground-state total cross sections the PSD was moved as close as possible to the target cell and a larger target cell exit aperture was used. The angular acceptance of the apparatus was then such that essentially all fast O atoms were detected.

RESULTS AND DISCUSSION

In Fig. 2 the DCS's for charge-transfer scattering of O^+ ions by H_2 are shown. These data are tabulated in Tables I and II. The vertical error bars reflect statistical uncertainty only. The horizontal error bars arise from the finite primary beam size and the angular resolution used for analysis. The present total cross sections together with those measured by other workers are shown in Figs. 3 and 4. The tabulated values appear in Table III. The uncertainties in the reported total cross sections due to counting statistics, measurement of target number density and of target length are generally quite small ($\pm 3\%$). The uncertainties in the total cross sections due to the uncertainty in the ratio of the ion to neutral detection efficiencies [10,11] and the repeatability of the measurements are significantly larger. The overall uncertainty in the $O^+(^4S)$ cross sections includes a component based upon estimates of the level of contamination of the $O^+(^4S)$ oxygen ion beam by metastable ions. The overall uncertainty in the cross sections for metastable O^+ ions includes the uncertainty in the determination of the fraction of

TABLE II. Laboratory frame differential charge-transfer cross sections for $O^+(^2D,^2P)$-H_2 collisions, where E is the projectile energy and the numbers in square brackets represent powers of ten.

Laboratory angle θ (deg)	$\frac{d\sigma(\theta)}{d\Omega}$ (Å^2 sr^{-1})				
	$E=0.5$ keV	$E=0.85$ keV	$E=1.5$ keV	$E=2.8$ keV	$E=5$ keV
0.045	9.86[5]	1.07[6]	1.31[6]	1.39[6]	1.05[6]
0.090	3.90[5]	4.19[5]	3.36[5]	2.26[5]	1.21[5]
0.135	2.04[5]	1.80[5]	1.03[5]	6.31[4]	3.14[4]
0.180	1.32[5]	7.70[4]	4.52[4]	2.58[4]	1.30[4]
0.225	8.17[4]	3.83[4]	2.32[4]	1.34[4]	8.07[3]
0.270	4.63[4]	2.21[4]	1.39[4]	7.99[3]	4.41[3]
0.315	2.59[4]	1.46[4]	9.23[3]	5.30[3]	2.98[3]
0.371	1.52[4]	9.22[3]	6.07[3]	3.81[3]	2.20[3]
0.438	9.66[3]	5.78[3]	3.75[3]	2.56[3]	1.51[3]
0.506	6.71[3]	4.13[3]	2.61[3]	1.87[3]	1.08[3]
0.573	4.98[3]	3.04[3]	1.99[3]	1.40[3]	8.04[2]
0.641	3.35[3]	2.37[3]	1.44[3]	1.08[3]	6.29[2]
0.708	2.67[3]	1.88[3]	1.28[3]	9.36[2]	4.46[2]
0.776	2.18[3]	1.53[3]	9.62[2]	6.52[2]	3.88[2]
0.843	2.02[3]	1.25[3]	7.78[2]	5.16[2]	3.05[2]
0.911	1.62[3]	1.10[3]	6.82[2]	4.73[2]	2.77[2]
0.978	1.24[3]	9.23[2]	5.23[2]	4.37[2]	2.25[2]
1.05	1.08[3]	7.60[2]	5.05[2]	2.76[2]	2.02[2]
1.11	9.66[2]	6.00[2]	4.13[2]	2.93[2]	1.78[2]
1.18	7.14[2]	6.00[2]	3.62[2]	2.83[2]	1.49[2]
1.25	6.91[2]	5.51[2]	3.02[2]	1.96[2]	1.42[2]
1.32	6.68[2]	4.65[2]	3.06[2]	2.17[2]	1.42[2]
1.52	5.03[2]	3.65[2]	2.21[2]	1.46[2]	9.49[1]
1.85	3.68[2]	2.46[2]	1.55[2]	9.22[1]	6.80[1]
2.19	2.56[2]	1.66[2]	1.20[2]	5.15[1]	4.44[1]
2.53	2.01[2]	1.44[2]	9.44[1]	5.43[1]	4.61[1]
2.87	1.38[2]	1.00[2]	7.05[1]	2.69[1]	2.91[1]
3.20	1.55[2]	9.87[1]	7.23[1]	2.95[1]	2.45[1]

metastable oxygen ions in the beam.

In each frame of Fig. 2, the DCS for charge-transfer scattering of ground-state O^+ ions is compared to that for metastable O^+ ions at the indicated projectile energy. The DCS for scattering of metastable oxygen ions is, in all cases, much greater at small angles than that for ground-state ions. As with scattering of O^+ ions by N_2 [1], grazing angle collisions of ground-state ions, in which little kinetic energy is lost by the projectile, are much less likely to result in charge transfer than those involving metastable ions because the ground-state reaction has a significant energy defect, whereas the metastable reaction is essentially resonant. As one might ex-

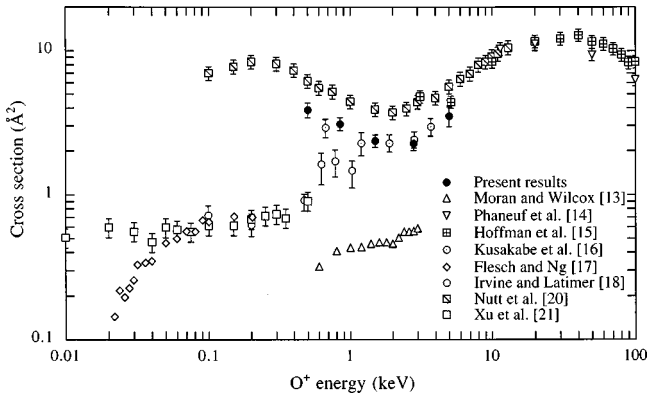

FIG. 3. Absolute $O^+(^4S)$-H_2 total charge-transfer cross sections.

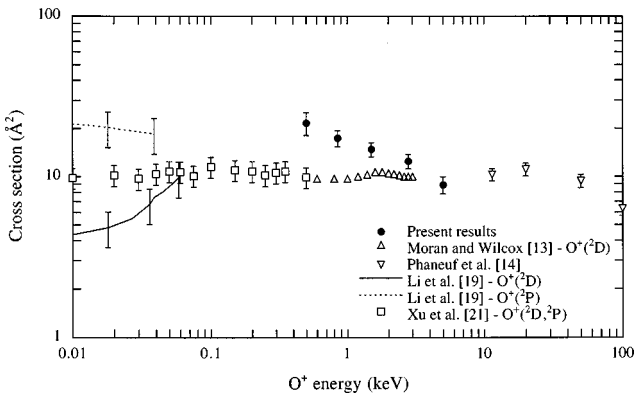

FIG. 4. Absolute $O^+(^2D,^2P)$-H_2 total charge-transfer cross sections.

TABLE III. Absolute O^+-H_2 charge-transfer cross sections. The $O^+(^4S)$ data are total cross sections. The $O^+(^2D,^2P)$ cross sections are integral over 0°–3.4°.

Projectile energy (keV)	Cross section (Å^2) $O^+(^4S)$	$O^+(^2D,^2P)$
0.50	3.9±0.5	21.6±3.5
0.85	3.1±0.3	17.4±2.0
1.5	2.4±0.2	14.8±1.5
2.8	2.3±0.2	12.5±1.3
5.0	3.5±0.5	8.9±1.1

pect, the disparity between these DCS's is most evident at 0.5 keV impact energy, and becomes systematically less pronounced as the impact energy is increased. It can also been seen from Fig. 2 that the ground-state DCS's fall sharply at angles beyond a few degrees, consistent with the maximum possible laboratory scattering angle of 7.2°. A similar effect has been observed previously in this laboratory for the case of direct scattering of O by H_2 [12]. No other experimental or theoretical data were found with which to compare the differential cross sections reported here.

A comparison with earlier measurements of the total cross sections is made in Figs. 3 and 4. The experiments fall into three broad classes; those in which the cross section is determined from measurements of the fast neutral products [13–16], those in which the slow product ions are detected [17–19], and those in which cross sections are obtained by measuring attenuation curves for the fast ion beam [20,21]. Clearly, whenever a projectile ion is neutralized in a charge-transfer collision a secondary ion results. Measurements of fast neutrals or of slow secondary ions should thus yield identical values of the charge-transfer cross section. Measurements of beam attenuation, however, may be complicated by the fact that collisions other than charge transfer may result in beam attenuation, and care must be taken to ensure that the measured attenuation is due solely to charge-transfer processes.

The various studies can also be distinguished by the particular technique used to obtain the state-specific cross section. For the case of ground-state O^+ ions, five separate methods have been employed. Moran and Wilcox [13], Hoffman, Miller, and Lockwood [15], and Kusakabe *et al.* [16] utilized controlled electron impact ion sources to produce pure $O^+(^4S)$ ground-state beams. Flesch and Ng [17] and Irvine and Latimer [18] produced their ground-state ions by dissociative photoionization of O_2. Li *et al.* [19] used a method which combined dissociative charge transfer with an rf octopole ion trap. Nutt, McCullough, and Gilbody [20] and Xu, Thomas, and Moran [21] relied on attenuation curve data alone, while in the present study the ground-state ion beam was obtained by filtering out the metastable component from a mixed state beam. For the case of metastable state O^+ ions, three general methods have been employed. Moran and Wilcox [13] and the present study measured the cross section for a mixed state ion beam and then used the fractional abundance of ground-state and metastable ions in that beam to determine the cross section for the metastable component. Xu, Thomas, and Moran [21] used the attenuation curve method. The most recent metastable measurement, that of Li *et al.* [19], used dissociative charge exchange in combination with a technique based on the rf octopole ion trap. Phaneuf, Meyer, and McKnight [14] did not use any type of state selection but, at the higher energies these investigators studied, the internal energy of the O^+ ions should have only a very slight effect on the total cross section.

It can be seen from Fig. 4 that the metastable cross-section data of Moran and Wilcox [13] and Xu, Thomas, and Moran [21] are not in good agreement with the present data. It should, however, be borne in mind that the Moran and Wilcox [13] data are probably subject to a large degree of uncertainty. Moran and Wilcox [13] state that the absolute error associated with their cross sections is approximately ±7% due to uncertainties in the absolute determination of the target gas concentration and the assessment of their neutral beam flux. However, they do not state the accuracy of the ground- and metastable-state fractions used in their work, which are crucial to the determination of the metastable cross section. As Turner, Rutherford, and Compton [22], from whom Moran and Wilcox [13] derived their ground- and metastable-state fractions, indicate that the fractions which they quote strictly apply only to the experimental configuration used in that work, it seems clear that there must be a considerable degree of uncertainty associated with the Moran and Wilcox data [13]. The data of Phaneuf, Meyer, and McKnight [14] and the data of Li *et al.* [19], in which the electronic state of the oxygen ions was well specified, are not inconsistent with the data presented here. Given the difficulties in separating the effects of ground-state ions from those of metastable ions and in ascertaining precisely which metastable species are present, the agreement between the various measurements of the cross section for the metastable ions may be viewed as tolerable although further work is needed to completely resolve the factor of 2 discrepancies which remain.

For the ground-state ions the situation is obviously far less satisfactory than that for the metastable ions. In fact more than an order of magnitude difference exists between the various measurements at energies below a few hundred eV. The present ground-state cross sections are in excellent agreement with those of Kusakabe *et al.* [16]. The data of Nutt, McCullough, and Gilbody [20] share the same energy dependence as the present results and those of Kusakabe *et al.* [16] but lie somewhat higher. During the present investigation it was observed that accurate determination of the ground-state cross section using an attenuation technique similar to that employed by Nutt, McCullough, and Gilbody [20] was extremely difficult because direct scattering contributed significantly to the observed attenuation. The agreement with Xu, Thomas, and Moran [21], Irvine and Latimer [18], and Flesch and Ng [17] is poor. The ground-state data of Moran and Wilcox [13] lie significantly lower than the present cross sections and those of all other investigators. A similar situation exists with respect to the Moran and Wilcox [13] measurements of the charge-exchange cross section of O^+ with N_2 [1].

It appears likely that the explanation for much of the discrepancy between these ground-state measurements is to be found in the differential cross-section data in Fig. 2, which show that, for $O^+(^4S)$ projectiles, a high percentage of the fast neutral products are scattered through appreciable

angles. It follows from simple kinematics that in such collisions the slow product ions must receive significant energy. Consider the case of an incoming 500 eV $O^+(^4S)$ ion. Based upon the DCS data presented here, it is estimated that 50% of the ''slow'' H_2^+ product ions must have energy in excess of 3.4 eV and 25% of the H_2^+ product ions must have energy in excess of 9.5 eV. This is a vastly different situation from that normally encountered in which the product ions formed in charge-transfer collisions have near thermal energies. Even in these, more typical, cases the total collection and mass analysis of product ions present formidable experimental difficulties; when the ions to be detected and mass analyzed have energies of several eV, the potential for incomplete collection, and thus underestimation of the cross section, is immense. Such considerations may explain the low cross sections observed by Irvine and Latimer [18], Flesch and Ng [17], and Li *et al.* [19]. The results of Xu, Thomas, and Moran [21] and Moran and Wilcox [13] are obviously not influenced by these arguments and an alternative explanation is needed.

An intriguing aspect of the $O^+(^4S)$ total cross section is the undulation clearly evident over the energy range of the present study. This behavior was first observed by Nutt, McCullough, and Gilbody [20] and has been corroborated by both Kusakabe *et al.* [16] and the present work. It has been suggested by Nutt, McCullough, and Gilbody [20], based on an analogy with the cross section for scattering of He^+ by H_2 [23], that the lower energy maximum in the ground-state cross section they report could be due to dissociative charge transfer. This analogy may not be entirely justified as the large ionization potential of He makes dissociative charge-transfer channels much more accessible than in the present case.

CONCLUSION

The absolute differential cross sections for charge-transfer scattering of 0.5-, 0.85-, 1.5-, 2.8-, and 5-keV $O^+(^4S)$ and $O^+(^2D,^2P)$ by H_2 have been determined. Below about 5 keV the ground-state cross section is much smaller than that for the metastable state and the effect of electronic excitation of the projectile on the angular distribution of the scattered neutrals is very significant. These results are of fundamental interest and have potential application in the area of tokamak plasmas and planetary aeronomy. The literature values for the ground-state total cross section are widely scattered. The ground-state data presented here are in excellent agreement with the data of Kusakabe *et al.* [16], and an explanation, based on the kinematics of the collision, is provided to explain the discrepancy between these measurements and those of some of the other investigators.

ACKNOWLEDGMENTS

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