Charge transfer in keV $O^+(^4S,^2D,^2P)$-He collisions

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Absolute differential cross sections (DCSs) are reported for charge-transfer scattering of (1–5)-keV $O^+(^4S)$ ground-state and $O^+(^2D,^2P)$ metastable-state ions by helium atoms at angles between 0.2° and 6.3° in the laboratory frame. Estimated ground-state and metastable-state total cross sections are derived from these measurements. The present ground-state cross sections agree satisfactorily with previous measurements for energies above 2 keV and the metastable-state cross sections are consistent with the mixed-state data of Kusakabe *et al.* [J. Phys. Soc. Japan **59**, 1987 (1990)]. The large differences between the ground- and metastable-state cross sections predicted by theory are not observed.

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I. INTRODUCTION

The ubiquity of charge-transfer reactions in natural and man-made plasmas is well known and these reactions are of enormous importance. Kimura *et al.* [1] have pointed out the particular significance of O^+-He charge transfer, and the reverse reaction, in environments as diverse as supernovae, the upper atmosphere, and controlled fusion plasmas [2]. The basic physics of the charge-transfer process is understood for simpler cases, but most of the more complex reactions, such as that considered here, while not less significant, are very poorly comprehended. The challenges presented to experimentalists by these complex systems often result in large disparities between the results of different workers, which ensure that even when theories are developed they cannot be tested with any degree of sensitivity.

Perhaps the single most formidable obstacle to obtaining accurate cross section measurements for O^+ projectiles is the fact that O^+ ions may be present in both the ^4S ground-state and in one of two long-lived metastable states [3] and that in collisions with atoms and molecules the behavior of the O^+ ions in these various states differs, occasionally markedly so [4]. Another issue relevant to O^+-He studies is that the cross sections are very small and measurements are therefore more prone to experimental errors. For the O^+-He system the following reactions, with corresponding asymptotic energy defects, are most probable [5]:

$$O^+(^4S)+\mathrm{He}\rightarrow O(^3P)+\mathrm{He}^+-10.96\ \mathrm{eV}, \quad (1)$$

$$O^+(^2D,^2P)+\mathrm{He}\rightarrow O(^3P)+\mathrm{He}^+-(7.64\ \mathrm{eV},5.94\ \mathrm{eV}). \quad (2)$$

Pertinent measurements have been performed by Jorgensen *et al.* [6], Kusakabe *et al.* [7], and Wolfrum, Schweinzer, and Winter [5]. Kimura *et al.* [1] have also calculated cross sections for $O^+(^4S)$ ground-state ions and for ions in both metastable states. When these studies are compared, it becomes apparent that there are primarily two outstanding issues: Kusakabe *et al.* [7] find that the metastable cross section is similar to or even greater than that for the ground-state, but Wolfrum, Schweinzer, and Winter [5] find that it is much smaller than the ground-state cross section; and Kimura *et al.* [1] calculate metastable cross sections which are much greater than appears compatible with even the data of Kusakabe *et al.* [7].

Differential cross-section (DCS) measurements and estimated total cross sections are reported here for scattering of $O^+(^4S)$ ground-state, and $O^+(^2D,^2P)$ metastable oxygen ions with He. Since it is not possible, using the techniques employed here, to differentiate between them, the two metastable cross sections reported pertain to an unspecified mixture of $O^+(^2D)$ and $O^+(^2P)$. A greater level of control over the metastable populations is certainly desirable, but as only qualitative metastable observations have been reported previously these measurements represent a significant advance.

II. APPARATUS AND EXPERIMENTAL METHOD

The apparatus, shown in Fig. 1, and the experimental method have both been described in detail previously [4]. CO is admitted to a magnetically confined plasma ion source. 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 form a beam with an angular divergence of ≈0.03°. The collimated O^+ beam passes through a short target cell and then impacts a position-sensitive detector (PSD), located 13 cm beyond the target cell. The PSD 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. An electric field estab-

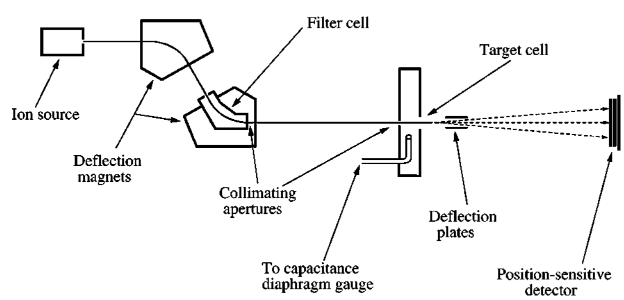

FIG. 1. Schematic of the apparatus.

lished between 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, He 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 it 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, comprises both $O^+(^4S)$ ground-state and $O^+(^2D,^2P)$ metastable ions. Cross sections for the different components of the ion beam are obtained by taking advantage of the fact that $O^+(^4S)$ ground-state ions have a much smaller charge-transfer cross section with N_2 than $O^+(^2D,^2P)$ metastable ions. In order to measure the ground-state cross section the filter cell is filled with several millitorr 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 neutral species, which because of the apparatus geometry do not enter the target cell [4]. The $O^+(^2D,^2P)$ cross section is then determined by evacuating the filter cell, measuring the effective cross section for the mixed composition beam, and subtracting the contribution these ground-state ions have made to the total scattering signal. The metastable analysis also requires knowledge of the fraction of ions in the ground and excited states, which is usually measured during the course of the experiment. However, due to the technical constraints of this study it was not possible to carry out the determination *in situ*. Instead, fraction measurements obtained earlier under similar ion source conditions were utilized. As fraction measurements have been performed many times and found to correlate reasonably well with a given set of ion source conditions, this deviation from normal procedure was adopted. It is also worth noting that the fraction measurements have been used solely to determine the fraction of excited-state O^+ ions in the beam, rather than the cross sections themselves, and any error resulting from their use in this context should be smaller than the quoted uncertainties in the data presented here.

Due to the finite angular range subtended by the detector in this type of experimental arrangement, it is not possible to collect all of the fast O atom products; the fraction that escapes detection is, however, typically quite small and the measured cross sections are essentially equivalent to total cross sections. In the present experiment, because the projectiles are four times more massive than the target atoms, the maximum possible laboratory scattering angle is only 14°. Given that the maximum possible detection angle is 6° or so, *and* that the scattering is not found to be sharply forward peaked, it is therefore clear that a significant fraction of the fast O atoms is not detected. The fraction that escapes detection is between 11% and 29%, depending upon the collision energy. Total cross sections are estimated by combining these fractions with the measured integral cross sections [4]. At the very lowest collision energy, however, it was not feasible to

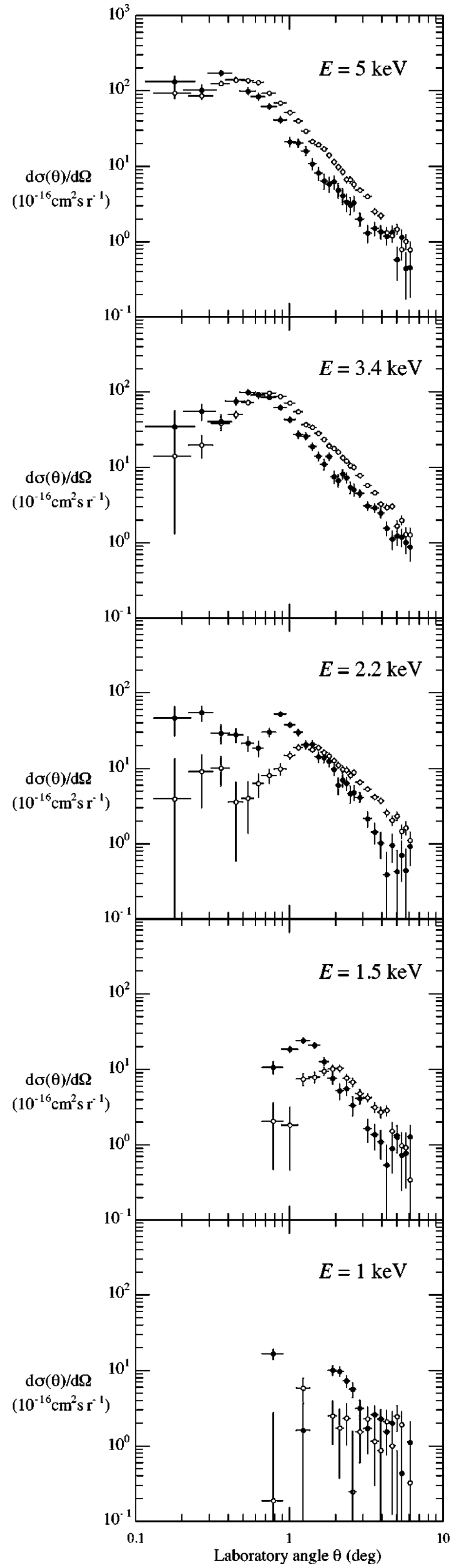

FIG. 2. Absolute differential cross sections for charge-transfer scattering of O^+ ions by He at the projectile energies indicated. $O^+(^4S)$ ground-state data are shown as open circles and $O^+(^2D,^2P)$ metastable data are shown as full circles.

TABLE I. Laboratory frame differential $O^+(^4S)$-He charge-transfer cross sections, where E is the projectile energy and the numbers in square brackets represent powers of 10.

Laboratory Angle θ (deg)	$d\sigma/d\Omega$ (10^{-16} cm^2 sr^{-1}) $E=1$ keV	$E=1.5$ keV	$E=2.2$ keV	$E=3.4$ keV	$E=5$ keV
0.27±0.05			9.1±6.0[0]	1.97±0.65[1]	8.57±0.92[1]
0.36±0.05			1.0±0.4[1]	3.82±0.51[1]	1.25±0.08[2]
0.54±0.05			4.0±2.6[0]	7.25±0.55[1]	1.37±0.07[2]
0.78±0.12	0.2±2.6[0]	2.1±1.6[0]			
0.87±0.08			9.8±1.7[0]	8.76±0.38[1]	6.94±0.33[1]
1.23±0.12	5.9±2.1[0]	7.5±1.3[0]			
1.28±0.08			2.04±0.17[1]	3.68±0.22[1]	2.94±0.18[1]
1.68±0.08		9.5±1.2[0]	1.63±0.14[1]	2.34±0.14[1]	1.71±0.12[1]
2.87±0.08	1.5±0.9[0]	4.8±0.6[0]	6.55±0.44[0]	7.83±0.41[0]	4.84±0.33[0]
3.94±0.19	0.87±0.82[0]	2.7±0.5[0]	3.71±0.31[0]	3.25±0.27[0]	2.22±0.21[0]
6.09±0.19	0.33±1.06[0]	0.35±0.54[0]	1.11±0.32[0]	1.27±0.28[0]	0.79±0.22[0]

obtain a reasonable estimate of the total cross section using this approach.

III. RESULTS AND DISCUSSION

The DCSs for charge-transfer of $O^+(^4S)$ and $O^+(^2D,^2P)$ with He are shown in Fig. 2 and selected values are tabulated in Tables I and II. Besides the statistical uncertainties shown on the graphs there are additional systematic uncertainties of ±15% and ±25% in the absolute magnitudes of the ground-state and metastable-state DCSs, respectively. The angular uncertainties arise from the finite primary beam size and the angular resolution used for analysis. It can be seen that the DCSs are not forward peaked, in sharp contrast to other differential cross-section measurements, which we have reported. This observation is consistent with the fact that the O^+-He charge-transfer process is extremely nonresonant; in fact, it is, to our knowledge, the most nonresonant reaction for which high-resolution DCSs have been obtained. The DCSs do, however, exhibit clear maxima at angles in the region of 0.5°–2°, and the angles at which these maxima occur decrease with increasing impact energy. For the most part the ground-state and metastable cross sections are fairly similar and no dramatic variation in magnitude comparable to that in O^+-N_2 [4] and O^+-H_2 [8] collisions is seen. There are, however, some obvious differences between them, particularly in the location of the maxima near 2 keV. No other experimental or theoretical data were found with which to compare these DCSs.

The present total cross sections and their associated uncertainties are tabulated in Table III. The uncertainties in the ground-state cross sections are primarily due to the uncertainty in the ratio of the ion to neutral detection efficiencies [9] and the repeatability of the measurements. The overall uncertainty in the metastable cross sections includes an additional component to account for the determination of the metastable fraction in the ion beam.

Both ground-state and metastable cross sections are quite small, which is consonant with the large energy defects in-

TABLE II. Laboratory frame differential $O^+(^2D,^2P)$-He charge-transfer cross sections, where E is the projectile energy and the numbers in square brackets represent powers of 10.

Laboratory Angle θ (deg)	$E=1$ keV	$d\sigma/d\Omega$ $E=1.5$ keV	(10^{-16} cm^2 sr^{-1}) $E=2.2$ keV	$E=3.4$ keV	$E=5.0$ keV
0.27±0.05			5.47±1.20[1]	5.54±1.33[1]	1.03±0.16[2]
0.36±0.05			2.94±0.83[1]	4.03±0.95[1]	1.72±0.14[2]
0.54±0.05			2.16±0.47[1]	9.95±0.87[1]	9.93±1.00[1]
0.78±0.12	1.66±0.27[1]	1.07±0.20[1]			
0.87±0.08			5.24±0.39[1]	6.25±0.48[1]	4.12±0.41[1]
1.23±0.12	1.6±2.0[0]	2.41±0.20[1]			
1.28±0.08			2.06±0.27[1]	2.59±0.26[1]	1.60±0.22[1]
1.68±0.08		1.27±0.16[1]	1.39±0.21[1]	1.10±0.17[1]	6.39±1.45[0]
2.87±0.08	3.14±0.89[0]	4.14±0.66[0]	4.12±0.59[0]	4.52±0.49[0]	2.01±0.40[0]
3.94±0.19	2.27±0.77[0]	1.10±0.46[0]	1.03±0.39[0]	2.48±0.33[0]	1.36±0.28[0]
6.09±0.19	1.12±0.98[0]	1.28±0.53[0]	0.93±0.41[0]	0.88±0.31[0]	0.46±0.27[0]

TABLE III. Absolute O^+-He charge-transfer cross sections. The angular range for the integral cross sections is 0°–6.3°.

Projectile Energy keV	Integral cross section (10^{-17} cm^2) $O^+(^4S)$	Integral cross section (10^{-17} cm^2) $O^+(^2D,^2P)$	Estimated total cross section (10^{-17} cm^2) $O^+(^4S)$	Estimated total cross section (10^{-17} cm^2) $O^+(^2D,^2P)$
1.0	0.44±0.07	1.14±0.28		
1.5	1.04±0.16	1.16±0.29	1.47±0.48	1.45±0.51
2.2	1.81±0.27	1.41±0.35	2.48±0.77	1.59±0.56
3.4	3.07±0.46	1.70±0.43	3.78±0.91	2.14±0.75
5.0	2.39±0.36	1.26±0.32	2.78±0.57	1.47±0.51

volved in the O^+-He collision system. However, the concept of energy defects cannot easily explain why the ground-state cross section is greater than that for the metastable ions because the ground-state reaction is associated with a larger energy defect than the metastable reaction [Eqs. (1) and (2)] and might therefore be expected to have a smaller cross section than that for the metastable ions. This is most definitely not borne out by the experiment and clearly indicates the limitations of using the asymptotic energy defects in this way. Consideration of the DCSs, however, does suggests a reasonable physical explanation as to why the simple energy defect model should break down for O^+-He even though it has been quite successful in more than a few other instances [4,8,10]. From the DCSs it can be seen that, on average, the O atoms are scattered at large angles indicating a very intimate collision. This is, of course, consistent with the requirement that significant kinetic energy be made available to the collision partners for this very endothermic charge-transfer reaction to take place. Under such conditions, the interaction clearly cannot be approximated by the glancing collision model often adopted and, in this case, the asymptotic energies thus cease to have an obvious direct correlation with the collision event.

The three prior experimental studies [5–7] and the calculation by Kimura *et al.* [1] are compared to the present data in Fig. 3. Of these the earliest is that of Jorgensen *et al.* [6] who report cross sections in the (50–400)-keV range. These workers did not use any type of state selection but at such high energies internal energy effects should be minimal and their data are shown for comparison on both ground- and metastable-state plots. Consideration of the ground-state data [Fig. 3(a)] reveals that, although there is considerable scatter between the measurements, they are all consistent with each other above 2 keV. The present data and those of Kusakabe *et al.* [7] seem to deviate from one another below 2 keV, especially when the 1-keV integral value (Table III) is considered. This is perhaps not too surprising given that the cross section in this region is very small and that these are the lowest energies at which either apparatus is capable of operating.

The metastable measurements are shown in Fig. 3(b) and the present data are again seen to be in accord with those of Jorgensen *et al.* [6]. While there are no other metastable measurements available, qualitative observations have been reported by Kusakabe *et al.* [7] and Wolfrum, Schweinzer, and Winter [5]. Kusakabe *et al.* [7] found the mixed-state cross section to be similar to the ground-state cross section above 2 keV or so, from which it may be concluded that the metastable cross section is similar to the ground-state cross section at these energies. They also found the mixed-state cross section to be greater than the ground-state cross section at lower energies, indicating that the metastable cross section is significantly larger than the ground-state cross section below 2 keV. By contrast, Wolfrum, Schweinzer, and Winter [5] found the metastable cross section, in the (2–6)-keV range, to be so much smaller than the ground-state cross section that they were unable to measure it and they then postulated the idea of suppressed electron capture in $O^+(^2D,^2P)$-He collisions to explain their observations. The qualitative observations of Kusakabe *et al.* [7] firmly support our findings that the metastable cross section is of similar

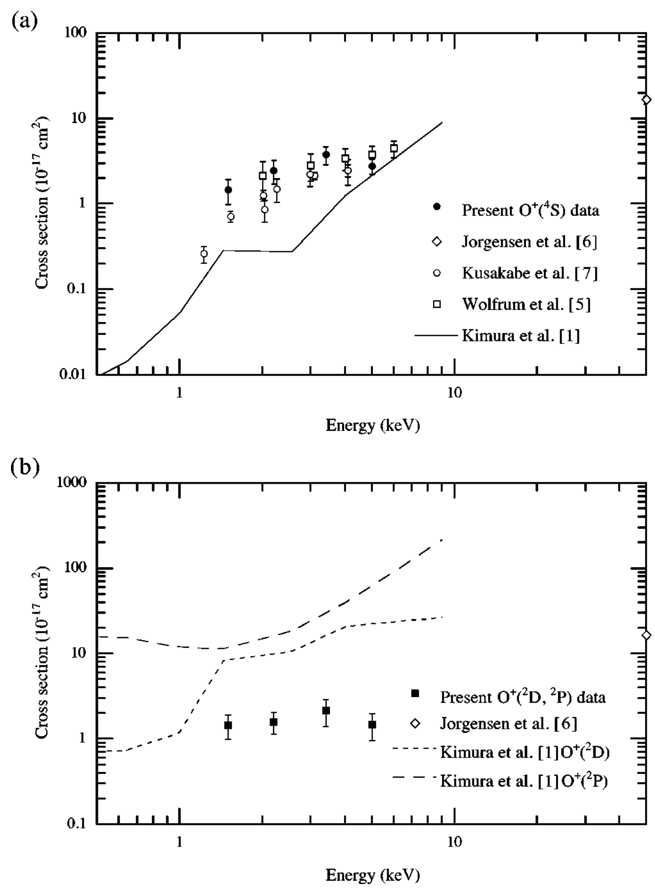

FIG. 3. O^+-He total charge-transfer cross sections: (a) $O^+(^4S)$-He and (b) $O^+(^2D,^2P)$-He. Note that only the lowest-energy measurement of Jorgensen *et al.* [6] is shown.

magnitude to the ground-state cross section. Furthermore, Kimura *et al.* [11] have suggested a plausible explanation as to why Wolfrum, Schweinzer, and Winter [5] found the metastable cross section to be much smaller than it actually is.

The calculations of Kimura *et al.* [1], obtained using a semiclassical molecular representation, are seen to underestimate the ground-state cross section below a few keV by as much as a factor of 5. Their calculated energy dependence also seems more pronounced than the measurements would indicate, especially when the data of Jorgensen *et al.* are considered. Comparison of the experimental and theoretical metastable data is a little more problematic because Kimura *et al.* [1] were able to calculate cross sections for each individual metastable state, whereas the experimental data pertain to an undetermined mix of $O^+(^2D)$ and $O^+(^2P)$ ions. Both calculated metastable cross sections are therefore shown and it is evident that, even if all of the metastable ions in our beam are assumed to be in the ^2D state, theory overestimates the cross section by a large factor. The calculations predict the $O^+(^2D)$ cross section to be approximately 40 times greater than the $O^+(^4S)$ cross section, whereas both the present study and that of Kusakabe *et al.* [7] indicate that the two cross sections are comparable. No explanation is offered as to why the calculations do not achieve a reasonable physical description of O^+-He collisions other than to note that Kimura *et al.* [1] considered the treatment of this system particularly challenging because of the open-shell, multielectron nature of oxygen and the correspondingly complex electronic structure of the collision system.

IV. CONCLUSION

Absolute differential cross sections (DCSs) are reported for charge-transfer scattering of (1–5)-keV O^+ ions by He at angles between 0.2° and 6.3° in the laboratory frame. Estimated total charge-transfer cross sections are derived from these measurements and are compared with previously published data. Cross sections for both $O^+(^4S)$ ground-state and $O^+(^2D,^2P)$ metastable ions are presented. The present ground-state cross sections are consistent with previous experimental data above 2 keV. The metastable-state cross sections are found to be only slightly smaller than those for the ground-state over most of the energy range studied. The large differences between the behavior of ground- and metastable-state ions predicted by theory are not observed.

It should be noted that additional work is needed on these reactions. The work presented here clearly advances our understanding of them but the uncertainties attributed to the data are undesirably high and furthermore the role of the different metastable states must be more fully understood.

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