

Collisions of kilo-electron-volt H^+ and He^+ with molecules at small angles: Absolute differential cross sections for charge transfer

R. S. Gao, L. K. Johnson, C. L. Hakes, K. A. Smith, and R. F. Stebbings

*Department of Physics, Department of Space Physics and Astronomy, and Rice Quantum Institute, Rice University,
P.O. Box 1892, Houston, Texas 77251*

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Absolute differential cross sections for charge-transfer scattering of H^+ and He^+ from molecules are reported over the laboratory angular range 0.02° – 1.0° . Cross sections have been determined at 0.5, 1.5, and 5.0 keV for collisions of H^+ with N_2 and O_2 , and at 1.5 keV for collisions of H^+ with CO, CO_2 , NO, and CH_4 , and of He^+ with H_2 , N_2 , O_2 , CO, and NO. The data exhibit considerable structure over the experimental angular range. Absolute integral cross sections from 0° to 1.0° have been obtained and are compared with the published total cross sections.

INTRODUCTION

This paper reports measurements of differential cross sections for charge transfer in collisions of H^+ and He^+ with several molecules. Differential scattering in ion-molecule collisions has been the subject of considerable experimental effort. In particular, at the University of Connecticut, collisions of H^+ and He^+ with molecules have been studied at laboratory angles of 2.5° , 3° , and 5° in the energy range 1–200 keV as a function of final charge state by Ziemba *et al.*;¹ while collisions of He^+ with H_2 and N_2 and of H^+ with N_2 at keV energies have been investigated by Pollack and co-workers.^{2–5} Collisions of He^+ with H_2 , N_2 , O_2 , CO, and NO and collisions of H^+ with N_2 and CO at keV energies have been studied by Barat and co-workers at the Centre National de la Recherche Scientifique (CNRS) at Orsay, France.^{6–9} Pollack and co-workers^{2–5} and Barat and co-workers^{6–9} measured relative differential cross sections over the range 0° – 3° , and both groups measured energy loss in order to obtain information on the reaction channels. The angular resolution in their experiments was about 0.3° , and consequently structure at scattering angles below about 1° was not completely revealed.

Previous papers originating in our laboratory described techniques for measuring keV-energy absolute differential cross sections for heavy-particle scattering at very small angles with high angular resolution. Differential cross sections for scattering of H and He projectiles by a variety of rare-gas and diatomic molecular targets were reported,¹⁰ as were direct-scattering and charge-transfer data for collisions of H^+ and He^+ with the rare gases.¹¹ The angular range of these published measurements was typically $0.02^\circ \leq \theta \leq 1^\circ$, and at small angles the angular resolution was about 0.02° . These differential cross sections have been used to evaluate theoretical models and interaction potentials.^{10,11} In addition, integration of the differential cross section over angle has been carried out to provide integral scattering cross sections which, since the scattering is generally strongly peaked in the forward direction, represent a large fraction of the total cross section.

In this paper, differential cross sections for charge-transfer scattering of H^+ by N_2 and O_2 at 0.5, 1.5, and 5.0 keV, for 1.5 keV H^+ collisions with CO, CO_2 , NO, and CH_4 and He^+ collisions with H_2 , N_2 , O_2 , CO, and NO are reported.

EXPERIMENTAL METHOD

The apparatus, shown in Fig. 1, and the experimental method have been described fully in previous publications,^{10,11} and only a brief description is given here. Ions emerging from the source are accelerated to the desired energy and focused electrostatically. The resulting ion beam is momentum analyzed by two sector magnets and the mass-selected beam of H^+ or He^+ passes through a collimating aperture before arriving at the target cell (TC), which is approximately 0.4 cm long. The entrance aperture of the TC (30 μm diameter) and the collimating aperture (20 μm diameter) are separated by 49 cm, limiting the angular divergence in the beam at 0.003° . A 4.0-cm-diam. position-sensitive detector (PSD) located at 109 cm from the TC monitors both the primary beam and fast collision products. An electrostatic field may be established between deflection plates (DP's) to prevent primary and scattered ions from striking the detector. A Digital Equipment Company LSI 11/2 microcomputer sorts the arrival coordinates of each detected particle into bins in a 90×90 array. The minimum physical bin size for the present experiments is $109 \times 109 \mu\text{m}^2$. Under the thin-target conditions used in this experiment, the differential cross section is determined from the measured quantities according to the relation

$$\frac{d\sigma(\theta)}{d\Omega} = \frac{\Delta S(\theta)}{SnL\Delta\Omega},$$

where S is the primary ion-beam flux in particles per second, $\Delta S(\theta)$ is the neutral flux scattered at angle θ into a solid angle $\Delta\Omega$ sr, n is the target density determined by a measurement of the gas pressure in the TC, and L is the target cell length.¹⁰ For the present geometry, the product nL has been found to be an accurate representation of the target thickness.

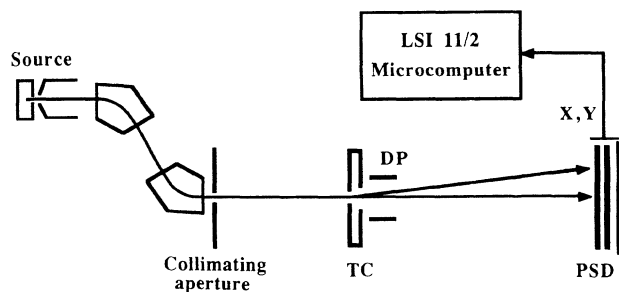
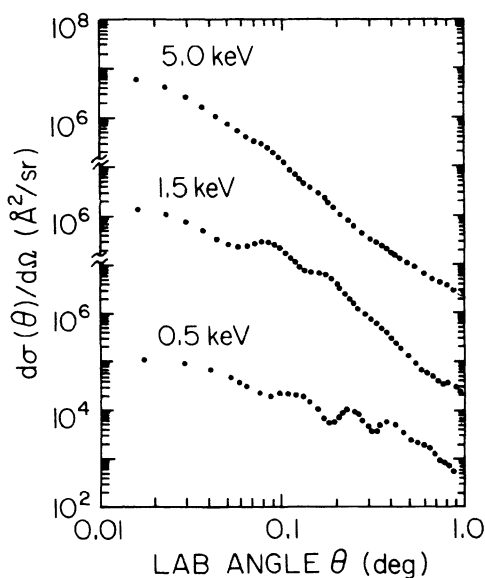
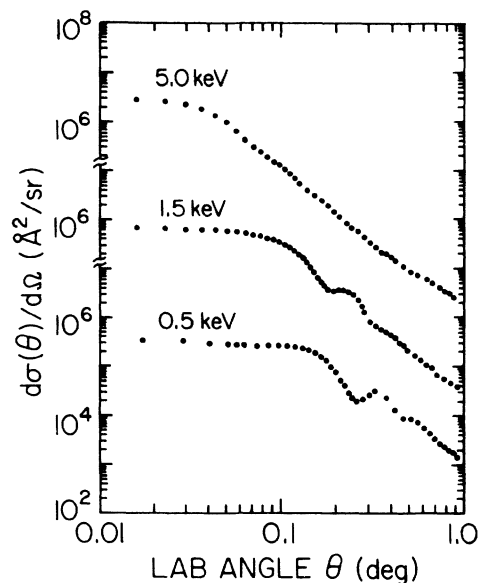


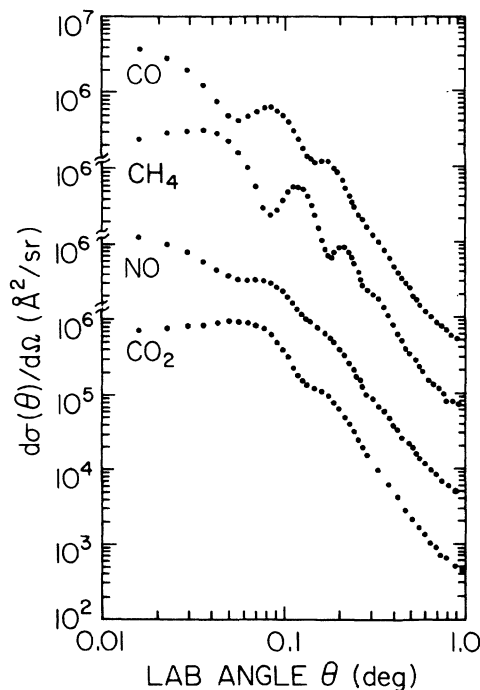
FIG. 1. Schematic of the apparatus.

The data acquisition procedures have been described in detail in previous publications,¹⁰ and are only outlined here. Two data files consisting of 90×90 arrays of particle position data, one with gas in the TC and one without, are accumulated. The scattered flux $\Delta S(\theta)$ at each scattering angle is obtained by partitioning the 90×90 arrays into rings concentric with the beam center, summing the counts in each ring individually, and then subtracting the ring sums for gas-out data from those for gas-in data. The experimental uncertainty in the number of counts at a particular angle is primarily statistical, and ranges from 1% near 0.02° to 10% near 1° . The angular uncertainty arises from the finite width of the primary ion beam, the discrete width of the analysis rings, and electronic errors in the detector's position encoding circuits. The collective uncertainty amounts to about 0.03° at the smallest scattering angles. The issue of possibly different detection efficiencies of the PSD for neutral and charged species has been discussed previously¹¹ with the conclusion that at 5.0 keV the two efficiencies are considered equal, and at 1.5 and 0.5 keV the uncertainties are $\pm 5\%$ and $\pm 10\%$, respectively.

FIG. 2. Differential cross sections for charge-transfer scattering of H^+-N_2 at 5.0, 1.5, and 0.5 keV. Note the shift of the Y axes.FIG. 3. Differential cross sections for charge-transfer scattering of H^+-O_2 at 5.0, 1.5, and 0.5 keV. Note the shift of the Y axes.

RESULTS AND DISCUSSION

Differential cross sections for charge transfer are shown in Figs. 2–6. The present apparatus distinguishes between charge states, and not between electronic states; and so no information about the electronic states of the reaction products is obtained. The reactants in the

FIG. 4. Differential cross sections for charge-transfer scattering of H^+-CO , CH_4 , NO , and CO_2 at 1.5 keV. Note the shift of the Y axes.

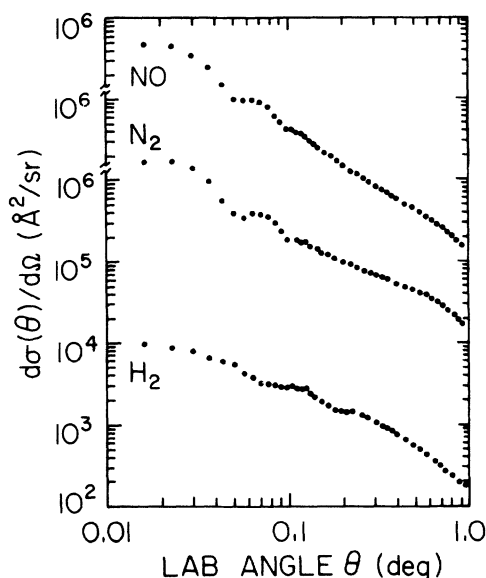
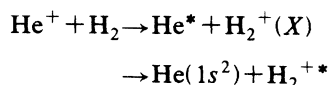
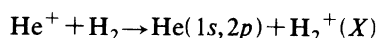


FIG. 5. Differential cross sections for charge-transfer scattering of He^+ -NO, N_2 , and H_2 at 1.5 keV. Note the shift of the Y axes.

present work are assumed to be initially in their ground states. For He^+ -molecule collisions, the work by Pollack and co-workers²⁻⁴ and Barat and co-workers⁶⁻⁸ provides valuable information about the final states. For the He^+ - H_2 reaction, they found^{4,7} that at reduced scattering angles $\tau < 2$ keV deg, there are several channels open to the reaction, of which both



are important. As τ increases, the reaction



becomes dominant. For the He^+ - N_2 reaction, the near-

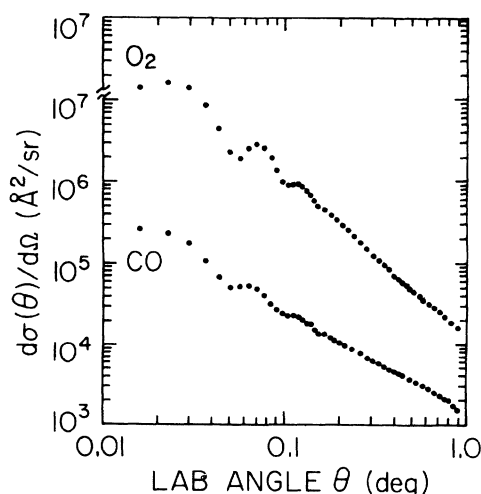
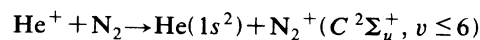


FIG. 6. Differential cross sections for charge-transfer scattering of H^+ - O_2 , and CO at 1.5 keV. Note the shift of the Y axes.

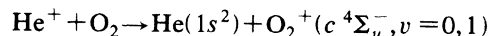
resonant channels



are always open at keV energies.^{4,6} At $\tau \geq 0.2$ keV deg, there are several competing channels,⁶ including

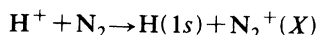


The dominant channel for He^+ - O_2 at $\tau \leq 1$ keV deg has been identified as^{6,12}

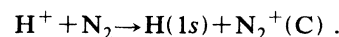


followed by dissociation. For higher τ , a variety of final states is produced. A rich spectrum of final states has also been observed⁷ for both He^+ -CO and He^+ -NO collisions, but little definite state identification has been done. Interested readers will find more discussion in Refs. 4-8.

The only charge transfer measurements of H^+ -molecule collisions in which both scattering angle and energy loss are determined are those of Quintana, Andriamasy, and Pollack⁵ for N^+ - N_2 . They found that at 1 keV the dominant charge-transfer process is



and there is some small contribution from process



The integral cross sections from 0° to 1.0° are obtained using the formula

$$\sigma_{0-1^\circ} = \Delta S / S n l,$$

where ΔS is the sum of all neutral particle signal scattered by less than 1° , and are listed in Table I. While it is not possible to reliably extrapolate the present data to larger angles for the purposes of determining total cross sections, it is nevertheless illustrative to compare the present integral cross sections with total cross sections determined elsewhere. This comparison is made in Table I. For H^+ projectiles the present integral cross sections agree well with published total cross sections. The good agreement is attributed to the small energy defects for these reactions which enable the reactions to occur at large impact parameters. The differential charge-transfer cross sections are consequently so strongly peaked in the forward direction that a large fraction of the total cross section is contained in the laboratory angular range below 1° . For He^+ projectiles, good agreement between our integral cross sections and total cross sections measured by other groups is found only for He^+ - O_2 which is known to be near resonant.⁶ For the other He^+ -molecule reactions, the integral cross sections are smaller than the total cross sections. This result is viewed as a consequence of the nonresonant nature of these processes. The results of Pollack and co-workers and Barat and co-workers show that the dominant channels for these reactions are nonresonant and are enhanced at large scattering angles. The present absolute differential cross sections at 0.1° and 0.8° are given in Table II as an aid to other investigators.

All the differential cross sections shown in Figs. 2-6

TABLE I. Absolute integral cross sections in \AA^2 , $0^\circ \leq \theta \leq 1.0^\circ$, in the laboratory frame and total cross sections in \AA^2 .

Process	Projectile energy (keV)	Integral cross section	Total cross section
$\text{H}^+ - \text{N}_2$	0.5	2.5	2.6 ^a
	1.5	8.1	10 ^b
	5.0	11	12, ^c 13, ^d 18.5 ^e
$\text{H}^+ - \text{O}_2$	0.5	12	16 ^a
	1.5	9.5	
	5.0	9.0	9.4, ^c 11 ^e
$\text{H}^+ - \text{CO}$	1.5	12	14 ^f
$\text{H}^+ - \text{CO}_2$	1.5	14	13 ^f
$\text{H}^+ - \text{NO}$	1.5	7.3	
$\text{H}^+ - \text{CH}_4$	1.5	23	23 ^g , 19 ^f
$\text{He}^+ - \text{H}_2$	1.5	0.49	2.1 ^h
$\text{He}^+ - \text{N}_2$	1.5	3.7	8.7, ^h 10 ⁱ
$\text{He}^+ - \text{O}_2$	1.5	8.3	11 ^h , 10 ⁱ
$\text{He}^+ - \text{CO}$	1.5	3.4	6.5 ^h , 4.6 ^j
$\text{He}^+ - \text{NO}$	1.5	4.8	8.7 ^h

^aKoopman (Ref. 15). ^bGordeev and Panov (Ref. 16). ^cStier and Barnett (Ref. 17). ^dMcNeal and Clark (Ref. 18). ^eRudd *et al.* (Ref. 19). ^fMcNeal (Ref. 20). ^gEliot (Ref. 21). ^hMoran and Conrads (Ref. 22). ⁱStebbing, Smith, and Ehrhardt (Ref. 12). ^jCoplan and Ogilvie (Ref. 23).

exhibit oscillatory structure. One type of structure results from the fact that transitions between the initial and final states of the system may occur as the reactants approach or recede from each other and the scattering amplitudes corresponding to the different reaction pathways interfere. Landau-Zener¹³ (curve crossing) or Demkov¹⁴ (noncrossing) models of this phenomenon assume that transitions occur over a well-localized range of internuclear separations. Structure in the cross sections can also arise from quantum or classical effects involving the at-

tractive and repulsive parts of the potential surface(s) relevant to the collision, such as diffraction or rainbow scattering. Such effects have been observed in this laboratory for other ion-neutral processes. The evidence presented in Refs. 2–9 shows clearly that in charge-transfer reactions of H^+ and He^+ with simple molecules, that there can be a multiplicity of final states. Because several final states are accessible, it is in some respects surprising that the well-defined oscillations are observed at all. One should note, however, that the observed oscil-

TABLE II. Values of the differential cross sections in $\text{\AA}^2/\text{sr}$ at the indicated laboratory angles. The statistical uncertainty in the values is 5% at 0.1° and 10% at 0.8° .

Process	Projectile energy (keV)	$d\sigma(\theta=0.1^\circ)$ $d\Omega$	$d\sigma(\theta=0.8^\circ)$ $d\Omega$
$\text{H}^+ - \text{N}_2$	0.5	2.1×10^4	8.0×10^2
	1.5	2.1×10^5	3.6×10^2
	5.0	1.4×10^5	3.8×10^2
$\text{H}^+ - \text{O}_2$	0.5	2.6×10^5	2.2×10^3
	1.5	3.3×10^5	5.5×10^2
	5.0	1.2×10^5	3.5×10^2
$\text{H}^+ - \text{CO}$	1.5	4.6×10^5	6.0×10^2
$\text{H}^+ - \text{CO}_2$	1.5	3.6×10^5	6.6×10^2
$\text{H}^+ - \text{NO}$	1.5	2.3×10^5	6.4×10^2
$\text{H}^+ - \text{CH}_4$	1.5	3.9×10^5	7.5×10^2
$\text{He}^+ - \text{H}_2$	1.5	2.8×10^3	2.4×10^2
$\text{He}^+ - \text{N}_2$	1.5	1.8×10^4	2.4×10^3
$\text{He}^+ - \text{O}_2$	1.5	9.4×10^4	2.1×10^3
$\text{He}^+ - \text{CO}$	1.5	2.4×10^4	2.0×10^3
$\text{He}^+ - \text{NO}$	1.5	4.2×10^4	2.1×10^3

lations are most pronounced at the smaller scattering angles where the quasis resonant channels are likely to dominate, thus restricting the number of final states available. In addition, the structure is much less apparent at higher collision energies and in cases such as He^+-H_2 where there is no dominant quasis resonant channel.

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