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Charge Transfer in Collisions between Diatomic Molecular Ions and Atomic Hydrogen using Merged Beams

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Abstract. A merged-beams technique is used to measure charge transfer (CT) cross sections for the (O_2^+ , D) and (CO^+ , D) systems over a wide range of collision energy from 20 eV/u to 2 keV/u. At the higher energies where the collision is ro-vibrationally frozen and the differences in the Q-values of the CT process can be neglected, the cross sections all converge to $(7.5 \pm 0.5) \times 10^{-16} \text{ cm}^2$ at 2 keV/u and are consistent with a ro-vibrational frozen (H_2^+ , H) calculation. Toward lower velocities, (O_2^+ , D) and (CO^+ , D) have consistently similar cross sections but diverge below 60 eV/u. In contrast, previously reported merged-beams measurements for (D_2^+ , H), a system with fewer electrons on the molecular core, no electronic excited states and relatively fewer charge transfer channels, shows a decreasing cross section toward lower energies. These different trends are compared to previous merged-beams measurements of charge transfer with H for several atomic 4+ ions (Si^{4+} , Ne^{4+} , N^{4+} and C^{4+}) which have a variety of electrons on the core.

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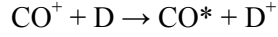
I. INTRODUCTION

Knowledge of charge transfer (CT) for molecular ion-neutral collisions is necessary for the modeling of high density-low temperature plasmas as can be found in cold divertor regions of a fusion tokamak. Accurate cross sections of the entire spectrum of excited molecules is necessary for understanding the formation of the detachment layer desired for the reduction of heat loads on divertor plates [1]. CT with an atomic hydrogen-target is of special interest because of its fundamental aspect and its importance in interstellar clouds. In the low density of the interstellar medium including dense clouds, the majority of interstellar molecules are formed by ion-neutral reactions including charge transfer processes efficient at low temperatures (UMIST database) [2].

Measurements of molecular ion charge transfer with atomic H is a particular experimental challenge. An H target can be difficult to produce and characterize resulting in few measurements at limited collision energies. There have been ion-flow drift tube studies where the amount of atomic H was estimated by observing the COH^+ products from the introduction of CO_2^+ . In this way, for example, the O^+ , CO^+ , and CH^+ reaction rates with H have been measured at 300 K [3]. For the fundamental system $\text{H}_2^+ + \text{H}$, which has been studied using a variety of detailed theoretical approaches [1,4-6], there is surprisingly little experimental measurement for comparison. Using a crossed-beam technique where a thermal energy hydrogen beam was produced using a tungsten tube furnace, McGraph et al. [7] performed one of the first experimental investigations on the $\text{H}_2^+ + \text{H}$ collision system in the energy range between 10 keV and 50 keV. Their results indicate that the dissociative and non-dissociative CT channels dominate collision reactions toward lower energies. These measured cross sections for dissociative and non-dissociative CT in $\text{H}_2^+ + \text{H}$ at the higher energies are also consistent but yet significantly below the sudden approximation calculations [6]. At the lower energies where the ro-vibrational states become important theory becomes difficult. A fully quantal state-to-state calculation [4] has been performed for collision energies from 0.2 eV/u to 10 eV/u and extended [5] up to 100 eV/u using a semiclassical straight-line trajectory approximation for the projectile motion. Tabulated results for both calculations are available on the Oak Ridge National Laboratory' (ORNL) Controlled Fusion Atomic Data Center (CFADC) website [5]. While ORNL merged-beams measurements are now in progress for CT in $\text{H}_2^+ + \text{H}$ collisions, a detailed comparison has previously been made for $\text{D}_2^+ + \text{H}$ from 0.1 eV/u to 2 keV/u [8]. This first molecular ion measurement with the merged-beams apparatus benchmarks the sudden approximation theory appropriate at the higher energies and the vibrational specific adiabatic theory for the $(\text{H}_2\text{-H})^+$ complex at the lower energies. ORNL CT merged-beams measurements have also been reported for $\text{D}_3^+ + \text{H}$ from 2 eV/u to 2 keV/u [9].

In this paper, we report the measurements of the cross section for (O_2^+, D) and (CO^+, D) over an extended energy range from 20 eV/u to 2 keV/u using the ORNL ion-atom merged-beams apparatus and

compare to our previous results for the fundamental (D_2^+ , H) system [8] . Note, for the measured charge transfer reaction, e.g. (CO^+ , D),



we only measure the D^+ product of the reaction. Therefore our measurements do not distinguish between dissociative and non-dissociative channels of the product CO^* . Dissociation may occur if energetically possible. Ionization of D is not expected at these low collision energies.

A succinct description of the upgraded ORNL ion-atom merged-beams apparatus is given in Section II of this paper. In Section III, our measurements are compared with available theories at higher energies where the ro-vibrational states can be considered frozen and at lower energies where the collision times are comparable to characteristic ro-vibrations. Our conclusion is drawn in section IV.

II. DESCRIPTION OF THE EXPERIMENT

The Multi-charged Ion Research Facility (MIRF) ion-atom merged-beams apparatus at ORNL has been successful for many years [10-13] in producing absolute CT cross sections for a variety of atomic ions on H and D. More recently with the development of intense molecular ion-beams produced by an Electron Cyclotron Resonance (ECR) ion source [14], the apparatus has been used for molecular ion studies [8,9]. The description of MIRF ion-atom merged-beams apparatus upgraded to accept beams from the CEA/Grenoble all-permanent magnet ECR ion source mounted on a 250 kV High Voltage (HV) platform has been detailed elsewhere [15]. Only a brief description is given here. A schematic of the current apparatus is shown in Fig. 1. In the merged-beams technique, fast (keV) atomic or molecular ion beams are merged with H or D neutral beams producing center-of-mass collision energies from meV/u to keV/u. The ion beam accelerated up to 150 kV from the HV platform is merged using electrostatic spherical deflectors with the neutral ground-state H or D beam. For the O_2^+ and CO^+ measurements reported here, deuterium was used to obtain the slower velocities needed for velocity matching with the relatively heavy singly charged molecular ions. For example, for the measurements with $O_2^+ + D$, the 9.1

keV D beam is merged with a 16.7 keV to 125 keV/u O_2^+ beam resulting in collisions in the center-of-mass from 1966 eV/u to 22 eV/u, respectively. Also, for the CO^+ measurements, CO (M=29) enriched with ^{13}C was used in the source to avoid contamination with any N_2^+ (M=28) ions which may be present.

The D beam is obtained by photodetachment of a 9.1 keV D^- beam as it crosses the optical cavity of a 1.06 μm 100W CW Nd:YAG laser, where kilowatts of continuous power circulate. The voltage cell labeled in Fig.1 can be used to fine tune the velocity of the D^- beam at meV/u center-of-mass collision energies. However, for these measurements at eV/u center-of-mass energies and above, the cell is not used and is grounded. The 9.1 keV D^- beam is produced by a commercial duoplasmatron ion source with mass selection by a 30° bending magnet. After photodetachment surviving D^- ions are removed from the neutral D beam using electrostatic deflection. The ion and neutral beams interact along a field free merge-path, after which D^+ product ions are magnetically separated from the primary beams. An electrostatic deflector produces the final deflection out of the plane of magnetic dispersion (see Figure 1). The product signal D^+ ions are detected with a channel electron multiplier (CEM) operated in pulse counting mode. The beam-beam signal rate (Hz) is extracted from (kHz) backgrounds with a two-beam modulation technique. D^+ backgrounds come from D stripping on background gas in the merge path. Vacuum pressure in the merge-path is kept around 4×10^{-10} Torr with beams.

The independently absolute cross section is calculated from directly measurable parameters including the D^+ product count rate, the measured overlap of the two beams, the velocities of the ion and neutral beams, and the beam intensities. The velocities of the beams are calculated from the acceleration potentials including estimated plasma shifts in the ion sources [see e.g., 11]. The overlap of the two beams is measured from profiles obtained using two mechanical slits (labeled as Slit Scanner M1 and M2 in Fig. 1) and one dual rotating wire scanner [16]. The neutral beam intensity is measured by using secondary electron emission from a stainless steel plate; the secondary emission coefficient is calibrated in situ [17]. While merged-beam measurements typically can access meV/u to keV/u collision energy, the measurements here were limited. Our measurements for CO^+ and O_2^+ could not extend below 20

eV/u due to the requirement that the signal D^+ at 9.1 keV has to be deflected to 62.5° by a magnetic field which is not always strong enough to deflect the entire O_2^+ or CO^+ primary beam into the de-merge Faraday cup. This is a problem for the higher molecular ion velocities needed to access lower center-of-mass collision energies.

Although the ECR source produces molecular ions by energetic electron impact in diffuse plasma, there is no direct coupling between the power injected into the source and the internal energies of the ions themselves. These molecular ions are produced with certain vibrational and rotational excitations. We know of no investigation of the electronic states of ECR produced molecular ions. However, the distribution of the ro-vibrational states should be similar to the ones produced by conventional electron impact ion sources which have been observed to be determined by Frank-Condon transitions between the electronic ground states of D_2 and D_2^+ , O_2 and O_2^+ , CO and CO^+ [18-23]. In addition to the electronic ground state $X^2\Sigma^+$, calculations [24] show that CO^+ excited states $A^2\Pi$ and $B^2\Sigma^+$ can also be formed by the electron impact. The excited state $CO^+(B^2\Sigma^+)$ with a measured lifetime of 45 ns [25,26] is not expected to survive the 5-15 μs time-of-flight to the merged-beams apparatus. However, some excited $CO^+(A^2\Pi)$ with a measured lifetime of several microseconds [27] might be present during measurements. For O_2^+ , Wim et al. [20] have observed the production of $O_2^+(a^2\Pi_u)$ metastable in a conventional electron impact ion source, for which the electronic ground state is $O_2^+(X^2\Pi_g)$. The amount of $CO^+(A^2\Pi)$ or $O_2^+(a^2\Pi_u)$ decreases with the electron impact energy and is not significant if the electron energy is below the required energy to ionize CO or O_2 to these electronically excited states.

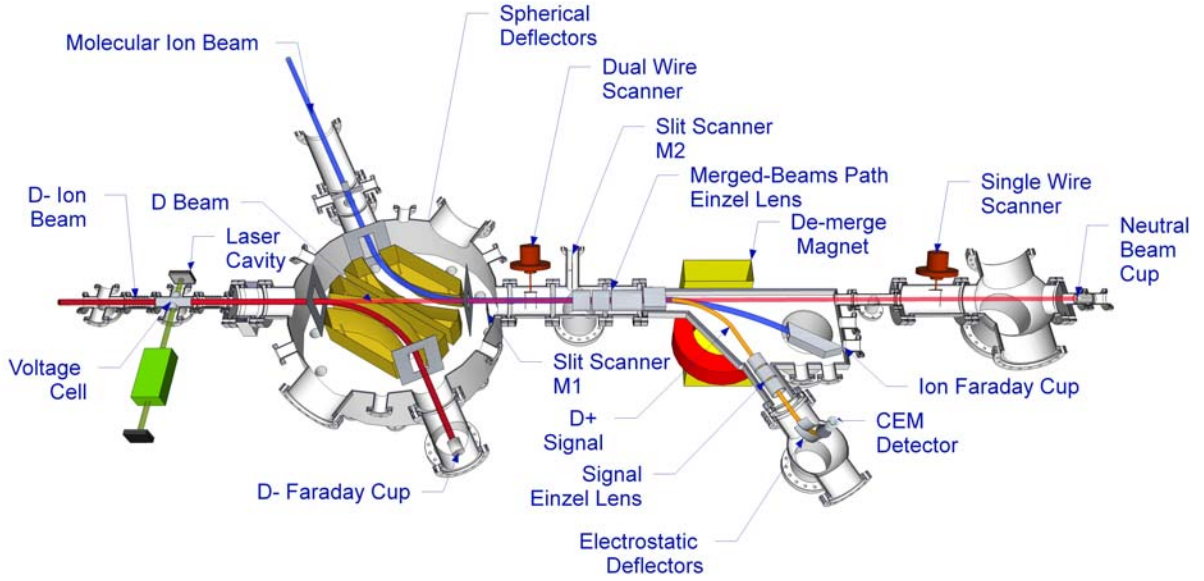


Figure 1. “Color online” Schematic of the current ORNL ion-atom merged-beams apparatus. The merge-path (32.5 cm) begins at the exit of the electrostatic spherical deflectors and ends in an Einzel lens just before the de-merge magnet. A discussion of the various elements can be found in the text.

III. RESULTS AND DISCUSSION

Our results for (CO^+, D) and (O_2^+, D) are presented in Table 1 and 2, respectively, along with relative and absolute errors at a 90% confidence level [10]. Figure 2 shows our measurements for CO^+ and O_2^+ in comparison with our previous $\text{D}_2^+ + \text{H}$ measurements and various theories for $\text{H}_2^+ + \text{H}$ and $\text{CO}^+ + \text{H}$. Previous experimental and theoretical studies of CT with O_2^+ in the literature include Cs, H_2 , and O_2 targets [24, 32-34], but we know of no CT study for $\text{O}_2^+ + \text{H}$ or isotopic D. Preliminary measurements for $\text{CO}^+ + \text{D}$ were reported in conference proceedings [9].

Table 1. Measured charge transfer cross sections for $\text{CO}^+ + \text{D}$. Relative and statistical errors are listed below and correspond to a 90% confidence level.

Energy (eV/u)	Cross Section (10^{-16} cm^2)	Relative Error (10^{-16} cm^2)	Absolute error (10^{-16} cm^2)
19.8	2.0	0.2	0.3
30.7	2.5	0.3	0.4
41.3	4.1	0.4	0.6
48.5	5.2	0.3	0.7
70.7	5.7	0.5	0.9
98.0	5.9	0.3	0.8
131	6.5	0.3	0.9
169	6.7	0.5	1.0
213	6.9	0.4	0.9
324	6.7	0.3	1.0
459	6.9	0.3	0.9
657	7.5	0.4	1.0
900	8.0	0.4	1.0
1224	7.8	0.4	1.0
1672	7.7	0.4	1.0
1923	7.9	0.4	1.0

Table 2. Measured charge transfer cross sections for $\text{O}_2^+ + \text{D}$. Relative and statistical errors are listed below and correspond to a 90% confidence level.

Energy (eV/u)	Cross Section (10^{-16} cm^2)	Relative Error (10^{-16} cm^2)	Absolute error (10^{-16} cm^2)
21.7	6.9	0.5	1.0
29.4	6.3	0.4	0.9
35.1	7.1	0.4	1.0
52.0	5.2	0.4	0.7
72.8	5.2	0.5	0.8
97.0	6.8	0.6	1.0
127	6.4	0.4	0.9
199	7.5	0.5	1.0
328	7.7	0.5	1.1
473	8.2	0.4	1.1

567	8.0	0.4	1.0
761	7.4	0.4	1.0
986	7.3	0.5	1.0
1385	7.3	0.5	1.0

While the Q-value of the $O_2^+ + D$ endoergic reaction is -1.5 eV, the Q-values for the exoergic reactions $D_2^+ + H$ and $CO^+ + D$, are 2.0 eV and 0.4 eV, respectively for $v = v' = 0$ [4,18,20,31-34]. At keV/u energies, the collision can be considered ro-vibrationally frozen and the differences in the Q-values are not a factor as all three cross sections converge to $(7.5 \pm 0.5) \times 10^{-16} \text{ cm}^2$ at 2 keV/u and are consistent with the $H_2^+ + H$ calculations [6] at keV/u energies (see Fig. 2). The discrepancy of the $CO^+ + D$ measurements with the $CO^+ + H$ state-to-state calculations with Lin et al. [33] is not understood. The calculations shown here are for an angle-averaged 300 K Boltzmann distribution of the initial vibrational states. A more detailed discussion is found in Ref. [9].

As seen in Fig. 2, below 2 keV/u the cross sections for O_2^+ and CO^+ are relatively flat while the D_2^+ measurements and H_2^+ theory decrease. Vibrational times of the molecular ions can be compared to collision times to assess the possible role of vibrational states. The energy of the excited vibrational level with respect to the ground vibrational level determines the characteristic vibrational time of the molecular ions. The first vibrational states in O_2^+ , CO^+ , and D_2^+ are comparable and correspond to 0.23 eV, 0.25 eV, and 0.19 eV, respectively [18-20,31,33]. Assuming an average vibrational energy level of 0.5 eV corresponds to about 50 a.u. in time. This vibration time can be compared to the various collision times calculated for our range of collision energy and assuming an interaction length of 1 a.u.. From Table 3 we note that the typical vibrational time of 50 a.u. is significantly longer than the collision times at and above 100 eV/u and therefore it seems reasonable that the collision can be considered frozen in this energy range. Indeed, a theoretical study by Lin et al. [33] indicates that for (CO^+, H) , the collision can be considered frozen down to 100 eV/u. In addition, the measurements for D_2^+ agree with the H_2^+ calculations which are based on a sudden approximation (ro-vibrational modes frozen) and straight line

trajectories [6]. The fact that the collision can be considered vibrationally frozen above 100 eV/u suggests that the difference between the measured CT cross sections for O_2^+ , CO^+ , and D_2^+ is not due to dynamical vibrational states.

Table 3. The collision time is shown as a function of collision energy and can be compared to the characteristic vibrational time of 50 a.u. (see text for details).

Collision energy	20 eV/u	100 eV/u	400 eV/u	2,000 eV/u
Collision time	20 a.u.	10 a.u.	5 a.u.	2 a.u.

Relatively flat decreasing CT cross sections at collision energies below the CT maximum have been previously observed for other systems. A flat cross section is consistent with an absorbing-sphere model [35] which assumes that there are sufficient crossings between the initial and final potential energy curves that CT can always occur. If favorable crossings are not present then a decreasing cross section would occur until the “reaction” window moves to a favorable crossing. This Landau-Zener curve crossing picture is often used to understand CT with atomic ions, e.g., it is used to explain observed Z-oscillations in CT with highly charged ions on H [36]. Figure 3 shows a variety of 4+ ions where the CT cross section with H (or D) was measured [37-40] with the ion-atom merged beams over an extended energy range. Note that the cross sections are similar at keV/u energies but can be different by about two orders of magnitude depending on the number of electrons on the core. The cross sections for Si^{4+} and Ne^{4+} are generally larger than the N^{4+} and C^{4+} with fewer electrons on the core. While detailed molecular orbital close-coupling calculations exist for these systems, one may try to understand CT using a Landau-Zener curve-crossing model. The flat energy dependence for the ions with multielectron cores indicates an abundance of favorable crossings. The larger quantum defect for the Ne^{4+} and Si^{4+} ions leads to larger

splittings between possible capture states. Larger splittings spread potential crossings over a wider range of internuclear separation.

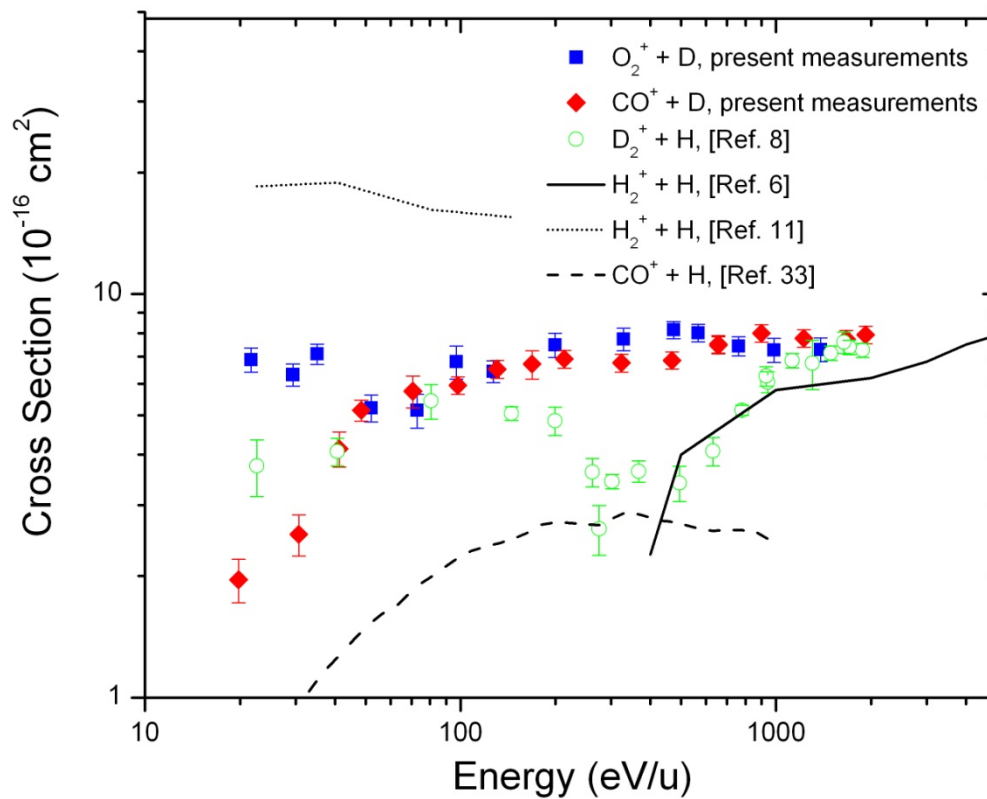


Figure 2. “Color online” ORNL ion-atom merged-beams cross sections for (O_2^+ , D), (CO^+ , D) and (D_2^+ , H) compared to theory for (H_2^+ , H) by Errea *et al.* [Ref. 6] and by Krstic [Ref. 11] and for (CO^+ , H) by Lin *et al.* [Ref. 33].

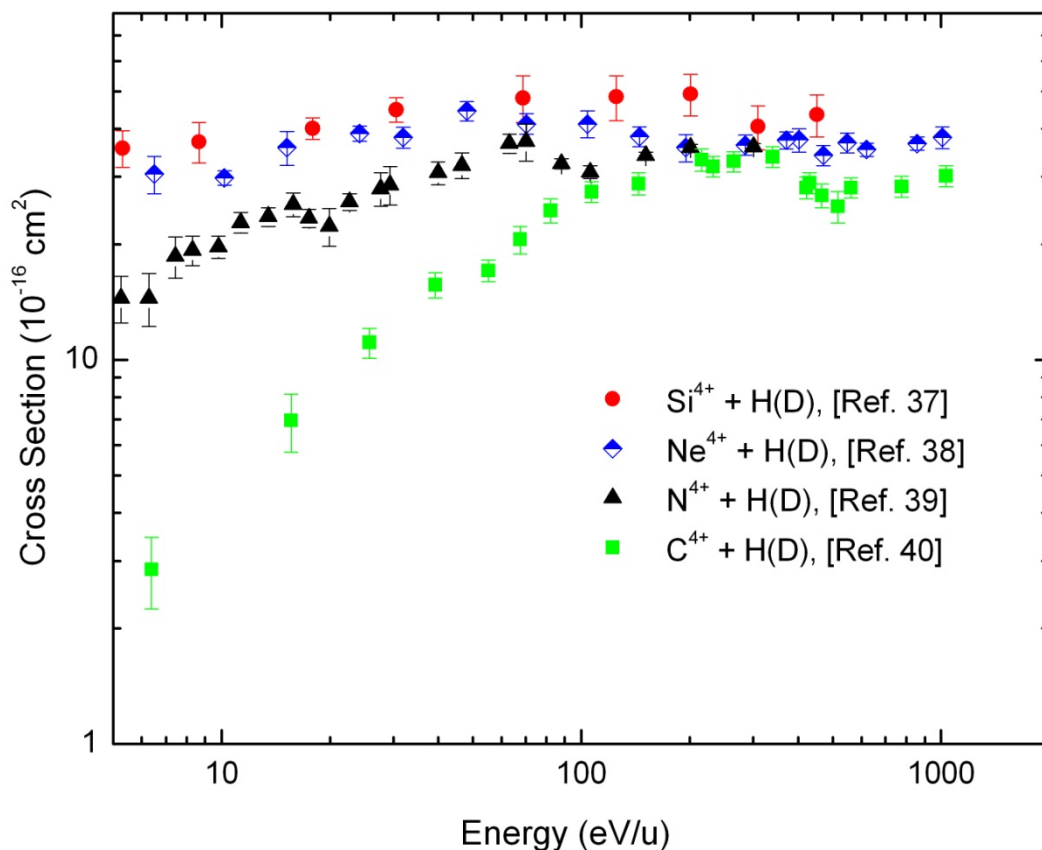


Figure 3. “Color online” ORNL ion-atom merged-beams measurements for various $q=4+$ ions with H(D) over an extended energy range.

Without the availability of detailed molecular potentials one might expect that the multielectron cores of O_2^+ and CO^+ would lead to more states for the complex $(\text{O}_2, \text{D})^+$ and $(\text{CO}, \text{D})^+$ than for $(\text{H}_3)^+$ and to an abundance of favorable crossings for charge transfer similar to the $4+$ atomic ion cases. The resultant cross section is relatively flat compared to the $\text{D}_2^+ + \text{H}$ cross section which involves only two electrons (see Fig.2).

While all three CT cross sections for diatomic molecular ions coincide at 100 eV/u, O_2^+ and CO^+ diverge below 60 eV/u with their different atomic cores. CT for O_2^+ remains relatively constant whereas CT for CO^+ decreases. At these energies we expect the ro-vibrational states to play an important part, as predicted by theory and through the collision times estimated in Table 3. Detailed state-to-state molecular-orbital close-coupling calculations are required for reliable calculations. Toward lower energies, the O_2^+ cross section is expected to decrease due to the fact that the reaction is endoergic by 1.5 eV. Such thresholds have been observed for (D_3^+, H) CT measurements [19] using the ORNL ion-atom merged-beams.

IV. CONCLUSION

Absolute CT cross sections during (CO^+, D) and (O_2^+, D) collisions were measured from 20 eV/u to 2 keV/u collision energy, using the merged-beams technique. A discrepancy is seen with $\text{CO}^+ + \text{H}$ state-to-state and angle-averaged calculations of Lin et al. [33]. Results are compared with our previous measurements for $\text{D}_2^+ + \text{H}$ and theory for $\text{H}_2^+ + \text{H}$. Toward higher collision energy, the CT cross sections all converge to $(7.5 \pm 0.5) \times 10^{-16} \text{ cm}^2$ at 2 keV/u. At lower energy, the flat energy dependence of the CT cross sections observed with the CO^+ and O_2^+ ions suggests a multitude of favorable crossings, an assumption inherent in the absorbing-sphere model and previously observed for CT with atomic ions with many electrons on the core. Relatively poor agreement with the sparse theory suggests more work is needed for understanding molecular-ion neutral reactions.

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