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Relative Differential Cross Section Measurements in H^+- CO_2 Low Energy Collisions

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Abstract

We have investigated the relative dissociative electron capture cross section in proton – CO_2 collisions within the energy range of 2-9.5 keV. Time of flight mass spectroscopy was employed in the measurements of the ionic fragments of the molecular target. Relative differential cross sections of ionic fragments were obtained. Fragment ions of CO^+ are found to be the dominant fragments. Ratio of CO_2^+ / CO_2^{++} was found to be of 3×10³. O_2^+ Fragments were observed.

Keywords: Experimental Physics, Electron capture, low energy collision physics, mass spectroscopy, Martian atmosphere.

1. Introduction

In spite of the fact that collision induced dissociation (CID) between ions and molecules has been widely studied, it is yet not completely understood due to the complexity of the interactions between charged particles and neutral atoms and molecules. It has been stated that the identification of the excited electronic states of CO_2 is still controversial [Green et al, 2002].

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Determination of the target beam interaction, in the identification of electronic states from target fragments and in measurements of cross sections (CS) related with their formation has been studied. Equally, interactions between ions and atoms as well as molecules are of fundamental interest in chemical reactions, astrophysics and plasma injection heating [Garcia, Mejia-Ospino, Guerrero, Alvarez & Cisneros, 2007].

Cross section measurements in low keV $H^+ - CO_2$ interaction were investigated by Van Zyl, Neumann, Le and Amme (1978), and single electron capture cross sections (SEC) were determined in the energy range 0.05-3 keV.

Other groups have studied the charge exchange cross section (CE); Basu, Jasperse, Robinson, Vondrak and Evans (1987) in the energy range 1-200 keV, Rees (1989) between 10 eV and 10 keV, Gao, Johnson, Hakes, Smith and Stebbings (1990) in the range of 0.5-5 keV, Luna et al (2003) between 10 and 100 keV, and Lindsay, Yu and Stebbings (2005) between 1 and 5 keV.

On the other hand, the fragmentation of CO_2 into C^+, O^+ and O in collisions with protons was investigated using an ion-ion triple coincidence technique by Moretto-Capello, Bordenave-Montesquieu D. and Bordenave-Montesquieu A. (2000). Protons in collisions with CO_2 in the range of 50 to 6000 keV were also reported [Knudsen et al. 1995]. To the best of our knowledge, we are not aware of any published differential cross sections (DCSs) for electron capture by protons on CO_2 within the energy range of the present investigation. The only available DCSs are those of Knudsen et al. (1995) in which the ratio of fragments cross section to that of the ionization cross section was measured within the energy range of 50 to 200 keV. In the present work, relative differential cross sections are reported for electron capture by 2-9.5 keV protons incident on CO_2.

It is worth noting that the reported data are of importance in direct applications in models of the Martian atmosphere, which is largely composed of CO_2 [Rodrigo, Garcia-Alvarez, Lopez-Gonzalez, Lopez-Moreno, 1990; Nair, Allen, Anbar, Yung & Clancy, 1994], which is indirectly impacted by solar wind protons. Atmospheric models involving the interaction of the solar wind protons require knowledge of the processes that are studied here as well as for the interpretation of energetic neutral atom (ENA) measurements from the Mars Express Spacecraft.

1. Experimental Apparatus

The experimental apparatus shown in figure 1, consists of the following sections:

Ions are generated in the Colutron ion source that was operated at 180 mTorr with a mixture of 80/100 argon to hydrogen with typical filament current of 0.1 A, and anode voltage of 80 volts. Ions within 1 to 10 keV of energy were directed into the Wien velocity filter (both Colutron source and Wien filter not shown in figure 1). The protons were selected and allowed into a 10 degrees deflection angle in order to remove neutrals. Following that, they were allowed to pass through two sets of collimators of 1 and 2 mm diameters into the interaction region. The interaction of protons with target gas occurs under the end of a hypodermic needle jet of 0.1 mm diameter. The gas pressure is measured in the surrounding of the jet region. The proton beam and the resulting neutrals are monitored and measured downstream with a Faraday cup and a channeltron.

Pulsed generator (200-300 ns duration) is used to drive a pulsed power supply PVX-4140 DEI (0-3000 volts).

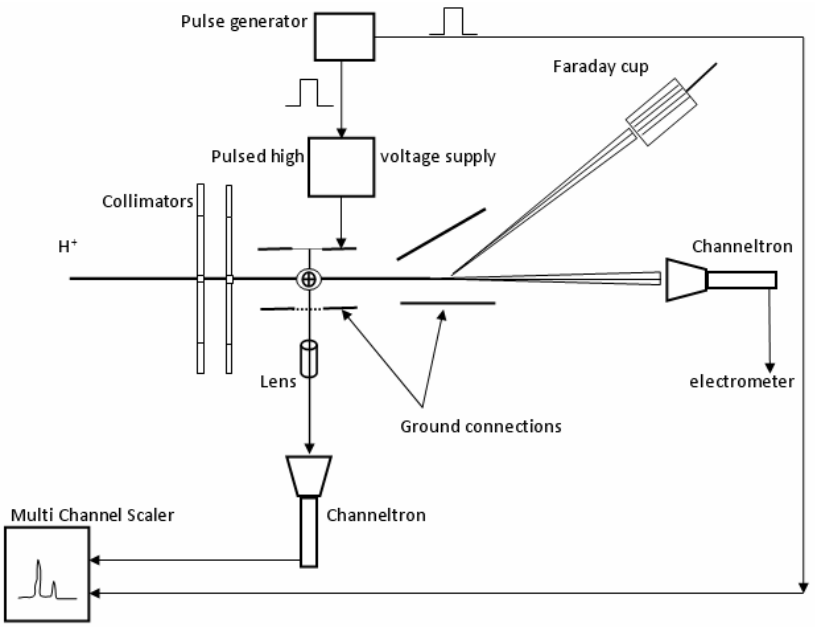

Figure 1: Schematics of the experimental apparatus.

The fragments of the ionization and dissociation of the target molecules are directed at 90° to the direction of the protons by the pulsed voltage that is applied on the left plate of the ionization region, while the right plate was maintained at ground potential. As the pulse generates the high voltage on the transverse electrostatic plates and at the same time initiates the flight of the ionic fragments into the time of flight region, which is 50 cm long, that ends with the detection of the target ionic fragments (TIF_s) by the channel electron multiplier. The same frequency of the pulse initiates the time scans of the Multi Channel Scaler (MCS-Ortec) in order to record the mass spectra of the target TIF_s. All the MCS scans were made at the same number of channels and the same dual time for all the acceleration energies. The proton beam was constantly monitored employing a PC interface and the average proton beam intensity was obtained during each measurement. The mass scale of the spectrum is calibrated by recording the mass spectrum of a reference species such as Ar under the same conditions of voltage extraction and pulse frequency.

The number of detected ions resulting from residual gases in our vacuum system that contributes to our total signal was recorded. In order to quantify these background signals, the mass spectra with and without target gas under the same experimental conditions were recorded. A subtraction of the ions from the residual gases is made by normalization to the intensity of proton intensity.

2. Results And Analysis

To measure the relative differential dissociative electron capture and ionization, CO_2 is admitted to the interaction region, after which the TIF_s are pulsed out into the time of flight region (TOF) and detected as a function of proton beam intensity and target gas pressure employing TOF mass spectroscopy.

The possible processes involved in the interaction between protons and CO_2 are:

$$H^+ + CO_2 \rightarrow CO_2^+ + H$$
$$H^+ + CO_2 \rightarrow CO^+ + O + H$$
$$H^+ + CO_2 \rightarrow CO + O^+ + H$$
$$H^+ + CO_2 \rightarrow C^+ + O_2 + H$$
$$H^+ + CO_2 \rightarrow C + O_2^+ + H \ ,$$

where the first reaction is ionization, while the remaining reactions are those of dissociative electron capture leading into the ionic fragments of C^+, O^+, CO^+, and O_2^+.

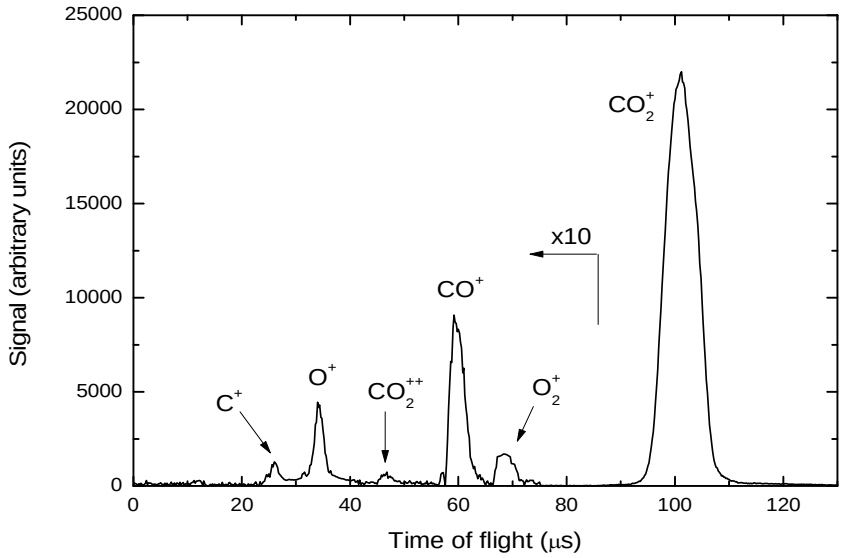

Figure 2: Recorded time of flight mass spectra identifying all of the detected ions.

Figure 2, shows an example of a recorded time of flight mass spectra identifying the ionization and the dissociative ionic fragments. The areas under the various peaks, after removing the background, were normalized to the same proton current and target gas pressure for all the collisional energies. Our measured relative cross sections for ionization and all of the dissociation fragments are presented in figure 3 within the energy range of 2-9.5 keV. It can be seen that $CO^+$is the dominant fragment, followed by O_2^+, O^+, C^+ and CO_2^{++}. Besides the statistical uncertainties as shown in figure 3 that are involved, there are additional systematic uncertainties that are mainly due to pressure measurements as well as to the proton beam intensity. The errors in the target gas pressures are identified by the pressure gauge manufacturers to be of ± 5 % and the variations of the gas pressure during each measurement. Therefore gas number density within the proton beam cross section would be influenced further by an estimated additional 10%. Measurements of the average proton beam intensity were determined employing a PC interface.

This allowed the evaluation of the average current of protons at the end of each scan. However the statistical uncertainties were at about 30 % at 2 and 2.5 keV of energy. These uncertainties fall down to 10% as the collisional energy was increased higher than 2.5 keV.

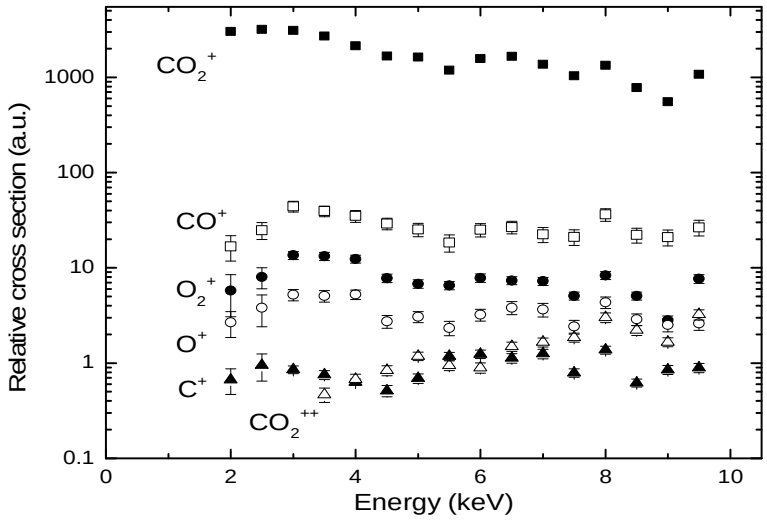

Figure 3: Our present measured differential dissociative electron capture and ionization.

The ionic fragments of CO^+,O_2^+, O^+ and C^+ show some increasing behavior as the collisional energy increases above 2 keV, reaching to a maximum at about 3 keV. It can be seen that the position of the above maxima is dependent on the mass of the ionic fragment. Following that, the cross section decreases to a minimum that is followed by a second increase to a second maximum. The only exception is that for CO_2^{++}, which continues to increase as the energy increases. Straub et al. (1996) measured the absolute partial cross section for product ions in the energy range 25-1000 eV for electron collision with CO_2 employing a time of flight (TOF) mass spectrometer.

Their absolute partial cross section at energies below 50 eV showed the fragment intensity preference of $CO^+ > O^+ > C^+$similar to our present work. Their results [Straub et al. 1996] above 50 eV electron collisional energies showed that the ratio $O^+/_{CO^+}$ exceeded 1.

Our highest proton energy of 9.5 keV would correspond to about 5.4 eV electron energy in collision with CO_2. That would put us at an energy well below the minimum energy of 25 eV reported by Rapp and Englander-Golden (1965). A critical survey of available experimental data reported by Lindsay and Mangan, (2003) with their partial cross sections based on the measurements of the product ions with time of flight mass spectroscopy by Straub et al. (1996), showed that for the total cross section below 50 eV, Lindsay et al. (2003) adopted the values of Rapp and Englander-Golden (1965). The cross sections for energies under 50 eV show a ratio of $O^+/_{CO^+}$ fragments to be higher than 1 compared to our measured ratio of $O^+/_{CO^+}$ of about 10. While the $CO^+/_{C^+}$ cross sections ratio in Lindsay et al. (2003) is about 2 at 50 eV and increasing to 10 at about 30 eV, compared with our present results for $CO^+/_{C^+}$ of about 20 over all of our energy range.

On the other hand, the observation of a significant O_2^+ fragment peak is rather interesting as it has not been reported by Straub et al. (1996) and Lindsay et al. (2003). It was stated by Straub et al. (1996) that their apparatus embodied a short path time of flight that was not sufficient to resolve the possible contribution from the O_2^+ from their adjacent and more dominant peaks of CO^+. While in our case, the peak of O_2^+ is well resolved since our TOF path is of 50 cm that is sufficiently enough to resolve the O_2^+ from the adjacent and more intense peak of $CO^+$and the estimated O_2^+ relative ratio with respect to CO_2^+ is obtained.

However optical emission spectroscopy of CO_2 glow discharge at low pressure [Reyes, Mendez, Oscorio-Gonzalez, Castillo & Martinez, 2008] shows a substantial contribution from the excited state $A^2\Pi_u$of O_2^+ resulting from the dissociative excitation according to the following reaction

$$e + CO_2 \rightarrow C + O_2^+ + 2e$$

Furthermore, CO_2^{++} fragments were also observed in the present work and were previously reported [Straub et al. 1996; Freund, Wetzel &Shul, 1990; Orient & Strivastava, 1987; Märk & Hille, 1978; Crowe & McConkey, 1974; Adamczyk, Boerboom & Lukasiewicz, 1972; Krishnakumar, 1990; Rudd, DuBois, Toburen, Ratcliffe & Goffe, 1983] with a ratio of 0.01 for $\sigma(CO_2^{++})/\sigma(CO_2^{+})$ [Rodrigo et al.1990; Nair et al. 1994] at about 120 eV electron energy. Our present measured ratio is between 3×10^{-4} and3×10^{-3} over all the energy range of 3.5 to 9.5 keV.

The excited channel responsible for the O^+ production is the low lying state $X^2\Pi_g$ leading to $O^+(4S_u)$ + CO $(X^1\Sigma^+)$, while the CO^+ is produced from the higher excited state of $b^4\Pi_u$through the channel leading to $O^3(P_g)$ + $CO^+(X^2\Sigma^+)$ and the channel $A^2\Pi_u$ leading to $O(^1D_g)$ + $CO^+(X^1\Sigma^+)$. The dissociation channel responsible for the C^+fragments is the ground state of CO_2^+ leading to $C^+(^2P)$ + $O_2(X^3\Sigma_g)$.

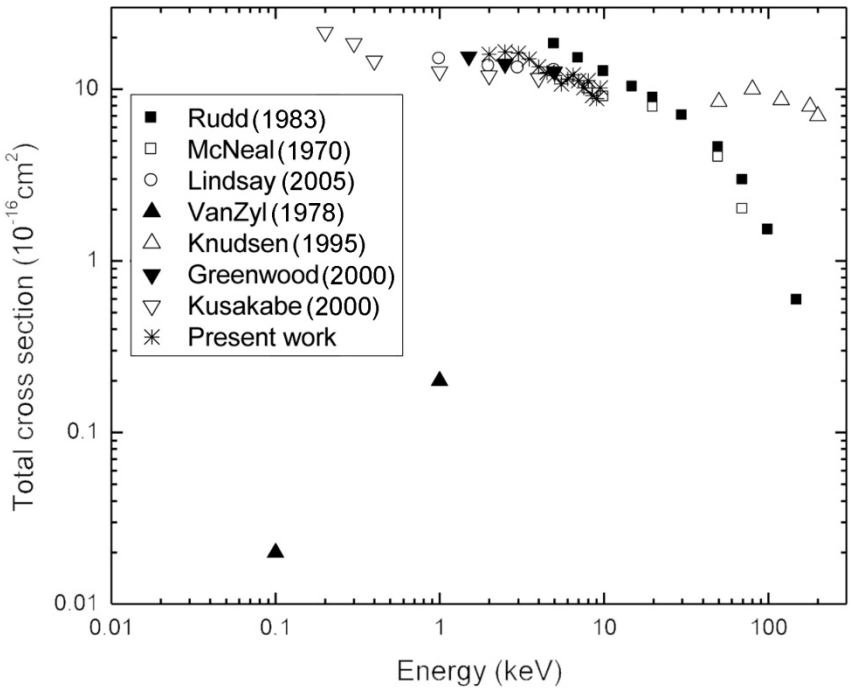

Figure 4. Present and previous data for H^+ in collision with CO_2.

We have multiplied the sum of our ionization and differential relative cross section data with a constant factor and superimposed these data in figure 4 in order to compare them with those of other investigators [Lindsay et al, 2005; McNeal, 1970; Greenwood, Chutjian & Smith, 2000; Kusakabe et al. 2000]. It can be seen that our present data overlap perfectly with them. The behavior of our total cross section agrees well with previously measured total cross section data as seen in figure 4.

3. Conclusions

Relative cross sections for the ionization and dissociation fragments are measured for protons in collision with CO_2 within the energy range of 2 to 9.5 keV for the first time.

The relative cross sections for all the dissociation fragments show minor changes within the energy range of this work.

Over all the energy range, the fragments show the following preference of

$$CO^+ > O_2^+ > O^+ > C^+.$$

Unlike the electron impact experiment, the O_2^+ show an interesting cross section that exceeds the O^+relative cross section and in accordance to the finding of the glow discharge experiments.

The sum of all the relative differential cross sections measured when multiplied by a certain factor falls perfectly with previously measured experimental data.

The dissociation channel responsible for the O_2^+ production is the excited state of

$$A^2\Pi_u \text{ or } X^2\Pi_g.$$

The dissociation channel responsible for the O^+ production is the low lying state $X^2\Pi_g$ leading to $O^+(4S_u)$ + $CO(X^1\Sigma^+)$, while the $CO^+$is produced from the higher excited state of $b^4\Pi_u$ through the channel leading to $O^3(P_g)$ + $CO^+(X^2\Sigma^+)$ and the channel $A^2\Pi_u$ leading to $O(^1D_g)$ + $CO^+(X^1\Sigma^+)$. The dissociation channel responsible for the C^+fragments is the ground state of CO_2^+ leading to $C^+(^2P)$ + $O_2(X^3\Sigma_g)$.

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