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Collisional induced double electron loss of NO^- and CH_4^- anions below 10 keV energies

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ABSTRACT

Double electron detachment cross sections for methane and nitric oxide negative ions passing through oxygen and nitrogen gas targets have been studied in the energy ranges of 2.0–10.0 keV for CH_4^- and 4.0–10.0 keV for NO^-. The signal growth rate method was employed. Fragmentation was only observed for NO^- anions colliding with both targets. In the case of CH_4^- anions no fragmentation was observed. The double electron detachment cross sections for methane ions is almost one order of magnitude larger than the one for nitric oxide ions. The fragmentation cross-sections for NO^- is larger than the double electron detachment cross sections.

1. Introduction

The process of losing two electrons from a negative ion, that here on will be called *double electron detachment* (DED), is of fundamental significance in atomic physics. For example, a conditional probability model based on combining *single electron detachment* (SED) with electron collision induced ionization cross sections describes, at least qualitatively, the process of DED [1]. In terms of applications, DED in low energy plasmas may induce a polarity change of a determined species from negative to positive, thereby increasing the thermal electron population. Polarity inversion is normally used in accelerators technology to increase ion beams kinetic energy, for this reason, details of DED are relevant for ion beam optimization. In the case of molecular anions, it is of significance to probe collisional DED because molecular anions have recently been found to play important roles in certain atmospheric environments [2]. For instance, molecular anions are abundant in the ionospheres compositions of Venus, Titan and Mars [3]. Also NO^- is important in film deposition plasma techniques [4,5] because it is a source of free thermal electrons causing electrical breakdowns.

The contribution of DED is believed to be negligible in plasma equilibrium, however, the available data of this process is scarce. To our knowledge, DED from CH_4^- and NO^- molecular anions have not been quantified, until now. In addition, it is interesting to point out that molecular anions in earth's atmosphere, are believed to have some positive influence in the human health [6,7].

There has been efforts to quantify the contribution of the DED process to the electron population in plasmas. For example, SED and DED from NO_x^- molecules like NO_2^- and NO_3^- in collisions with several targets have been measured by Bennett et al. [8] using the beam attenuation technique. They found that DED cross sections were 20 times smaller than SED's. Matic and Cobic also found that the σ_{-11} was an order of magnitude lower than SED [9]. This order of magnitude difference between SED and DED has confirmed in other investigations [1,10,11].

DED and fragmentation from a simple molecular anion may be important to understand the details of the total electron detachment process. For instance, theoretical models and many-electron loss collision phenomena involve negative ions on gases [12]. However, they have not been investigated as extensively as those involving positive ions [13,14]. DED may be a significant reaction channel in intermediate and low energy collisions between negative ions and neutral gases, therefore, experimental studies of collision systems of this kind may be useful for improving our understanding of the physics of negative ions interactions with atoms and molecules [15].

The present work reports measurements of collisional DED from CH_4^- and NO^- on O_2 and N_2 at energies below 10.0 keV. One fragmentation channel into a positive fragment cross sections from NO^- is also reported. These measurements may shed light in understanding more details of hydrocarbon and atmospheric plasma physics.

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2. Experimental method

The experiment was performed with an electrostatic accelerator that operates below 10.0 keV. Fig. 1 shows a schematic of the apparatus. A description of the details for this experiment will be discussed below; for a complementary description see Ref. [16].

The experiment initiated with a hot tungsten filament ion source, where molecular ion beams of CH_4^- or NO^- were generated. In the case of CH_4^- a 50% gas mixture of CH_4 and Ar was injected into the ion source chamber. In the case of NO^-, we injected a gas mixture of 3 ppm of NO_2 with N_2. The anion beam was accelerated to energies less than 10.0 keV. The negative ion beams were chosen with a magnetic mass selector. Next, the anion beam wends its way up to a gas-cell where the beam collides with high purity gas targets. The results of the interaction of the negative ion beam with the gas target was analyzed on a detection system comprising two channel electron multipliers: one located in the center axis of the accelerator (CEM-1) and the second placed in one side of the accelerator axis (CEM-2). Beam and charged particles were separated from the resulting molecules and fragments. The positive fragments resulting from the DED were deflected to CEM-2, with an electric field (E) generated by applying a voltage to a set of parallel plates, while the neutral beam resulting from losing electrons continues its trajectory into CEM-1.

When E of the analyzer plates was zero, the count rate of the central CEM-1 consisted of neutral molecules plus negative and positive ions. When E was turned on, the lateral CEM-2 measured the negative ions. With no gas load in the gas-cell, the counting rate in CEM-1 with $E = 0$ was verified to be similar to the sum of the counts rates of CEM-1 plus CEM-2 when E was turned on. This test was performed to verify similar gain and efficiency conditions on both CEMs. By sweeping the voltage in the plates it was possible to find fragmentation processes.

Since no absolute measurements were possible with the present experimental setup, we have calibrated our experiment to those of well known electron capture cross section of protons on Ne and CH_4 that were measured in the same energy range by Kusakabe et al. [18], Allison [19], Rudd [20] and Lindsay et al. [21]. The calibration was suitable for all cases.

2.1. Signal growth rate method

The *signal growth-rate method* (SGR) has been widely used to study the electron detachment of negative ions in collisions with atoms and molecules [22,23]. This method is based on approximations to the solutions to the equilibrium equations of the fraction of the resulting particles to the number of anions in the ion beam as a function of the target density Π. It consists basically in passing a pure incident anion beam (I_{-1}) through the collision gas-cell and measuring the signal growth rate of the new charge state fractions

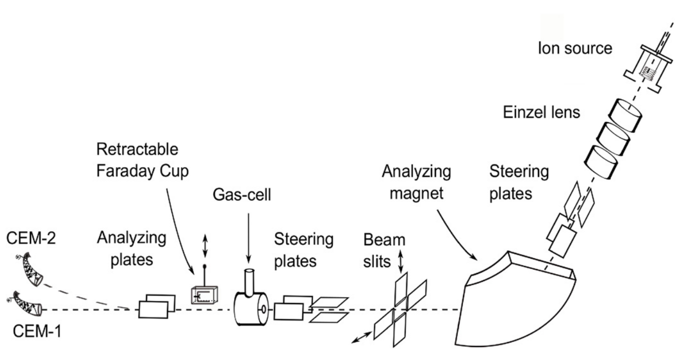

Fig. 1. Schematic drawing of the apparatus taken from [17].

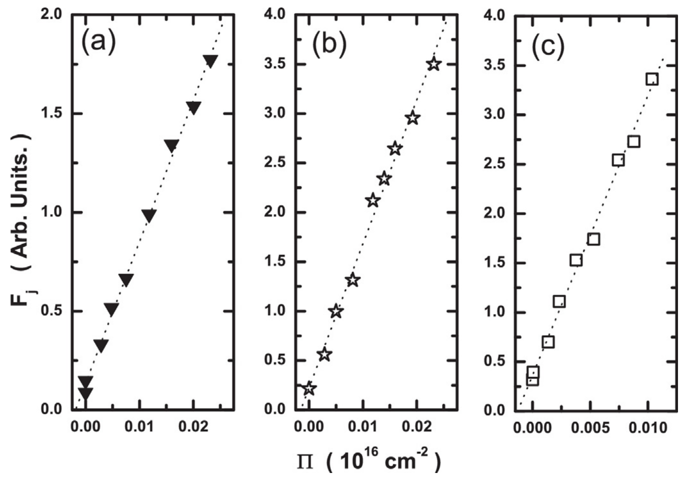

Fig. 2. Examples of grow rate curves for the case of CH_4^- projectile. The cross section corresponds to the slope of the curves according to Eqs. (3)–(5). (a) The resulting ion CH_4^+, (b) resulting ion NO^+ and (c) resulting ion N^+. The linear fit is represented for dotted grey line.

I_j, where $j = 1, f$ are the counts of NO^+ and N^+ respectively. Then, the cross section σ_{-1j} can be estimated from the slope of the linear portion in the F_j vs Π curve, see Fig. 2. $F_j \equiv \frac{I_j - I_b}{I_{-1}}$ and I_b is the signal counts rates resulting from the interaction with the vacuum residual gas. The cross section is derived from:

$$F_j = \frac{I_j - I_b}{I_{-1}} = \sigma_{-1j}\Pi + \Theta(\Pi^2) \quad (1)$$

Π denotes the target thickness defined by the ideal gas equation (2) and $\Theta(\Pi^2)$ are terms of quadratic order:

$$\Pi = \frac{lP}{\kappa T} \quad (2)$$

where l is the gas-cell effective length, P and T are the pressure and temperature within the gas-cell and κ is Boltzmann's constant. Π can also be expressed in terms of the gas density n as $\Pi = nl$. At a given kinetic energy the signal was measured as a function of the gas-cell pressure. Examples of F_j versus Π curves are shown in Fig. 2 where the cross sections σ_{-11} and σ_{-1f} correspond to the slopes of the curves. In this experiment, the data were taken at pressures below 10^{-2} Torr in the gas-cell. A check for second order contribution to the solutions of F_j had no-effect on the cross sections derived from a first order-fit. Hence, when a molecular negative ion beam passes through the collision chamber with gas pressures below 10^{-2} Torr, single collision regimen can be achieved.

3. Results and discussion

DED cross sections for CH_4^- and NO^- are presented in Fig. 3 and Tables 1 and 2 for target gases O_2 and N_2 respectively. We were able to measure fragmentation cross sections only for the case of NO^- anion beam into N^+ fragment and are presented in Fig. 2b and Table 2. Eqs. (3) and (4) describe DED processes for the present two target gases:

$$CH_4^- + X \rightarrow CH_4^+ + X^* + 2e^- \quad (3)$$

$$NO^- + X \rightarrow NO^+ + X^* + 2e^- \quad (4)$$

where X represents O_2 or N_2 gas target. Similarly, we describe the fragmentation process by Eq. (5)

$$NO^- + X \rightarrow N^+ + O^* + X^* + 2e^- \quad (5)$$

where X^* is used to indicate that the final internal states are unknown. In Fig. 3 we note first that the cross sections are