5.1 ELECTRON-MOLECULAR ION INTERACTION

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1 Introduction

The long storage times in heavy ion rings make them excellent tools to study intramolecular processes. Depletion of excited vibrational levels by radiative decay and well defined collision energy are the main advantages in studies of molecular ion - electron collisions [1,2,3]. In the last year the experiments concerned the following aspects of electron - molecular ion interactions: absolute cross sections for dissociative recombination (DR) as well as dissociative excitation (DE) and kinetic energy release in DR process. The study of product state distribution in DR of diatomic molecular ions has been successfully initialized by means of a method of translational spectroscopy.

2 Dissociative recombination

2.1 Cross sections

The molecular group has recently performed a DR study of the D_3^+ ion, which is an heavy isotopomer of H_3^+ with the same electronic structure. The H_3^+ ion has been recently detected in two interstellar clouds [4] and is believed to play the central role in the chemistry of these natural plasmas. The H_3^+ saga has been previously described elsewhere [5], so that a brief overview will be given here. For the so-called DR direct process to occur efficiently, a favorable curve crossing is needed between the ionic and the dissociative potential curves; this is not the case for the DR of a cold H_3^+ ion. To account for the manifold experimental evidences of its DR high efficiency at low energy, a new theory has emerged over the last couple of years, although no detailed calculations have been carried out. The new mechanism is called the tunneling mode and is based on non-adiabatic couplings between the inner wall of the dissociative state (X^2E') of H_3 and the ionic potential curve.

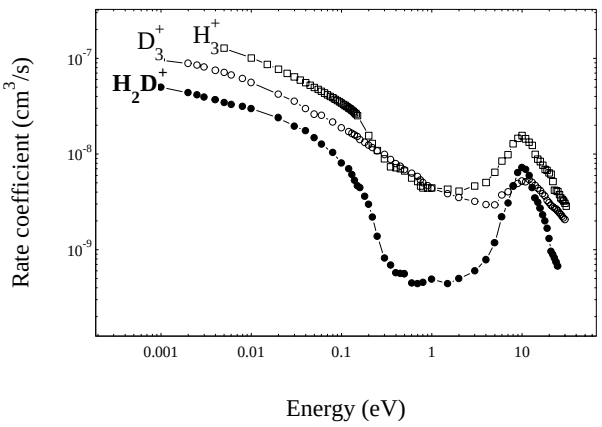

Figure 1. The absolute dissociative recombination rate coefficients measured for H_3^+, H_2D^+ and D_3^+.

Nevertheless, it is expected that the tunneling mode should be less efficient for D_3^+ than for H_3^+, because doubling the mass of the ion should reduce dramatically the rate of quantum tunneling through the barrier. One of our purposes was to check this prediction and the efficiency of the D_3^+ dissociative recombination has effectively been measured to be a factor of four smaller then the one of H_3^+. Another point was the search for a field effect which was previously found to influence some Canadian measurements [6] on H_3^+; some highly excited Rydberg

states involved in the recombination process were field ionized during their passage through an electrostatic deflector. We did not observe any evidence of field effect in our D_3^+ study.

Very recently, we have also looked into an ion of particular atmospheric interest: N_2^+. For instance, its dissociative recombination is believed to be responsible for the escape of atomic nitrogen fragments from the Martian atmosphere, when their kinetic energy is sufficient. Due to their lack of any dipole moment (infrared inactive molecules), vibrationally excited ions produced in a conventional source do not relax in the storage ring prior to the recombination measurements. Nevertheless and thanks to the new operating JIMIS hallow cathode source, we were able to produce relatively cold N_2^+ ions. This has been confirmed by the dissociative excitation data (threshold location) as well as by the 3D imaging data (see below). Our results are in perfect agreement with most of the other experimental results, which indicate a fairly high efficiency for the dissociative recombination of slightly excited N_2^+ ions. An accepted value of the corresponding rate coefficient at room temperature would be $2 \cdot 10^{-7}$ $cm^3\ s^{-1}$. Contrary to the H_3^+ case, the direct DR process is quite efficient for N_2^+ due to the presence of different dissociative routes in the vicinity of the v=0 level.

2.2 Imaging spectroscopy

The kinetic energy release in the dissociative recombination process was measured for $^3HeD^+$ for incident energies between 0 and 15 eV [7]. It was found that the $He(1s^2)$+D(n=2) channel completely dominates at zero electron energy. The data are in agreement with MQDT calculation that predict the dissociation route via $C^2\Sigma^+$ state, leading to the situation that the asymptotic state D(n=2) totally dominates. At collision energies above 10 eV, many channels leading to excited He atoms were find to contribute and a strong angular anisotropy of the dissociation products is observed.

The imaging spectroscopy has been used to measure the kinetic energy release in dissociative recombination of OH^+. It has been found that the metastable a $^1\Delta$ state is populated in the beam even after the storage time of 10 s. Unfortunately, calculations give a lifetime of 0.1s which disagrees completely with our statement. Our measurements permitted to calculate a new experimental value of the a $^1\Delta$ state excitation energy of 2.25 eV which is in good agreement with an electron spectroscopy measurement by Katsumata and Lloyd [8].

A three-dimensional, position-sensitive imaging detector has been used to measure the atomic limits and vibrational features of the DR process with N_2^+. A vibrational distribution of ions produced in the hollow cathode source JIMIS has been found to be v=0 (55%), v=1 (23%), v=2 (13%) and v=3 (10%).

3 Dissociative excitation

In the dissociative excitation process an electron excites an ion to a repulsive ionic state and thus results in dissociation. An alternative pathway is when a doubly excited neutral state is formed which then autoionizes to the vibrational continuum of the ground state ion. Both mechanisms of DE have been studied in CRYRING for $^4HeH^+$ [9] and D_3^+ molecular ions. Absolute cross sections measured for

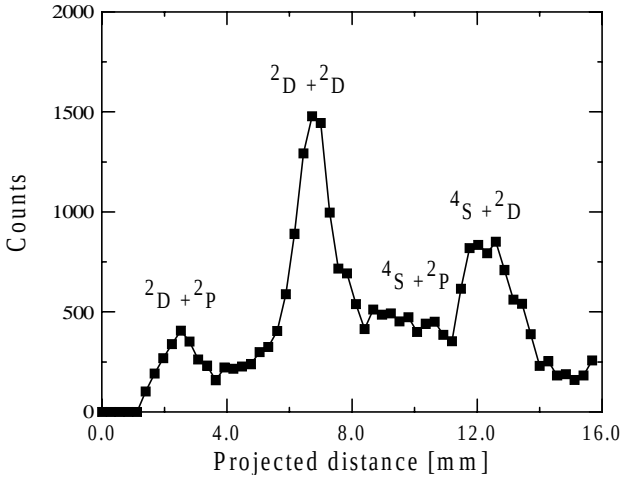

Figure 2. The distribution of projected distances between two nitrogen atoms following DR of N_2^+.

the ($^4He^+$+H) and (^4He+H^+) disscociation channels show step rises at energies corresponding to vertical excitation energies from the ground state to the excited ionic states (19 and 30 eV, respectively). The peak found in the (^4He+H^+) channel is due to an electron capture to Rydberg series converging to the first excited $a^3\Sigma^+$ and $A^1\Sigma^+$ ionic states, that then autoionize. This finding has been confirmed by the theoretical calculations by Orel and Kulander. In the case of D_3^+ both dissociation channels (containing D_2 and D, respectively) seem to have the same threshold for the direct excitation at about 14 eV. The low energy peak found in the D_2 channel at an

energy of 8 eV arises probably as a result of a competition between dissociation and autoionization of the resonant neutral states formed in the electron capture process.

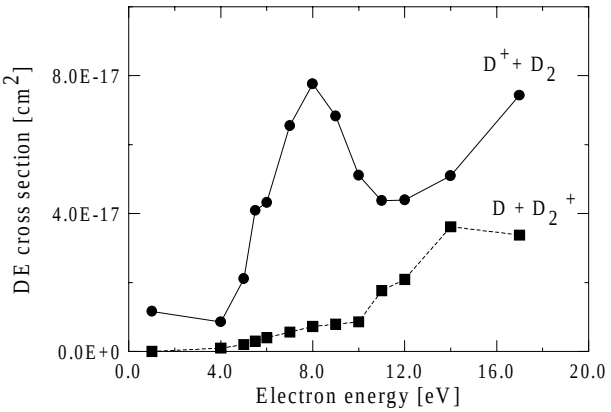

Figure 3. The absolute dissociative excitation cross sections measured for D_3^+.

4 Development of an imaging detector

An imaging detector for molecular fragmentation has been developed [10]. It is a time and position sensitive detector that determines both the transversal separation and the difference in time of arrival of molecular fragments. The detector has been used first time in December 96 to study dissociative recombination of N_2^+. Then the kinetic energy release in the reaction and hence the quantum states of the fragments have been measured.

The main part of the detector is a stack of three micro channel plates (MCP) and a phosphorous screen. The neutral particles, created in a DR event, produce a bunch of electrons in the MCP that are further accelerated to the phosphorous screen. The fluorescence light from the screen is detected by a charge couple device (CCD) camera. The frames are analyzed to find the position and the transversal separation of fragments according to the beam axis. To measure the difference in time of arrival there are a number of gold strips evaporated on the rear side of the last MCP. By fast electronics it is possible to measure the time difference from pulse from separate strips (fast preamplifier and constant fraction discriminator (CFD) and analog to digital converter (ADC)).

The spatial resolution of the camera is 160 μm as best and the time resolution is about 300 ps.

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