

Electron impact induced dissociation of N_2H^+ into NH^+

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Abstract. Dissociative excitation of N_2H^+ has been investigated in the energy range from 7.5 to 2500 eV using a crossed beams apparatus. Absolute cross sections are reported for the channel leading to NH^+ formation. Good agreement is found with the previous results of Fogle et al, from the Oak Ridge group [1]. The threshold measured for this channel indicates the presence of excited states in the target beam.

1. Introduction

Collisions between electrons and molecular ions lead to reactions which differ drastically with increasing energy. At the lowest energy range (from 0 eV), only the exothermic dissociative recombination (DR) process may take place but for higher electron energies, endothermic reactions appear to play a dominant role. Above a few eV, that is, in the energy domain where DR is seen to exhibit a vanishing importance, dissociative excitation (DE) first dominates the electron-molecular ion interaction by producing one ionic fragment. For a diatomic molecular ion DE can be written in the general form:



At slightly higher energies, ionization processes are energetically accessible and single ionisation (SI) may be observed:



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However, for diatomic systems, the doubly-charged molecule is generally unstable against dissociation so that dissociative ionisation (DI) generally dominates the reaction:



These three processes (DE, SI, DI) display individual characteristic thresholds since they involve a transition from the initial ion state to an upper excited or ionised state that is dissociative or not. Thus DE, SI and DI are likely to be significant reactions in the chemistry of environments, where high enough electron energies are encountered, for example, in divertor and fusion-wall plasmas or in stellar nebulae. Most of the experimental studies of these reactions have not been performed using single or multiple pass merged beam methods, but using the most appropriate crossed beams method. A review of work performed up to 1998 using these various techniques has been given by Dunn and Djuric [2] but there have been a number of studies published since then, especially by the groups of Louvain-la-Neuve and Oak Ridge.

In this paper, we report on new measurements for one of the dissociation channels arising from electron impact on N_2H^{+} . This ion has great significance in astrophysics, attested, for example, by the recent paper published in *Science* [3] describing astronomical measurements of its abundance around solar nebula that allowed the mapping of the so-called ‘‘Snow Line’’. N_2H^{+} is formed by proton transfer from H_3^{+} to N_2 but is destroyed by charge transfer to CO which is much more abundant. When CO has condensed out of the gas phase, within the ‘‘snow line’’, then the N_2H^{+} concentration becomes significant and can be measured, thus serving as a marker for this condensation, which is a precursor for planet formation.

Being protonated nitrogen, it is likely to be present in any plasma where hydrogen and nitrogen co-exist. Thus it is considered to be one of the ions likely to be present in fusion wall plasmas where nitrogen gas will be present either due to leaks or deliberately introduced as a coolant molecule. There is a need therefore for experimental data on its reactions with energetic electrons.

2. Experimental method

The crossed electron-ion beam apparatus at the Catholic University of Louvain has been used for this measurement. This apparatus has been described in detail elsewhere [4] so only a brief description of the technique will be given. Figure 1 shows the experimental layout.

Ions are formed in an ECR source (1) [5], extracted and accelerated to 8 keV through the focusing Einzel lens system (2), mass selected by a magnet (3), deflected by an electrostatic spherical deflector (4) to remove neutrals formed in transit and then pass through the collision region where they interact with the electron beam produced by the electron gun (5- also shown as an insert). The electron energy can be tuned from a few eV up to 2.5 keV. Collision fragments are separated from the primary beam by a 90° analyzing magnet (6). The primary beam is dumped and detected into a Faraday cup (7). The analyser slit (8) allows separation of the collision fragments from spurious ions coming from the primary beam. An additional 90° spherical electrostatic analyzer (9) separates signals according to the kinetic energy and focuses ions onto a channeltron detector (10). The analyzing magnet (6) produces a uniform magnetic field (B), which acts as the standard mass analyzer. Product ions are selected according to the mass over charge ratio, which is related to the field, the radius of the ion trajectory ($R_B = 0.3$ m) and the velocity (v):

$$\frac{m}{q} = \frac{B}{R_B v}. \quad (4)$$

In the electrostatic analyzer (9), the electric field (F) improves the resolution of the system by further selecting dissociation products according to their kinetic energy:

$$E_K = q F R_E. \quad (5)$$

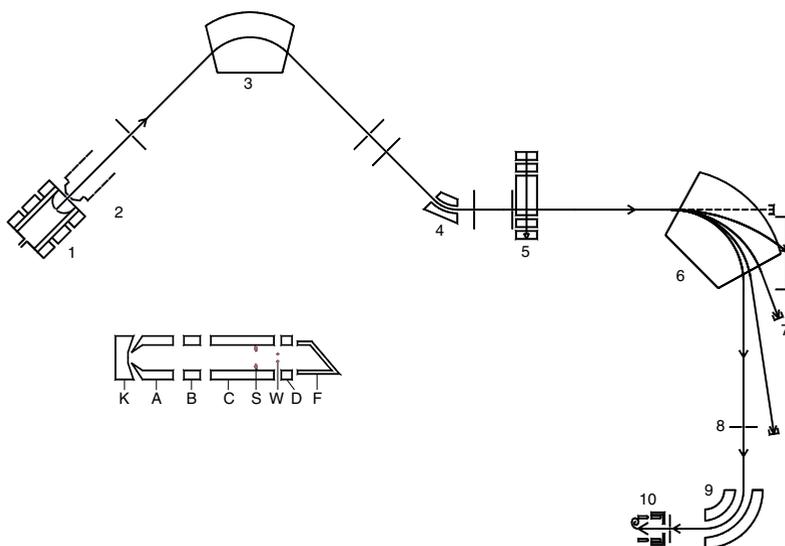


Figure 1. Crossed electron-ion beam apparatus at Louvain-la-Neuve.

In this expression, R_E is the radius of the ion trajectory ($R_E = 0.15$ m).

The experiments make use of the animated crossed Electron-Ion beam method full details of which are given in [6]. In SI experiments, absolute cross-sections are directly determined through the measurements of the total number of events produced during passage of electron beam across the ion beam. However, in dissociation experiments (DE and DI), the fragments are only partially collected, due to both the velocity and angular spread resulting from the kinetic-energy-release (KER) which is transferred to the products. This KER is associated with the shape of the potential surface of the upper state (excited or ionized) involved in the dissociation process. Hence, the velocity distribution of the ionic products is determined and further integrated in order to estimate the transmission factor, for each individual dissociation channel. This is done by scanning the analyser magnet and integrating the resulting product spectrum. This measurement also provides information on the KER distribution of the fragments [7].

In present type experiments, spurious contributions of molecular species formed with isotopes frequently pollute the experiment. In the present case, $^{14}\text{N}^{15}\text{N}^+$ ions should evidently interfere both with the primary N_2H^+ beam and with the NH^+ fragment, the resolving power of the analyzing magnets being not sufficient to discriminate between the species. For this reason, we have used isotopically pure $^{14}\text{N}_2$ as source gas, thus avoiding any contamination issues.

3. Results and discussion

First of all we did not observe any N_2H^{++} signal and indeed it seems that this ion is not stable. Theoretical calculations [8] did predict the presence of a weak deep well for this ion but also that it would rapidly dissociate. Present negative result indicates that its lifetime is certainly shorter than the transit time between the collision region and the detector ($\sim 9 \mu\text{s}$).

Electron impact cross sections for NH^+ formation are shown in Fig. 2 together with the results of Fogle et al. taken using the Oak Ridge apparatus [1]. A general satisfactory agreement is seen for the magnitude of the two sets of data, though the threshold region is better covered in this work. The thermodynamic estimation of the lowest DE threshold for this ion indicates that electrons must have

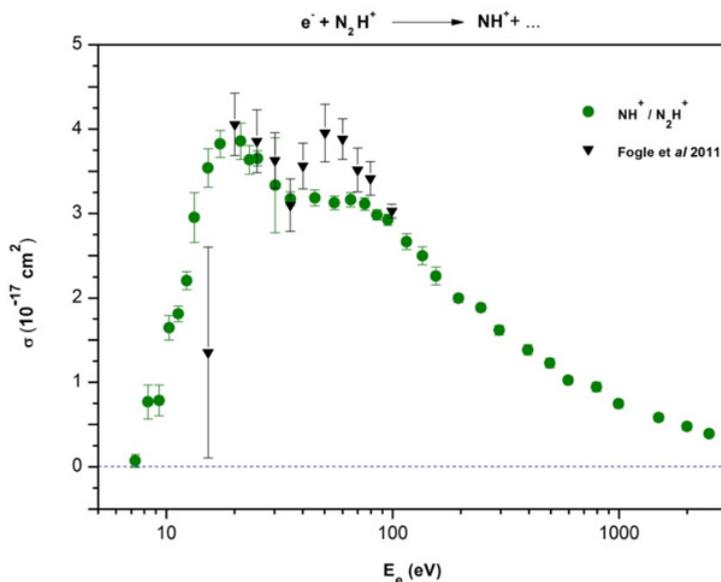


Figure 2. Measured absolute cross section for the electron impact dissociative excitation of N_2H^+ leading to NH^+ formation. The green full circles are our data, inverted triangles, those of Ref. [1].

a kinetic energy of at least 11.36 eV in order to produce NH^+ . However, this value does not take into account the shape of the potential surface of the upper transition state so that this estimation must be considered as a lower limit. Present experimental data show a steep rise at about 12 eV, but the significant signal observed below this value (down to 7 eV) indicates that target ions should have considerable internal excitation. This could be ro-vibrational excitation but populated metastable electronic states could also possibly contribute to the apparent sub-threshold cross-section. Such states have been identified by Moran and co-workers [9] in N_2^+ beams where states with lifetimes on the order of tens of microseconds were seen to survive the transit from the ion source to the target region. Such states might also be present in the present experiment. We shall examine this further in a subsequent publication where other measurements will be analysed.

The thermodynamic threshold for DI to produce $NH^+ + H^+$ is at 25.4 eV. As seen in Fig. 2, a peak appears in the data of Fogle with a threshold at about 35 eV. This onset appears at the same energy in our results but the result is rather a plateau. Such an ionisation process must of course pass through a transition to an upper excited state and this would indicate that this state has a steeply descending slope in the Franck-Condon region. To our knowledge, to date there have not been any calculations of potential energy surfaces leading to this channel. The full discussion of the present experiment will appear in a subsequent publication with details of the work performed with the deuterated ion N_2D^+ for other reaction channels. Kinetic Energy Release distribution of the collision fragments will also be presented and discussed.

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