Dissociation Cross Sections for Fast Hydrogen Molecular Ions Incident on Helium and Argon

N. V. de Castro Faria, I. Borges Jr.
Departamento de Física, Pontifícia Universidade Católica do Rio de Janeiro
Cx. Postal 38071, 22452 *Rio de Janeiro, RJ, Brasil*

L. F. S. Coelho and Ginette Jalbert
Instituto de Física, Universidade Federal do Rio de Janeiro
Cx. Postal 68528, 21945 *Rio de Janeiro, RJ, Brasil*

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We measured the transrnission of fast H_2^+ molecular ions (4.0-7.0 *a.u.*) in helium and argon targets. Total dissociation cross sections were obtained from the low-pressure region of the transmitted yield versus target pressure curve and were compared with the corresponding ones, at the same velocities and for the same targets, for H_3^+ ions. We note that a simple description is able to explain quantitatively the observed trends for both sets of experiments.

I. Introduction

The possib lity of producing atomic and molecular clusters with a chosen mass has created a new area of research, as they may be regarded either as large molecules or as fragments of condensed matter. Its implantation in solids, for example, produces a particular situation where the density of implanted nuclei could be extremely high and, consequently, new phenomena are expected. The hydrogen clusters H_n^+ ($n \geq 3$, odd), as the simplest rnolecular complexes[1], are specially important.

The study of cluster fragmentation has several motivations. In general terms the fragmentation of a cluster may show characteristics that are common to such distinct phenomen a as formation of asteroids, degradation of polymers and nuclear fragmentation. For example, the size distribiition of the fragments contains basic information on the fragmentation process and, in some circurnstances, is scale invariant[2]. Beyond this general behaviour, experimental information on the structure of H_n^+ clusters may be gathered from their fragmentation, as was already done for H_3^+ traversing thin foils[3]. Recently, Coulomb explosion after fragmentation was employed to study the fundamental building block of carbon clusters, i.e., C_3^+ [4].

Theoretical studies (see, for instance, reference 5) show that an H_n^+ cluster may have its structure described as H_2 molecules surrounding an H_3^+ ion, which is itself in the equilateral triangle configuration. Figure 1,

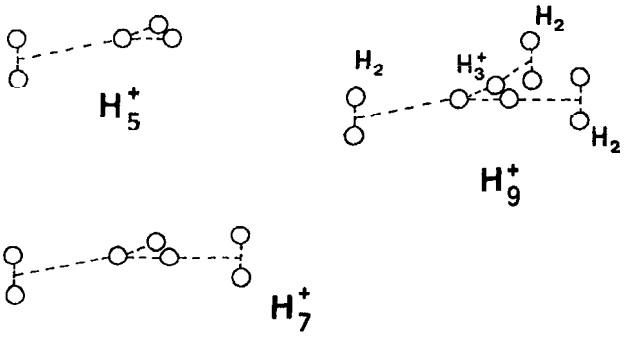

Figure 1: Structure of H_5^+, H_7^+ and H_9^+ ions, as calculated in reference 5.

taken from this reference, illustrates the H_5^+, H_7^+ and H_9^+ structures. Self-consistent-field Hartree-Fock calculations, with configuration interaction, were performed for these clusters and their energies obtained for the optimized geometries. However, this model has yet to be experimentally verified and a systematic study of the interaction of fast H_n^+ with foils and noble gases has just been started at the unique fast hydrogen cluster facility in the world, located in the IPN-Lyon[6]. In order to produce these fast ionized clusters, hydrogen gas is expanded from a reservoir at a stagnation temperature of

34 K through a conical nozzle into tlie vacuum and during the expansion a fraction of the gas condenses forming neutral clusters. These cluster bursts travel through an ionizer, where positive fragments are formed. The charged fragments are subsequently accelerated by an electrostatic field followed by a RFQ post-accelerator, corresponding to a. 5 MV potential.

As a first step in the study of H_n^+ cluster fragmentation it is interesting to analyse the high-energy fragmentation of the "basic clusters" H_2, H_2^+ and H_3^+ as function of the velocity and for several gas targets. In the Van de Graaff Laboratory of PUC-Rio we produced H_2^+ and H_3^+ ions in a radio frequency ion source and they are, subsequently, accelerated to velocities going from 2.5 to 7.0 atomic units. Concerning fast H_3^+ collisions, they have been systematically studied at this laboratory. Up to now the measurements, employing He, Ne, Ar and Xe gas targets, include the total destruction cross section[7], tlie center-of-mass distribution of H^- and H^+ ions[8], the production of H^- ions[9] and neutra1 fragments[10].

Similar systematic studies for fast H_2^+ ions would allow comparison and observation of general trends for the target Z, the projectile inolecular weight M, number of electrons n, minimum dissociation energy I and the velocity v. However, although the literature presents experimental results of collision processes at intermediate and high energies involving H_2^+ ions[11–14], and also for H_3^+ ions[7–10,14–17], few of these works deal with a wide range of energies and even less of them make a systematic choice of targets. In particular, the lack of systematic data for fast H_2^+ collisions with noble gas targets forbids comparing H_2^+ and H_3^+ data at the same velocities and colliding with the same targets, this comparison being needed in order to build models for molecular dissociation and cluster fragmentation.

For this reason, after studying the H_3^+ ion, we started now such experimental study of the H_2^+ dissociation. In this paper we present the total dissociation cross sections of H_2^+ (4.0 - 7.0 *a.u.*), the data being discussed in terms of simple models.

II. Experimental Arrangement and Methods

The experimental set-up is schematically shown in Figure 2 (a siniilar arrangement employed in this laboratory lias already been described[7]). The H_2^+ ions are produced by a radio frequency ion source, accelerated by the PUC-Rio 4MV Van de Graaff accelerator and momentum selected by a 90° magnet. The slit system limits the beam intensity to values as low as a thousand particles per second, allowing the use of a surface barrier detector for the transmitted ions.

For non-constant and intense beam currents a beam chopper could be used, with a surface barrier detector looliing at a rotating gold target and counting the Rutherford scattered (90°) particles. This arrangement was einployed for the previous[7] H_3^+ destruction measurement. The experiments described here were performed with improved accelerator conditions, allowing the assumption of constant intensity beams as will be discussed latter in this section.

The differentially pumpcd target system is composed by a gas cell 10 cm long, with entrance and exit openings of 0.5 and 2.0 mm respectively. As Figure 2 indicates, three diffusion pumps work in three different sections of the beam line, separated by two vacuum impedances.

Pressures inside the gas cell are a factor of a thousand higher than the ones outside. The cell pressure was measured employing a thermocouple device, calibrated against a high-precision Baratron capacitive manometer and the highly accurated calibration curves are shown in Figure 3. The uncertainty arising from this calibration procedure is smaller than 1%.

The scattering chamber is followed by a magnetic switch with seven exits, one at zero degrees and three on each side at the angles ±15°, ±30° and ±45°. Different fragments could be simultaneously measured at these exits, as was done in tlie references 8, 9 and 10 where the 0°, +15°, +30°, +45° and −45° exits were employed.

The H_2^+ transmission was measured choosing the magnetic field value able, for each energy, to send these ions to the +45° exit, where they were detected there by a large (25 mm diam.) surface barrier detector. For different target pressures (about ten values, the first and the last ones being the background) the number of transmitted particles was measured at fixed time intervals. The H_2^+ total destruction cross section was then obtained by fitting the transmitted fraction as an exponential function of the gas pressure (a typical result is shown in Figure 4). In order to account for possi-

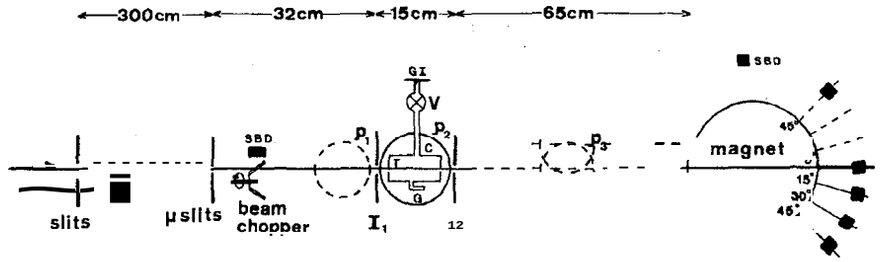

Figure 2: Scheme of the experimental set-up. The drawing is not to scale.

ble beam fluctuations several experimental runs were done - at least seven - for each target and energy value, leading to independently obtained cross section values. The final result, for a given target and a given energy, was the arithmetic average of these cross section values, their standard deviation accounting for statistical, fitting procedure and beam instability errors.

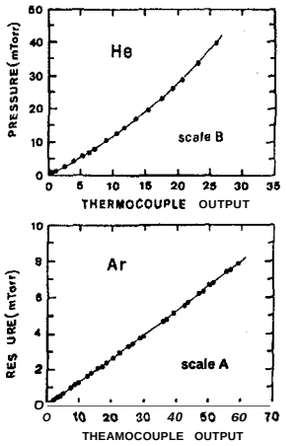

Figure 3: He and Ar pressure calibration curves.

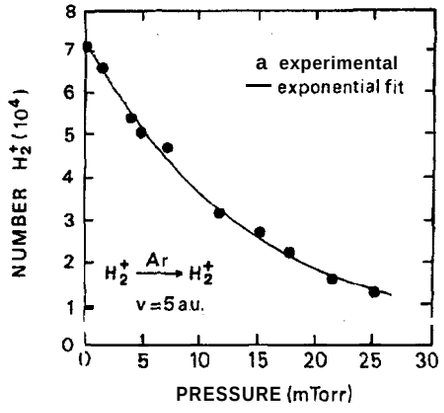

Figure 4: Typical signal versus pressure result.

III. Results and Discussion

The H_2^+ destruction cross sections for helium and argon targets, measured in this work, are presented in Table I. Previous experimental results[14], obtained for an argon target, agree very well with the present values.

Table I

H_2^+ destruction cross section for He and Ar targets.

Velocity $(a.u.)$	$\sigma_D^{H_2^+}$ $(10^{-16}cm^2)$	
	He	Ar
4.0	0.435±0.085	3.21±0.08
5.0	0.392±0.050	2.32±0.22
6.0	0.274±0.034	2.27±0.27
7.0	0.219±0.044	1.76±0.12

In order to interpret these data one may firstly look at the projectile dependence; it is useful to compare, for the same targets and velocities, the present H_2^+ data with the H_3^+ destruction cross sections. One may consider the collision as a free projectile electron moving with the projectile velocity and colliding with a *static* noble gas target[18]. A simple expression for the destruction of molecular projectiles[18], below detailed, then allows this comparison to be made and the results are shown in Table II. Secondly, one may look at the target dependence of these two collision processes. The ratio of the destruction cross sections in Ar and He, both for H_2^+ and H_3^+ projectiles, is shown at Table III.

Table II

Ratio of the norinalized destruction cross sections S for He and Ar targets and for H_2^+ and H_3^+ projectiles

Velocity (*a.u.*)	$S_{H_3^+}/S_{H_2^+}$	
	He	Ar
4.0	1.11	1.09
5.0	1.04	1.05
6.0	1.01	1.02
7.0	1.00	0.99

Table III

Ratio of the Ar and He target destruction cross sections for the projectiles H_2^+ and H_3^+

Velocity (*a.u.*)	$\sigma_D(\mathrm{Ar})/\sigma_D(\mathrm{He})$	
	H_2^+	H_3^+
4.0	6.5	6.5
5.0	7.2	7.2
6.0	7.7	7.7
7.0	8.1	8.0

The Salpeter expression[18], based itself on a Bohr model for atomic ionization[19], predicts that molecular dissociation cross sections will be inversely proportional to the dissociation energy I, as was expected from classical models. This energy is defined as the one needed to excite the molecule, placed at its equilibrium internuclear distance, from its electronic and vibrational ground state to the first dissociative electronic state. The basic assumption of this model is the free movement of the projectile components, leading to dissociation cross sections directly proportional to the number of projectile electrons n. We then defined $\sigma I/n$ as a normalized cross section value S, presented in Table II as the $S_{H_3^+}/S_{H_2^+}$ ratio. In order to reduce experimental fluctuations, the σ values were obtained from the interpolated formula $1/\sigma = a + bv^2$ as will be explained in the next paragraph. The I values employed were[18,7] 12.5 eV for H_2^+ and 19 eV for H_3^+.

A semi-empirical formula for atomic ionization[20], already applied to the H_3^+ dissociation problem after some simplifications[7], described well the cross section velocity dependence as $1/\sigma = a+bv^2$. The assumptions leading to this expression are not very different from the ones of the Salpeter model. The present cross section values (given in Table I) are similarly fitted, the results being shown in Figure 5. This model[20] describes the target atom by a simplified form factor expression and uses from the projectile only the average velocity of its electron.

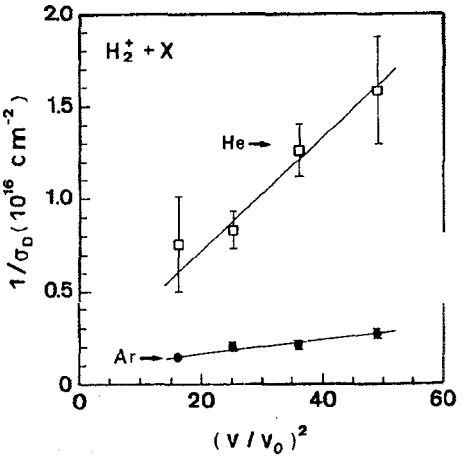

Figure 5: Experimental H_2^+ destruction cross sections for He and Ar targets, plotted as σ^{-1} versus v^2 and fitted to straight lines, as justified on a semi-empirical basis.

The most striking result of the normalization cross section procedure is that it works very well for both gases. As shown in Table II, the $S_{H_3^+}/S_{H_2^+}$ ratio of normalized cross sections is essentially equal to unity for the two targets and presents a very small velocity dependence, of the order of 10 %. The simple Salpeter approach accounts then for the main characteristics of the molecular projectiles, even neglecting the higher dissociative states and considering all molecular ions produced in the RF ion source to be in the lowest vibrational state (the vibrational excitation may lead to an internal energy around 1 eV, for a typical RF ion source[7]).

The destraction cross sections for fast molecular projectiles are expected to grow with the size of the atornic target and this has been observed to be roughly the case for H_3^+ projectiles[7]. The ratio of cross sections for Ar and He targets is presented in Table III for the two different projectiles H_3^+ and H_2^+. As expected, in all cases Ar gives much larger cross sections than He. However, we must point that, for all velocities, H_2^+ and H_3^+ data present the same target dependence. The reason for this behaviour may be that the size of the compound system formed during the collision is the relevant one and both projectiles have similar sizes (the average internuclear disfances in H_2^+ and in H_3^+ are respectively 1.1 and 0.8 Å) This size similarity may explain the lack of projectile dependence at Table III, the target dependente being due to the large difference of the He and Ar atomic sizes.

These results motivated us to compare the molecular ion destruction with the corresponding processes in the atomic systems H and H−. It seems that the model works very well for H atoms colliding with Ar, giving almost identical S values, and a reasonable agreement for a He target, which gives S values 30% larger than the molecular ones. Difficulties arise however when we try to make a simple description of H^- as a two-electron systern with an electron affinity of 0.755 eV. The experimental results of reference[21] have shown a very strong electron-electron correlation underlying the mechanisnis governing the electron loss from H−. The semi-empirical treatment of reference[20] for H and H^- electron loss also points in that direction, with the projectile orbital electron velocities that fit the model differing only by a factor of two. In fact, when we tentatively calculate the ratio of the normalized cross sections S_{H^-}/S_H for both targets, considering 0.755 and 13.6 eV a:; the respective binding energies of the ejected electroiis, we get values an order of magnitude lower than the same ratios for H_2^+ and H_3^+ ($S_{H_2^+}/S_H$ and $S_{H_3^+}/S_H$). This shows that the strong H^- electron correlation forbids the use of this simple independent-particle model for the H^- projectile.

IV. Conclusions

Total dissociation cross sections for fast H_2^+ ions (velocities in the 4.0 to 7.0 *a.u.* range) incident on He and Ar targets have been measured. The simple model proposed by Salpeter a long time ago[18] is able to reproduce well our present H_2^+ results, and also our previous H_3^+ results. In both cases the electronic wavefunctions occupy almost the same volume, and this, together with the small correlation of the H_3^+ electrons and the possibility of defining a dissociation energy, leads to a normalization procedure for the cross sections which then becomes projectile-independent. This procedure, consisting in defining a normalized cross section $\sigma I/n$, also gives a good agreement for the smaller H atomic projectiles but does not work for H^- projectiles, a large atomic ion with strong correlation between its two electrons. The target dependence of the destruction cross sections, as given by the ratio of the Ar and the He targets data, is remarkably identical for H_2^+ and H_3^+ projectiles. The absence of projectile dependence for these Ar/He ratios may be, together with the success of the simple normalization procedure here employed, a guide for more sophisticated models taking into account accurately the projectile and target form factors.

On the experimental side, in the future the destruction of H_2 molecules will be studied, for the same target gases and velocity range. These molecules are important firstly as building components of the H_n^+ clusters. Secondly, their similarity with the H_2^+ and H_3^+ ions, diatomic as the former and possessing two electrons as the latter, leads to the question whether they will behave similarly to these molecular ions.

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