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Dissociation cross sections of ionic hydrogen clusters by collision with helium at 60 keV/amu

S. Ouaskit[1], B. Farizon, M. farizon, M.J. Gaillard, E. Gerlic

Institut de Physique Nucléaire de Lyon, IN2P3 - CNRS et Université Claude Bernard,
43, boulevard du 11 Novembre 1918, 69622 Villeurbanne Cedex, France

Abstract

The dissociation of fast ionic hydrogen clusters H_n^+ by collision with helium atoms has been studied for various incident cluster masses (n = 5, 7, ...31) at 60 keV/amu energy. Absolute dissociation cross sections have been measured. No shell effect is observed. As expected, the cluster dissociation cross section increases with n but exhibits a quasi linear dependence in contrast with the $n^{2/3}$ dependence previously observed (M. Chevallier et al, Z. Phys. D2, 87 (1986) with argon atoms in the same velocity range.

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[1]Permanent address : Université Hassan II, Faculté des Sciences II,
B.P. 6621 - Sidi Othamane, Casablanca, Maroc

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I. INTRODUCTION

Mass-selected ionic hydrogen clusters of 60 keV/amu are now available up to mass 31 at the high-energy cluster-beam facility of the Institut de Physique Nucléaire de Lyon. That gives a new interest in cluster-atom collisions. Indeed, in the velocity range involved here, the relative velocity between the projectile and the target atom is around or greater than the velocity of the electrons in the cluster (≅ Bohr velocity). Then, the typical time for a collision with a target atom is short enough, compared to the typical time of the motion of the protons in the cluster, so that during the collision the protons can be considered to be stationary in the projectile frame[1]. Moreover, in this velocity range, the collision can induce large electronic excitations[2] up to ionization. Thus, processes such as ionization of the incident cluster followed by dissociation of the resulting unstable multicharged cluster can be involved.

Up to now, collisions of ionic hydrogen clusters with helium atoms have been studied at much lower velocity where molecular scattering and unimolecular dissociation are mainly involved[3]. Also, the dissociation of ionic hydrogen clusters by collision with argon atoms has been studied at 20 keV/amu for masses smaller than 25 [4].

II. EXPERIMENTAL METHOD

We report here on the dissociation of fast ionic hydrogen clusters H_n^+ by collision with helium atoms for various incident cluster masses (n=5, 7, ...31) at 60 keV/amu. Neutral clusters are formed in a cryogenic source and then ionized by electron impact. The beams of 0.18 to 1.86 MeV energies are delivered by the Cockroft Walton accelerator associated to the new radiofrequency (rf) quadrupole post accelerator [5]. The beam is pulsed with a cycle resulting from the combination of the cycle of the cluster source and of the rf power one. After momentum analysis by a magnetic field, the incident beam is defined by two collimating apertures which ensure an angular dispersion of ± 0.8 mrad. Before reaching the gas target, the beam goes through two parallel plates. A voltage which depends on the energy of the beam can be applied or set to zero between these two plates with a fast voltage amplifier. Then, the cluster bursts can either be deflected in the monitoring surface-barrier detector to measure the number of incident clusters or reach the target. The gas target is a jet which has been described in detail previously

[6]. In short, this gas jet is formed by expansion of helium in vacuum through a thin tube. Constant pressure in the gas reservoir at the inlet of the capillary is maintained by an electromagnetic valve connected to a pressure/flow controller. The gas jet is mounted on a micrometric goniometer that allows all possible translations (X,Y, Z) of the jet by steps of 50 μm. The capillary is set in order to get the maximum beam-jet overlap. During the experiments the vacuum in the collision chamber varied from 2.10^{-6} to 7.10^{-5} Torr, depending on the inlet pressure of the gas jet. A differential pumping system is able to maintain a residual-gas pressure of 5.10^{-7} Torr in the region close to the gas jet and along the beam line. The transmitted beam is magnetically analyzed one meter after the target in a detection chamber. The corresponding time of flight is 0.3 μs. The undissociated clusters are then detected with a movable surface-barrier detector. The contamination of the incident cluster beam in the range of masses 15-19 and 27-31 can be rejected by the detection system. Indeed, as previously observed[8], the signal of the surface-barrier detector is different for a molecular ion including oxygen, carbon or nitrogen from the signal obtained with an hydrogen cluster of the same mass and same velocity.

For a given mass-selected H_n^+ beam, the H_n^+ transmitted fraction F(x) is measured for various target thicknesses (x). The gas-jet thickness depends on two parameters, the inlet pressure which is measured with a precision better than 0.3% and the position of the jet capillary with respect to the incident beam which is known with a precision better than 4%. The position of the jet capillary with respect to the incident cluster beam is kept the same for the various cluster masses. The dissociation rate of the mass-selected incident H_n^+ in the beam line, by interaction with the residual gas or by spontaneous molecular evaporation, has been measured for all incident clusters. It has been found to increase with the cluster mass from 1% up to 5% of the incident beam. In all cases, the transmitted fraction F(x) is found to decrease exponentially when the target thickness x increases, as shown in fig. 1 for the H_9^+ and H_{25}^+ incident clusters. The dissociation cross section σ_n^{dis} is then deduced from the relation

$$F(x) = F(0) \exp(-\sigma_n^{dis} x) .$$

The determination of the target thickness and tests of the absolute calibration are described in ref. 6. The errors in the cross section values (±8%) arise mainly from the uncertainty in the target thickness and from the fluctuations of the beam intensity.

III RESULTS AND DISCUSSION

The dissociation cross sections for the H_n^+ clusters, n ranging from 5 to 31, at 60 keV/amu (1.55 Bohr velocity) are presented in fig. 2. The value obtained for H_3^+, $\sigma_3^{dis} = (2.5 \pm 0.2)\ 10^{-16}\ cm^2$, is in agreement with the value $(2 \pm 0.3)\ 10^{-16}\ cm^2$ linearly extrapolated from the only available data[7] at higher energy (>120 keV/amu).

The ionic hydrogen clusters are described as a nucleation of H_2 molecules surrounding an H_3^+ core[9,10]. Then, the cluster dissociation cross sections should be compared to the dissociation cross section of an H_2 molecule, but only the cross section of the single ionization of H_2 molecules by collision with helium atoms at 60 keV/amu ($\sigma_2^{ion} = 1.5\ 10^{-16}\ cm^2$) is available in the literature[11]. The quantity $[\sigma_3^{dis} + ((n-3)/2))\ \sigma_2^{ion}]$ is plotted (dashed line) versus n in fig. 2. The experimental data lie above this line. One might have expected the cluster dissociation cross sections to be even much larger than this quantity since ionic hydrogen clusters are very weakly bound. Thus, one would deduced that the ionization process seems to play an important part in the cluster dissociation. Nevertheless, one has to notice that

- the quantity $\sigma_3^{dis} + ((n-3)/2))\ \sigma_2^{ion}$ is rather simplistic to describe the cluster ionization

- the H_2 molecules in the cluster are in the lower vibrational state[12] while the H_2 ionization cross section has been measured with mainly excited H_2 molecules since the neutral beam was obtained by charge exchange from an H_2^+ beam. Then, this H_2 ionization cross section used for comparison is probably too large for the H_2 molecules of the cluster.

As expected, the dissociation cross section increases with the mass of the cluster. While shell effects have been predicted in the theoretical description of the structure of the ionic hydrogen clusters[10] and have been observed in the dissociation-energy measurements[13], no shell effect is observed in the present results. Such fine effect connected to a small energy gap ($\approx$ 0.1 eV) between open and closed shells is probably hidden by the large range of impact parameters and energy transfers which can lead to the dissociation. For n >9, the dissociation cross section increases proportionally to n . The $n^{2/3}$ dependence previously observed in the same velocity range with argon atoms[4] is not observed (see fig. 2). The linear dependence of

the dissociation cross section by collision with helium versus n seems to show that the H_2 molecules in the cluster contribute in an equivalent way to the dissociation, independently of their position in the incident cluster. This must be connected to the fact that these small clusters cannot be approximated by spherical droplets[10,12] and is probably due, as previously assumed, to an effect of average over the various impact parameters. Nevertheless, specific cluster effects should be observed in the analysis of the various dissociation channels.

To summarize, we have completed systematic measurements of dissociation cross sections of ionic hydrogen clusters at high energy, for a large range of masses. We have found a different behaviour compared to previous measurements at lower energy or with other type of gas target.

III. ACKNOWLEDGMENTS

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FIGURE CAPTIONS

Figure 1: Normalized H_{25}^+ and H_9^+ transmitted fraction F(x)/F(0) versus x the target thickness.

Figure 2: H_n^+ dissociation cross sections versus n, at 60 keV/amu. The dashed line displays the quantity $[\sigma_3^{dis} + ((n-3)/2))\ \sigma_2^{ion}]$.

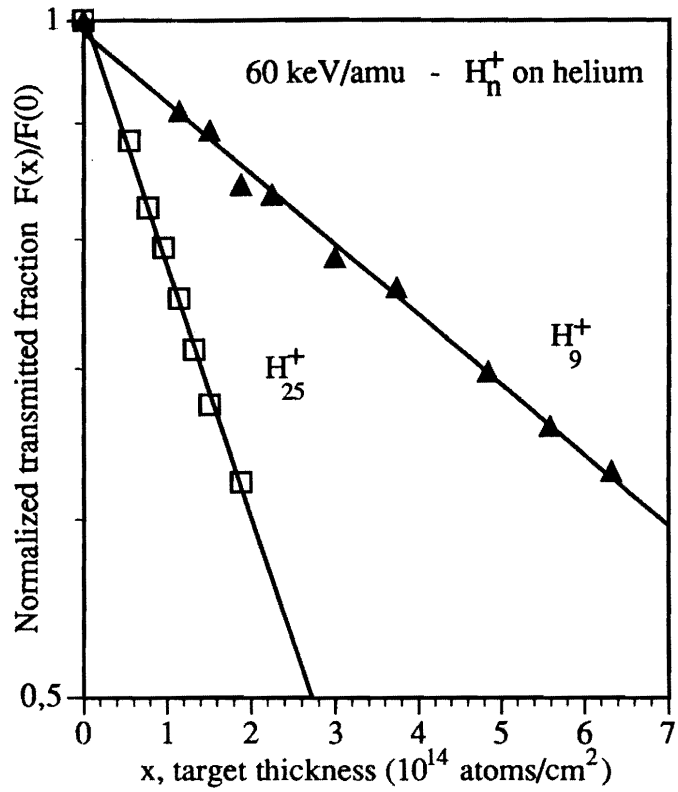

Figure 1 : S. OUASKIT et al.

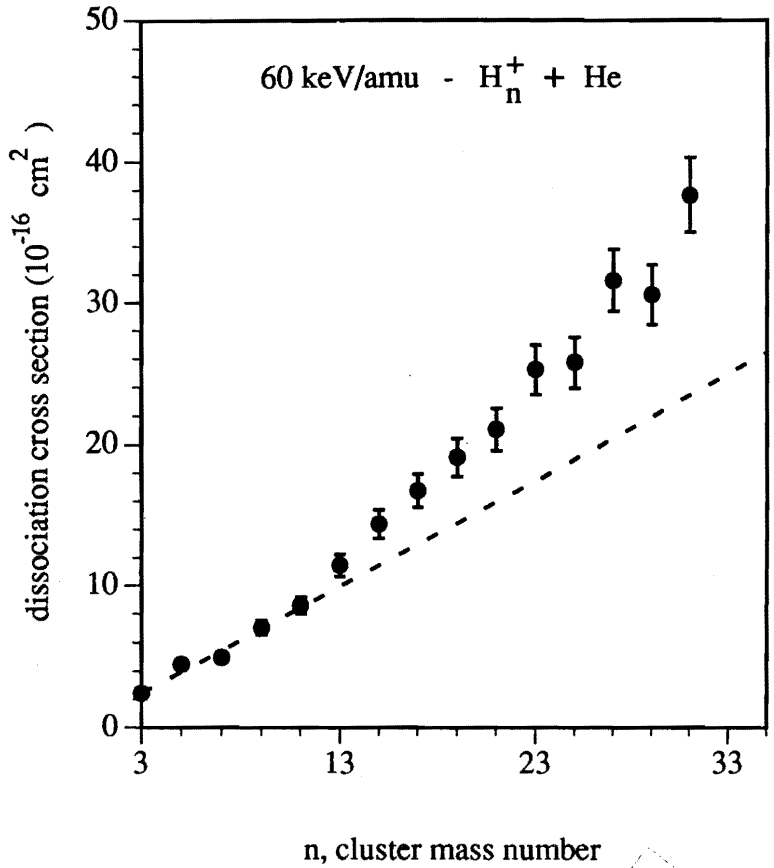

Figure 2 S. OUASKIT et al.