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EXPERIMENTAL STUDY OF THE DISSOCIATION OF 100-600 keV HYDROGEN CLUSTER IONS IN AN ARGON GAS TARGET(*)

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ABSTRACT

We have studied the break-up of accelerated hydrogen cluster ions passing through an argon gas target. The absolute dissociation cross section has been measured for a wide variety of H_n^+ (odd masses only) cluster ions, with n between 5 and 23 and with projectile velocities ranging from 1.5 to 5 x 10^8 cm/s. We discuss the dissociation processes and the dependence of their cross-sections upon the cluster mass and velocity.

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Since the 1960's, the production, the stability and the properties of molecular clusters have been the subject of many experimental and theoretical studies. The main reason for this interest is that the study of these clusters may help filling the gap between molecular and solid state physics.

Hydrogen clusters are of special interest, for both experiment and theory, because of their relative simplicity. All the light hydrogen clusters that have been produced and observed up to now are singly charged species of odd mass numbers [1,2], and their stability, according to the theoretical predictions, is due to the clustering of H_2 molecules around a positive ionic core, H^+ or H_3^+. The calculations give predictions about the structure, the binding energy and the vibrational frequencies of the lightest clusters H_5^+, H_7^+ and H_9^+.

We report here on experimental results obtained with fast analysed H_n^+ clusters. We measured the transmission of light hydrogen clusters through an argon gas target, for projectile velocities between 1.5 and 5 x 10^8 cm/s, i.e around and above the Bohr velocity v_0($v_0 = e^2/\hbar$). We have deduced the dissociation cross sections σ_d from our measurements. The knowledge of these cross sections is not only useful for evident experimental reasons but also because the velocity dependence of σ_d for a given cluster can help understanding the break up process and the variation of σ_d with n, the cluster proton number, at a given projectile velocity, yields information on the cluster structure.

The mass analysed pulsed H_n^+ beam (burst duration of ~ 60 ms with a repetition rate of ~ .2 Hz) was delivered by the 1 MV cluster accelerator of the Institut de Physique Nucléaire de Lyon [3]. This accelerator is composed of an 1 MV open air cascade, a cluster beam source, an ioniser consisting of a transverse electron beam and a high-gradient accelerator tube (figure 1). The accelerated ion cluster beam was analysed in energy and mass by the association of an electrostatic (E) and of a magnetic (B1) analyser. The selected H_n^+ beam (with n ⩽ 23) was tightly collimated by C_1 and C_2 before entering a gas target filled with argon gas at variable and controlled pressure. The beam transmitted through the target was then magnetically analysed by B2. The surviving H_n^+ cluster ions (i.e those with the mass and charge of the incident projectiles) are detected by the surface barrier detector D_2 whereas the detector D_1, located on the beam axis, was used to detect the neutral species of the transmitted beam.

At this point one must consider that beside dissociation two other collisional processes could prevent an incident H_n^+ ion from reaching D_2. First it could be scattered, without being destroyed, and miss the detector. Scattering of hydrogen clusters has been experimentally studied [4,5] at much lower velocities. However we estimate that, in our conditions, scattering without dissociation can occur only at angles that are much smaller than the angular acceptance of D_2. Secondly it could capture a target electron to form the neutral cluster H_n. The detector D_1, mainly devoted to the possible observation of stable H_n clusters, did not reveal the existence of this process. A very important consequence is that our experiment deals in fact with *dissociation, the only process that attenuates the incident H_n^+ beam.* The number of transmitted H_n^+ ions was measured for a given number of beam bursts. The stability of the beam intensity was checked by repeating the measurement for a given target thickness, the target pressure being increased then decreased step by step. The experimental procedure, including the calibration of the gas target thickness, was checked by observing the dissociation of H_2^+ and H_3^+ projectiles and we found our data to be in very good agreement with data obtained in a previous study performed with a Van de Graaff Accelerator in our laboratory [6].

In all cases the transmitted fraction $I(x)/I_o$ was found to decrease exponentially when the thickness x increases, as shown for example in figure 2 for 12 keV/u H_{19}^+ ions. We can then derive the dissociation cross sections σ_d from the relation $I(x) = I_o \exp(-\sigma_d x)$.

We have measured the dissociation cross sections of H_n^+ clusters in the range of masses and velocities allowed by our experimental set-up. These data are presented in fig. 3 and fig. 4. Figure 3 shows the variation of σ_d with n for five values of the projectile velocity. As expected and discussed later σ_d increases with n. In figure 4, the velocity dependence of σ_d is shown for n = 5, 7, 9 and 11 clusters (the only clusters we could study for a significant range of velocities). These data show that the dissociation cross section seems to be independent of the cluster velocity in the region of v_o, the Bohr velocity, and decreases when the velocity increases above v_o. From these features and from what is known of atomic and molecular collisional processes one can try to deduce which processes are involved in the break up of these clusters in this velocity range.

Experimental studies [7, 8] of the collisional processes experienced by 5 to 30 keV H_2^+ ions incident on a xenon gas target have shown that the dissociation cross section depends very little on the velocity and that the main dissociation process is the dissociative capture of a target electron $H_2^+ + e^- \rightarrow H^o + H^o$. It is then reasonable to think that the target electron capture is also responsible for the dissociation of H_n^+ in the same range of velocity. This is even more specially true here because all electron capture events lead to dissociation (which is confirmed by the non observation of neutral clusters H_n, as mentioned earlier), contrary to the H^+_2 case where bound states can obviously be formed. It is worth noting that our group had *earlier proved that, except a negligible fraction of long-lived states* [6], the H_3 molecule is unstable.

The observed decrease of σ_d when the velocity exceeds v_o, is too moderate to be conclusive. However it is quite compatible with the v^{-1} dependence that has been observed in the dissociation of H_2^+ and H_3^+ of higher velocities [6]. It has been shown [9] that H_2^+ ions of velocities equal to a few times the Bohr velocity dissociate via two collisional processes : the projectile electron loss and the electronic excitation into a repulsive molecular state. The same processes are probably responsible for the break up of H_n^+ projectiles in the same velocity range if one considers that their structure is too fragile to survive the removal or the excitation of one of its electrons.

The only other published result concerning the hydrogen cluster dissociation is, to our knowledge, the work of Van Lumig and Reuss [4] on very slow clusters. From their data, obtained at cluster energies of 10 to 200 eV/u one can extract that, for a given cluster, the dissociation cross section increases with energy. This can be qualitatively understood if one considers the collision in the cluster frame of reference. At these very low velocities the energy loss of the argon projectile in the cluster target increases also with energy and is dominated by the so-called nuclear energy loss, due to elastic collisions that result in vibrational excitation of the cluster. It is then reasonable to assess that vibrational excitation is responsible for the dissociation of very low energy clusters. It is quite probable that vibrational dissociation contributes also in our energy range, along with electron capture or loss. It is clear that the above discussion is rather speculative [illegible] ut more experimental work, bearing particulary on the stud[illegible] the dissociation fragments would be needed to identif[illegible] dissociation processes.

In order to get a more detailed view on the dependence with n of the dissociation cross section we took advantage of its quasi-independence with respect to the velocity to join the cross sections measured for H^+_{21} and H^+_{23} at 12 keV/u to those measured at 20 keV/u. This set of data is presented in fig. 5. If one excepts the σ_d values obtained for n = 15 and 19, the variation of σ_d with n is very well fitted by the law $\sigma_d = Kn^{\alpha}$ with $\alpha = .66$, i.e. $\alpha = 2/3$ and $K = 8.65$ Å^2, as shown on fig. 5. Our analysis is the following :

As the cluster orientation with respect to the beam direction is randomly distributed, we may consider that the clusters are "statistically" spherical as far as the dissociation is concerned.

As a consequence, the observation of a $n^{2/3}$ dependence of the dissociation cross section reflects the invariability of the cluster compactness as n varies, just like the $A^{1/3}$ dependence of the nuclear radius reflects the invariability of density of nuclear matter. The lower values of the dissociation cross section for H^+_{15} and H^+_{19} could be due to more highly bound structures. However they would need to be confirmed by further experiments.

As to the value found for K, it leads to a density of 0.052 atom/Å^3 if one makes the hypothesis that the dissociation cross section is equal to the geometrical cross section of the "spherical" clusters. This value is in good agreement with the previous estimate by Van Lumig and Reuss [4] (0.025 H_2/Å^3) and compatible with the density of liquid hydrogen (0.042 atom/Å^3)% This noticeable result can be easily understood for larger clusters but is more surprising for small clusters where the geometrical cross section of the argon atoms is not negligible.

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FIGURE CAPTIONS

Figure 1 — Schematic description of the Lyon cluster accelerator and of the experimental set-up.

Figure 2 — Variation of the transmitted fraction $I(x)/I_0$ of H^+_{19} clusters with argon target thickness.

Figure 3 — Absolute dissociation cross sections σ_d for H^+_n (n = 2 to 23) clusters in argon gas at energies ranging from 12 to 120 keV/u.

Figure 4 — Velocity dependence of the dissociation cross section σ_d for H^+_5, H^+_7, H^+_9 and H^+_{11} clusters in argon gas.

Figure 5 — Mass dependence of the dissociation cross section of 20 keV/u H^+_n clusters in argon. The dashed line corresponds to a fit with a $n^{2/3}$ law (see text).

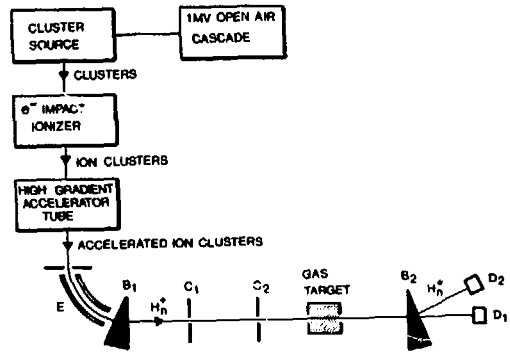

FIGURE 1

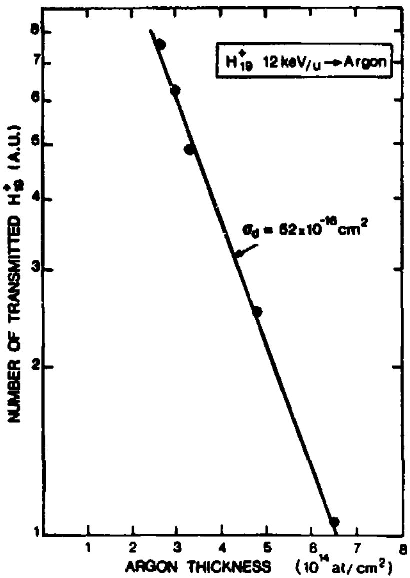

FIGURE 2

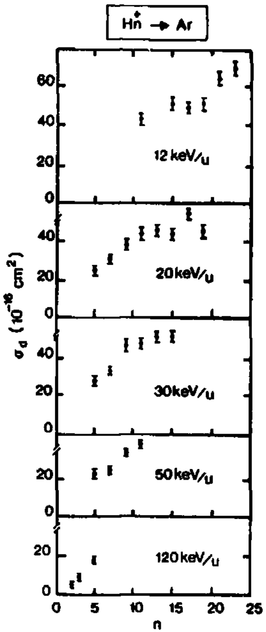

FIGURE 3

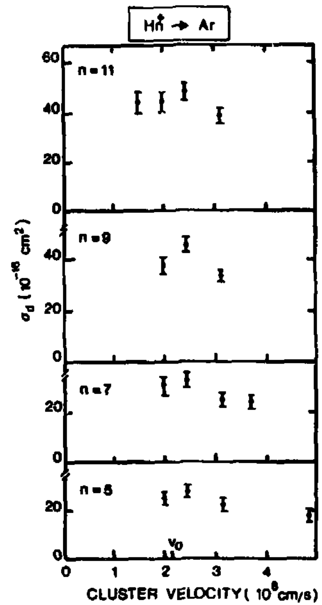

FIGURE 4

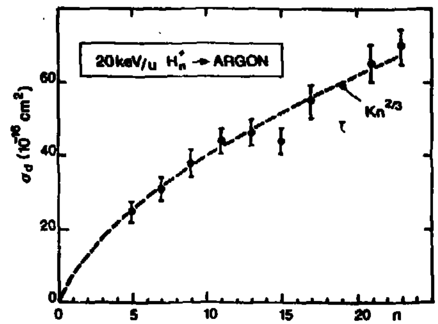

FIGURE 5