Enhancement of Lyman-α radiation following foil-induced dissociation of fast ionic hydrogen clusters H_n^+

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We have measured the Lyman-α radiation following foil breakup of hydrogen ionic clusters H_n^+ ($n=2$ and $n=3$ to 61, odd) with velocities above and around the Bohr velocity. An enhancement of this radiation was observed and could reach a factor of 3 with respect to the proton case of the same velocity. Cluster mass number, velocity, and thickness dependences of the relative population of the $2p$ state in hydrogen fragments following H_n^+ foil dissociation have been extracted. A specific collective effect on the $2p$-state hydrogen has been observed and interpreted in terms of charge-exchange processes.

I. INTRODUCTION

In recent years, molecular projectiles have been extensively used since they offer very specific possibilities for studying the basic aspects of atomic collisions in solids. The dissociation of the incident molecular projectiles is observed and electron-capture and -loss processes may be considered in the framework of repulsive molecular states.

Most experimental studies have dealt with charge or excitation states of molecular projectiles after dissociation in a thin solid target. Astner, Mannervik, and Veje[1] have performed beam-foil experiments in which they observed molecular effects in the formulation of excited states of transmitted atomic fragments. Brooks and Berry[2] have measured the carbon foil's thickness dependence of the Lyman-α ($1s$-$2p$) yield with H_2^+ and H_3^+ incident molecular ions. Clouvas *et al.*[3] have performed experiments in which beam-foil spectroscopy is combined with charge-state measurements, for molecular hydrogen projectiles. The evolution of the relative population of a given excited state of atoms (H^0) or ions (He^+) emerging from thin carbon foils bombarded with 1-MeV/amu molecular (H_2^+,H_3^+) beams has been observed in equilibrium and nonequilibrium situations that are known to occur in charge-exchange processes. Other experimental studies[4-8] have also revealed molecular effects in the formation of the excited state of transmitted atomic fragments.

In our laboratory, we were able to accelerate and analyze hydrogen clusters H_n^+ ($n=2$ and $n=3$ to 61, odd) with energies as high as 630 keV. In previous work[9] we have already measured the angular and charge distribution of hydrogen fragments resulting from the dissociation of fast H_n^+ clusters ($n \leq 21$) in a carbon foil and studied the velocity and n dependence of the emergent neutral fraction. When hydrogen ion clusters go through a condensed medium most of their electrons are immediately stripped off and the binding between the atoms of the clusters is disrupted. The ionic fragments are repelled from each other by a screened Coulomb interaction while they fly through the solid. When they emerge from the exit surface of the foil some fragments bind electrons that may originate from the incident projectile or that have been picked up from the target. From experiments performed with 1-MeV H_2^+ and H_3^+ beams, we know that the first process is dominant in the case of very small dwell times in the foil and is directly related to the transmission probability of the projectile electrons.[10] In the "pick-up" regime corresponding to thicker foils, the close proximity between the fragments at the exit surface tends to decrease their average final charge state.[11,12] With hydrogen clusters, we have already observed that protons have an angular distribution wider than hydrogen atoms, despite the fact that a cluster travels inside the foil as a swarm of individual protons, the volume of which increases with the penetration depth. Due to multiple scattering and energy straggling, the fragments progressively become randomly distributed both inside this volume and, in particular, at emergence, where their mutual separation settles the proximity effects on neutralization. The explanation could be searched for only beyond the foil; we therefore deduce that protons continue to repel each other in vacuum whereas, in a first approximation, the hydrogen atoms do not interact any more with the fragments. At these velocities, the angular distributions of hydrogen atoms thus reflect essentially the combined effect of multiple scattering and screened Coulomb repulsion in the foil. From the angular distributions one deduces the fraction ϕ_0^n of neutral atoms and its variation with n by integrating the angular distributions over the total volume of the explosion cone. One observes that

the neutral fraction increases first linearly with n, at least for the two low velocities studied (30–40 keV/proton), then tends to saturate beyond $n \sim 7$. This linear dependence was already observed for H^+, H_2^+, and H_3^+ and means that the average distance between the fragments at the exit surface of the foil is nearly the same whatever the cluster mass for a given velocity. In other words, the proximity effect which enhances the neutral production is proportional to the number of protons. This enhancement is smaller when the velocity is lower, essentially because the neutral fraction for incident protons is already very high at these low velocities and also because the average distance between the fragments increases when the velocity decreases. It is well known that the charge-exchange processes of correlated atomic projectiles are affected by the correlation.

In order to learn about collective effects in charge-exchange processes, we have investigated (in the present work) the velocity and mass number dependence of the relative population of the hydrogen $2p$ state following dissociation of hydrogen ionic clusters, which have not been studied to date.

II. EXPERIMENTAL PROCEDURE

In order to determine the relative number of $2p$ excited states among the H^0 fragments resulting from the passage of fast H_n^+ clusters through a thin carbon foil, we have measured the light emission from the Lyman-α transition. The mass-energy analyzed beams H_n^+ with $n=2$ and $n=3$ to 61 (odd number of protons), at incident energies of 10, 20, 30, 40, 60, and 80 keV/p, were delivered by the Institut de Physique Nucléaire de Lyon (France) accelerator. This accelerator is composed of a 630-kV (maximum) open air cascade, a cluster beam source, an ionizer consisting of a transverse electron beam and a high gradient accelerator tube. The clusters are initially produced by expanding hydrogen gas from a stagnation temperature of 34 K through a conical nozzle into the vacuum. During the expansion a fraction of the gas condenses in order to form clusters, while the major part is pumped on the cooled cryosurfaces which radially surround the beam. The cluster bursts last approximately 60 ms with a repetition rate of ~ 0.2 Hz. These bursts travel through an ionizer where positive cluster ions are formed by electron impact. After acceleration, the ion cluster beam is analyzed in energy and mass by the association of electrostatic and magnetic analyzers.

Figure 1 shows the experimental setup. The selected H_n^+ beam is tightly collimated by two apertures before reaching the solid target. Four carbon foils could be set on a target holder fixed on a goniometer that allows all possible translations of the targets. A double carbon foil made with two carbon foils separated by 1 mm was also put on the target holder. The purpose of this double foil system is to simulate a proton beam with the same analysis system configuration. The cluster projectiles explode in the first foil and the distance between the resulting fragments when they reach the second foil is sufficiently large that they behave like isolated protons (as far as charge exchange is concerned). The foils are thick enough to imply that the H^0 emergent species are protons having picked up a target electron (pickup regime). In our velocity range the negative ion H^- fraction is negligible and we may consider that the only emerging projectiles are H^0 and H^+ ions (molecular and cluster transmitted fractions were also negligible).

The transmitted beam current is measured with a Faraday cup equipped with an electron suppressor ring (S_1). In order to protect the target from the electrostatic field due to the high voltage of the suppressor ring, a metallic ring (S_2) connected to the mass is set between the target and the suppressor ring. The angular aperture of the device is about 16°. With this argument, we can deduce the number of incident particles after integration and correction of the neutral fraction previously measured[9] for these velocities and target thicknesses. The single targets

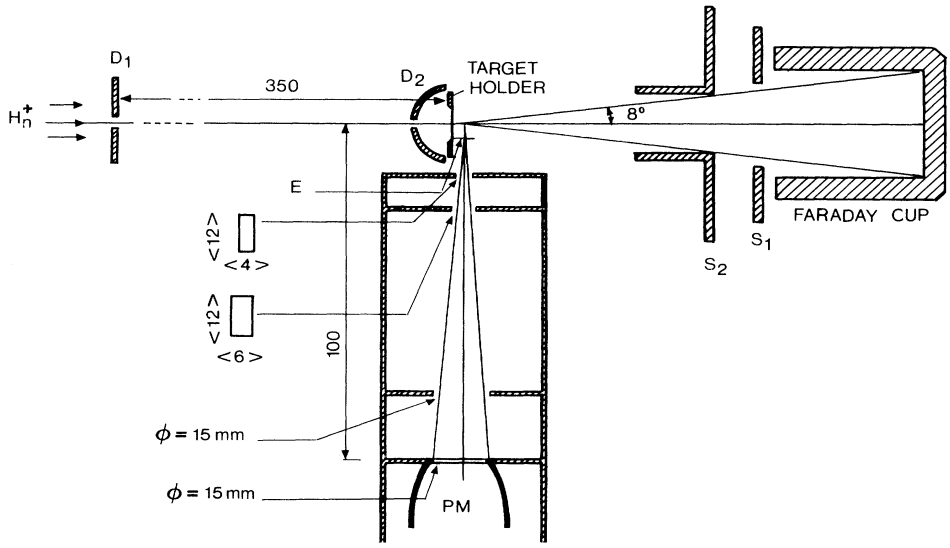

FIG. 1. Experimental setup (see text for details). Lengths are in mm.

have thicknesses of 2.1 ± 0.2 and 3.8 ± 0.4 $\mu g/cm^2$. The pressure in the chamber was better than 10^{-6} Torr.

A slit (E) placed at 2 mm from the beam direction defines the region of observation of the photomuliplier (PM) G-26E215, that has a spectral response curve centered on the Lyman-α wavelength (120 nm). The sapphire window has a wavelength cutoff lower than 105 nm that eliminates the Lyman-β, -γ lines. The tube of the PM is solar blind that eliminates the Balmer series. Three collimators placed in front of the photomulplier tube avoid edge effects from the holes drilled through the plates for the passage of the beam. The photomulplier has been placed 100 mm from the beam direction in order to optimize the solid angle of detection. The photomultiplier signal corresponding to the Lyman-α radiation coming from the beam was at least 100 times larger than the noise. The noise we consider is the photomultiplier signal when the beam passed through the target holder without carbon foil.

All the experiments were done with the photomultiplier looking at a region of the beam downstream of the foils, with a fixed angular aperture and at a fixed distance from the foil. These conditions were chosen in order to optimize the use of the accelerator time. The fixed distance does not need velocity correction since the data were always normalized to the proton case at the same velocity, i.e., double foil measurements. For each cluster mass and each velocity, the photon number emitted by the excited hydrogen atoms downstream of the single foil and the double foil was measured.

III. RESULTS AND DISCUSSION

Previously experiments have shown that the angular spread due to the multiple-scattering effects and the molecular breakup in the carbon foils does not affect the photon measurements. In Fig. 2, we have reported the Lyman-α photon yield obtained with 40 keV/p H_n^+ ($n=3$ to 15, odd) clusters impinging the double carbon foil for a fixed number of protons measured by a Faraday cup downstream of the foil. We observe that this yield is independent of the cluster mass number H_n^+ and equal to the one obtained with a 40-keV proton beam.

In order to determine the cluster effect in the population of the $2p$ state in H^0, excited after passage of H_n^+ clusters through carbon foils, we extracted the ratio $R_{2p}^n = N_{2p}^n / N_{2p}^1$, from measurements of N_{2p}^n and N_{2p}^1 Lyman-α intensities per incident proton, obtained by using cluster projectiles H_n^+ ($n=3$ to 61, odd) and protons of the same velocity, respectively (see Fig. 2). The atomic beam was obtained as mentioned above by employing the "double foil" technique.

The ratio R_{2p}^n versus the cluster mass number ($3 \le n \le 61$) is presented Figs. 3–5, for various projectile velocities (10–80 keV/p) and for carbon foil thicknesses of 2.1 and 3.8 $\mu g/cm^2$.

We generally observe an increase of the "cluster effect" R_{2p}^n (a) with the cluster mass number n for given projectile velocity and target thickness, (b) with the projectile velocity for given cluster mass number and target thickness, (c) with decreasing target thickness for a given projectile velocity and cluster mass number n. Indeed, for very thick targets, the average distance between the fragments of the cluster at the exit surface is so large that they behave like atomic projectiles as regards charge-exchange processes.

Up to now, there are no similar data available in the literature for incident H_n^+ ($n \ge 5$). However, for incident H_3^+ ions, by plotting our R_{2p}^3 values as a function of the projectile energy we observe, Fig. 6, that our results agree with the one obtained by Baudinet-Robinet and Dumont[8] with incident H_3^+ ions impinging on a carbon foil of the same thickness (~ 4 $\mu g/cm^2$).

The observed "cluster effect" R_{2p}^n could be simply due (in a first approximation) to an overproduction of emerging neutrals (in all nl states) per incident proton when incident clusters H_n^+ are used instead of a proton beam. In a previous experiment[9] we had deduced this overproduction by measuring the ratio $R^n = \phi_0^n / \phi_0^1$, (where ϕ_0^n and ϕ_0^1 are the neutral fractions for incident H_n^+ clusters and

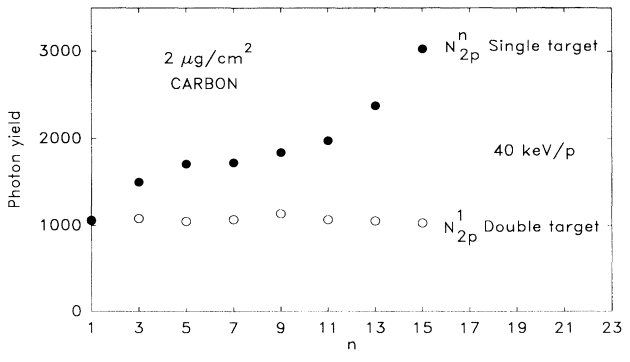

FIG. 2. Variation with n, the cluster mass number, of the number of $2p-1s$ photons emitted by emerging hydrogen fragments resulting from the collision of 40-keV/p H_n^+ ($3 \le n \le 15$) clusters with a single carbon foil (●) and a double carbon foil (○).

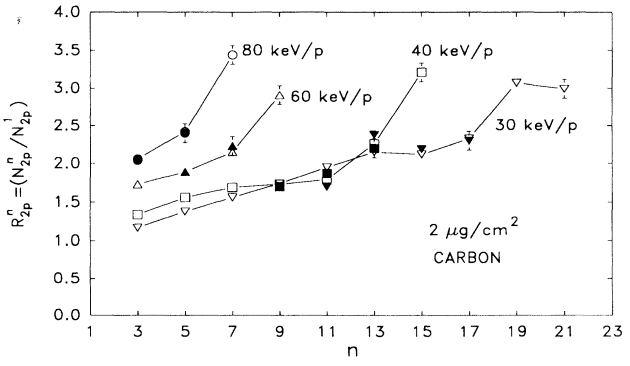

FIG. 3. Cluster mass number dependence of R_{2p}^n for various projectile energies (30, 40, 60, and 80 keV/p) impinging on a 2.1-$\mu g/cm^{-1}$ carbon foil. Open symbols correspond to results obtained with the slit E (prompt decay), and filled symbols with the slit E' (delayed decay).

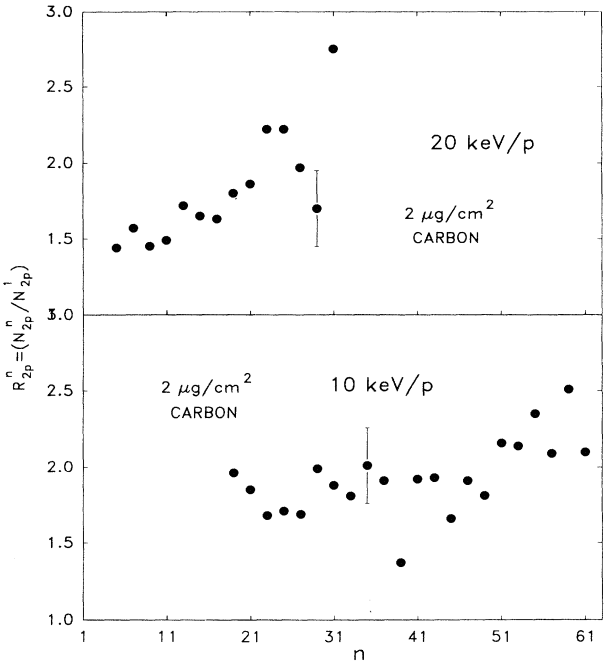

FIG. 4. The same as for Fig. 3 for projectile energies 10 and 20 keV/p.

a proton beam of the same velocity, respectively), obtained with H_n^+ clusters of the same velocities and carbon thicknesses as in the present experiment. In Figs. 7 and 8 we present the ratio $P_{2p}^n = R_{2p}^n / R^n$ for two target thicknesses (2.1 and 3.8 μg/cm^2) and various projectile velocities (30, 40, 60, and 80 keV/p). It is generally observed that for a given velocity, cluster mass number, and target thickness, the ratio P_{2p}^n is always smaller than the ratio R_{2p}^n but still greater than unity, which means that a cluster effect specific to the $2p$ state has also to be considered. In addition, we observe in Fig. 7 that, for a given projectile velocity, this specific $2p$-state cluster effect P_{2p}^n is constant for small cluster mass numbers and increases suddenly for higher cluster mass numbers, the

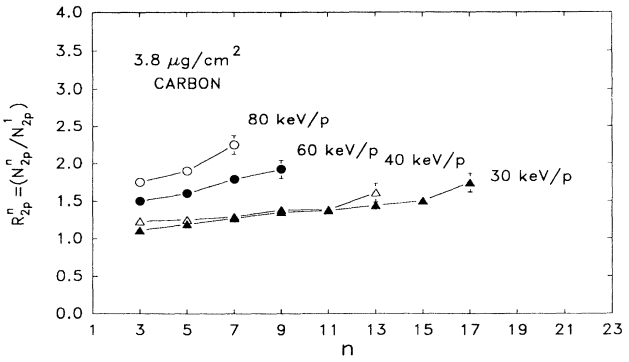

FIG. 5. The same as for Fig. 3 with a 3.8-μg/cm^2 carbon foil.

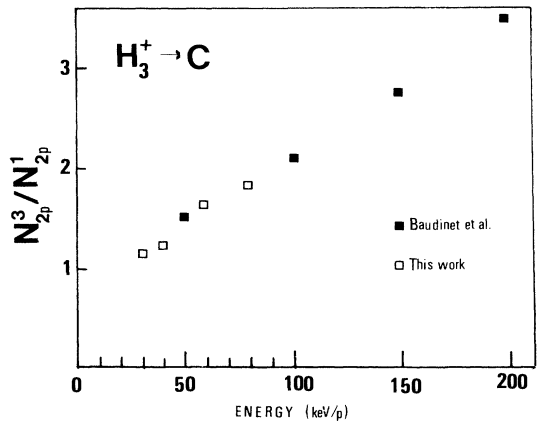

FIG. 6. Projectile energy dependence of $R_{2p}^3 (= N_{2p}^3 / N_{2p}^1)$ at emergence from a 3.8-μg/cm^2 carbon foil. The experimental results obtained by Baudinet-Robinet and Dumont shown for comparison are taken from Ref. 8.

threshold depending on the projectile velocity.

The specific cluster effect of the $2p$ state could be due to a proximity effect on charge-exchange processes inside the foil, or to an effect outside the foil such as the superradiance phenomenon.[13] It is well known that the normal decay of isolated atoms can be notably altered when the number of radiators in that sample become large enough. The collection from atoms then starts to radiate spontaneously much faster and stronger than the emission from independent atoms. To test this possibility we have performed two sets of experiments at 30, 40, 60, and 80 keV/p, where the photomultiplier looks at a region (slit E') of the beam far from the region usually studied (slit E). In Fig. 3, where we have reported the two sets of data with the slit E' and the slit E, no systematic deviation is observed. Probably such effects are too small to be observed with our experimental setup and it means that the collective effect observed is principally due to a prox-

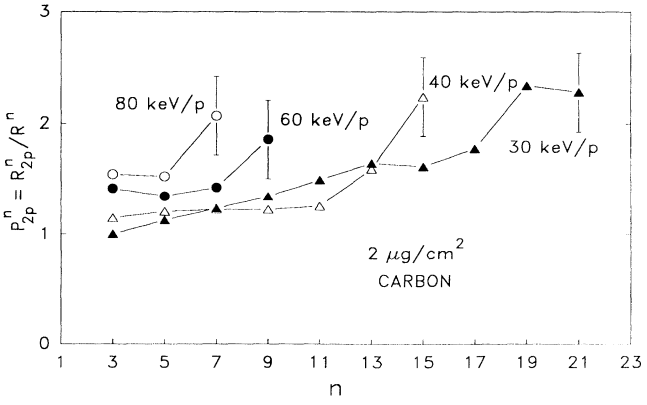

FIG. 7. Cluster mass number dependence of $P_{12p}^n (R_{2p}^n / R^n)$ various projectile energies (30, 40, 60, and 80 keV/p) impinging on a 2.1-μg/cm^2 carbon foil.

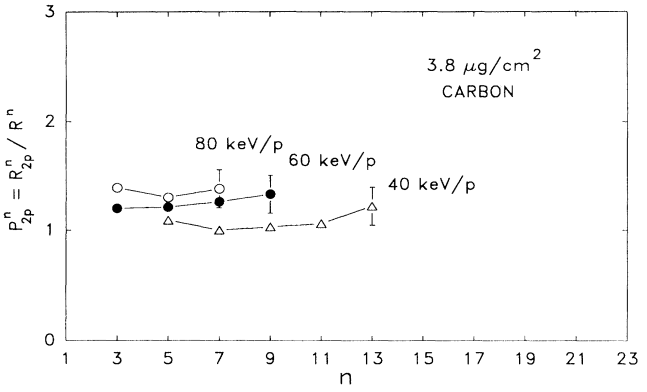

FIG. 8. The same as for Fig. 7 with a 3.8-$\mu g/cm^2$ carbon foil.

imity effect on charge-exchange processes inside the foil. Then, the P^n_{2p} ratio represents the relative population of the $2p$ state in the emerging H^0 fraction, obtained with H_n^+ incident clusters and normalized to unity for an incident atomic beam.

In a previous paper,[14] in order to explain the vicinity effects observed in the charge-exchange processes on atomic fragments resulting from the foil dissociation of H_2^+ ions at 30–1000 keV/p, we have considered that the electron-capture cross section is not affected by vicinity effects but that the electron-loss cross section is reduced due to the increase of the electron binding energy in H_2^+ compared to the H atom. Turning to $2p$-state results, the emerging H^0 atoms excited in the $2p$ state from the carbon foil dissociation of H_n^+ clusters with 10–80 keV/p may be produced by a direct (electron) capture of a target electron by a proton (originating from the dissociation of an incident cluster) in the last layers of the foil or by a two-step process. This last process involves capture of a target electron by a proton (surrounded by closest neighbors) in the $1s$ state and excitation of the resulting H^0 atom into the $2p$ state in the last layers of the foil. The "last layers" hypothesis originates from the fact that at these velocities (40–80 keV/p) the only hydrogenic state that can be bound in the foil, due to the screening of the projectile potential by the target electrons, is the $1s$ state. However, at the exit surface of the foil, the decrease of the electron density allows the $2p$ state to be bound again.[15]

From these considerations, the cluster effect R^n_{2p} (Figs. 3–5) as a function of the cluster mass, velocity, and thickness can be explained as follows. Increasing the projectile velocity reduces the repulsion time between the fragments (for a given target thickness) but increases the dynamical screening length and then the repulsion force. Increasing velocity also reduces the multiple-scattering effect. Indeed, from H_2^+ charge-exchange results,[14] we know that increasing velocity increases the cluster effect since it reduces the fragment separation at the exit of the foil. As for H_2^+, for a given cluster mass number, increasing velocity increases the electron binding of such a "cluster state," i.e., reduces the loss cross section and tends to increase the number of neutral fragments at the exit surface. Moreover, for a given projectile velocity, increasing the cluster mass number increases the electron binding energy, reduces the loss cross section, and tends to increase the number of atomic hydrogen emerging fragments. In fact, we observe that the ratio N^n_{2p} increases with velocity for a given cluster mass number and with cluster mass number for a given projectile velocity (Figs. 3 and 4). Moreover, increasing the target thickness increases the proton separation at emergence and then lowers the cluster effect (Fig. 5).

Concerning the specific $2p$ state with respect to all states of the emergent hydrogen, we suggest the following ideas to explain the results: since, as mentioned above, the $2p$ hydrogen state cannot be bound inside the foil (at low velocities) and is formed either by direct capture at the exit surface or from excitation of the $1s$ hydrogen state formed inside the foil. But we have observed that increasing velocity or increasing cluster mass number are two similar ways of reducing the loss cross section. In previous work,[16] we have built up a model which predicts whether the bound state of an emergent ion may have been formed inside the solid target or only at emergence. In the framework of this model, we have shown that the $He^+(3p)$ state can be bound inside solid carbon if the ion velocity is high enough and we have observed a velocity threshold. The discontinuity observed on the cluster mass dependence of the ratio P^n_{2p} reflects similar behavior, i.e., for high cluster mass number, the hydrogen $2p$ state surrounded by closed neighbors can be bound inside the solid.

IV. CONCLUSION

In the present work, we have performed the first systematic experimental study of the intensity of the Lyman-α radiation emitted by hydrogen fragments issued from H_n^+ cluster foil dissociation as a function of cluster mass number and velocity. We have clearly shown that the variation of the two parameters n (cluster mass) and v (cluster velocity) leads to different ways to observe an enhancement of the Lyman-α radiation due to charge-exchange processes. However, due to the complexity of the projectiles, no simple model can quantitatively describe these experimental results and we hope these measurements will stimulate theoretical studies on the cluster-foil interaction mechanisms.

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