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Neutral fragment production induced by collision of 60-keV/amu-H_{25}^+ clusters with helium

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Abstract

We report on measurements of the neutral fragment yields resulting from the collision of 60-keV/amu-H_{25}^+ clusters with helium. The sum of the mass numbers of all the neutral fragments ($S_{neutral}$) coming from each cluster is measured. The histogram obtained can be roughly divided into three $S_{neutral}$ value ranges corresponding to evaporation (2 amu$\leq S_{neutral} \leq$10 amu, even), electron capture ($S_{neutral}$=25 amu), and ionization (11 amu$\leq S_{neutral} \leq$23 amu, mainly). The ionization process which corresponds to both odd and even values of $S_{neutral}$ appears to be involved in about 83% of the dissociation events observed. These results, and their comparison with the inclusive ionic fragment distributions previously measured, show the importance of multi-ionization of the incident cluster during the collision. Moreover, the fragmentation of the instable multicharged cluster leads to the production of several light H_p^+ fragments ($p \leq 5$).

key words : high-energy cluster collision, fragmentation phenomena, cluster fragmentation, multi-ionization process, Coulombic explosion, ionic hydrogen clusters

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1. Introduction

Fragmentation [1-3] is a relatively ubiquitous phenomenon that underlies processes such as polymer degradation, breakup of liquid droplets, the crushing of rocks, and the fragmentation of nuclei. Fragmentation of atomic or molecular clusters is a rapidly growing research field [4-7]. The availability of high-energy mass-selected cluster beams is an interesting tool for such studies. In the MeV energy range, a single collision with a target atom can lead to the multi-ionization of the incident cluster. This allows the observation of the fragmentation of the resulting multicharged cluster.

In our previous experiments [6] on fragmentation of mass-selected 60-keV/amu-H_n^+ (n=9, 13, 21, 25, 31) induced by a single collision with helium, we have measured the inclusive mass distributions of the charged fragments. These distributions are very different from those obtained at a lower energy [8] where mainly H_2 molecule evaporation has been observed (ionic hydrogen clusters are a nucleation of H_2 molecules around a H_3^+ core). A prominent production of ionic fragments of masses which are intermediate between the masses of the evaporated dimers H_2 and the ones of the corresponding ionic fragments has been observed. Moreover, double and triple H_3^+ fragment production has been seen [9]. The formation of H_3^+ ions takes place during the fragmentation that occurs after ionization of the incident cluster, even for small species (n=9). The importance of the H_3^+ production could be related to the high efficiency of the $H_2^+ + H_2$ reaction [10]. This is evidence that a multifragmentation process (the triple H_3^+ production) induced by at least a double ionization of the ionic incident cluster occurs.

Here, we report on measurements of the neutral fragment production yields resulting from the collision of 60-keV/amu-H_{25}^+ clusters with helium atoms.

2 - Experimental apparatus

The experiment was performed at the high-energy cluster-beam facility of the Institut de Physique Nucléaire de Lyon [11]. The experimental apparatus is described in details in ref 12. In short, neutral clusters are formed in a cryogenic source and then ionized by electron impact. Cluster ion beams are obtained with the new radiofrequency (RF) quadrupole post accelerator

following the Cockroft-Walton accelerator. The beam is pulsed with a cycle combining the cluster source cycle and the cycle of the RF power. 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. Thus, a cluster burst can either be deflected into the monitoring surface-barrier (SB) detector or reach the target. The intensity of the collimated incident cluster beam corresponds to about 1000 clusters per second. The target is a gas-jet system described in detail previously [13]. 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 and the undissociated clusters are deflected into a movable (SB) detector. The neutral fragments resulting from the dissociation of a cluster are detected simultaneously with a SB detector centered in line with the incident beam and located one meter after the target. The corresponding time of flight is 0.30 μs. The signal is proportionnal to the total energy of the detected fragments. The detector is connected to a pulse height analyzer.

3. Results and discussion

The neutral fragment spectrum produced by collision of H_{25}^+ clusters with helium atoms is displayed in Fig. 1a. This spectrum has been obtained for a given number of incident clusters with a target thickness of $0.73 \pm 0.08\ 10^{14}$ atoms/cm^2. It shows 24 separate peaks where the total energies range from 0.12 to 1.5 MeV. All the neutral fragments coming from one incident cluster reach the detector and are simultaneously detected. The rate of pile up events corresponding to the simultaneous detection of two incident clusters in the monitoring detector has been measured and found to be less than 1%. Therefore, one event in this spectrum corresponds to the interaction of one incident cluster with the target. The number associated with each peak (Fig. 1a) corresponds to the value of the sum of the mass numbers of all the neutral fragments coming from one cluster ($S_{neutral}$). Later on, we will call the $S_{neutral}$ event a fragmentation event for which the sum of the mass numbers of all the neutral fragments coming from one cluster is equal to $S_{neutral}$.

The width of the peaks is nearly constant when $S_{neutral}$ increases. In the case of charged fragment production, we have observed [6] that the width of the peaks increased with the size of the charged fragment. It has been connected to a cluster effect on the response of the detector [14]. In the case of neutral fragments, the width of the peaks indicates that the $S_{neutral}$ events are mainly composed of light fragments such as H_2 or H.

In this spectrum (Fig.1a), we can observe two different parts. In the left part of the spectrum (2 amu$\leq S_{neutral} \leq$10 amu), the numbers of $S_{neutral}$ events, $N(S_{neutral})$, corresponding to even $S_{neutral}$ values are much higher than the numbers corresponding to odd $S_{neutral}$ values. $N(S_{neutral})$ tends to decrease with $S_{neutral}$ increasing. We note that the fragment H ($S_{neutral}$=1 amu) has not been measured but its production yield is expected to be small like the one of the odd $S_{neutral}$ values in this part of the spectrum. From $S_{neutral}$=11 to 25 amu, $N(S_{neutral})$ corresponding to odd $S_{neutral}$ values increases up to the same order of magnitude as the even ones. Moreover, in this part of the spectrum, $N(S_{neutral})$ increases with $S_{neutral}$, up to $S_{neutral}$=19 amu, and then decreases with increasing $S_{neutral}$, describing a characteristic "hump" shape.

Fig. 1b displays the spectrum obtained without a gas target for the same number of incident clusters. These spurious events correspond to about 14% of the incident clusters. The behaviour of this spectrum looks similar to the one obtained with the gas target except for the left part of the spectrum (2 amu$\leq S_{neutral} \leq$10 amu) which is strongly enhanced. We note that the relatively high number of spurious events is mainly due to the events with $S_{neutral}$=2, 4 and 6 amu (≈50%). These spurious events are likely due to collisions of incident clusters with the residual gas and to spontaneous evaporation of H_2 molecules after the last magnetic analysis of the incident beam.

The total neutral event cross section, $\sigma_{neutral}$, has been measured and used to deduce the $S_{neutral}$ event production yields, $\tau(S_{neutral})$, from the spectra 1a and 1b. The fraction of the total number of neutral events as a function of the target thickness x is displayed Fig. 2. We deduce the total neutral event cross section, $\sigma_{neutral}=(28.0 \pm 2.8)10^{-16}$ cm^2, from the slope of the straight line. The errors in the absolute cross sections deduced from these measurements vary from ±10% to ±15%, taking into account the target thickness and the statistical uncertainties [13].

The number of neutral events has to be compared to the number of dissociated clusters

for a given target thickness. In Fig. 3, the fraction F(x) of H_{25}^+ transmitted through the target is reported versus the target thickness x. The transmitted fraction decreases exponentially with x increasing and the dissociation cross section σ_{25}^{dis} is deduced from its slope. The value of σ_{25}^{dis} ($(29.5 \pm 3.0)10^{-16}$ cm^2) is in good agreement with our previous value ($(26.3 \pm 2.6)10^{-16}$ cm^2) [12]. The dissociated fraction calculated with the dissociation cross section for the target thickness used to obtain the spectrum displayed in Fig 1a is 19.3±2%. The number of neutral events in this spectrum corresponds to 20.3±2.0 % of the incident clusters. Thus, taking into account the uncertainties of the measurements, each cluster which dissociates is observed to give at least one neutral fragment. This could be connected to the relatively high value (0.3μs) of the time of flight between the target and the detection chamber.

The absolute $S_{neutral}$ event production yields, $\tau(S_{neutral})$, given by

$$\tau(S_{neutral}) = \left(N(S_{neutral}) \;/\; \sum_{S_{neutral}=2}^{25} N(S_{neutral}) \right) \sigma_{neutral} \qquad (1)$$

are deduced from the spectra 1a and 1b after substraction of spurious events. Since about 20% of H_{25}^+ incident clusters are dissociated, $\tau(S_{neutral})$ is measured under single-collision conditions and each $S_{neutral}$ event corresponds to the interaction of one incident cluster with one helium atom. The absolute neutral event yields, $\tau(S_{neutral})$, versus $S_{neutral}$ are shown in Fig.4.

The $S_{neutral}$ events correspond to the following fragmentation channels of the unstable clusters resulting from a collision :

$$(H_{25}^+)^* \rightarrow H_p^+ + (S_{neutral}/2)\, H_2$$

with $S_{neutral}=(25-p)$

corresponding to evaporation process,

$$(H_{25}^{q+})^* \rightarrow \Sigma_q H_p^+ + S_{neutral} \text{ (neutral fragments)}$$

with $S_{neutral}=(25-\Sigma q p)$, with p=1,2,3,5,...odd, and with q>1.

corresponding to fragmentation after q-1 ionizations of the cluster,

$(H_{25}{}^0)^* \rightarrow S_{neutral}$ (neutral fragments)

with $S_{neutral}$=25 amu

corresponding to cluster electron capture.

The processes described above, evaporation , ionization and electron capture can roughly be connected to three $S_{neutral}$ ranges in the histogram presented in Fig. 4 (2 amu$\leq S_{neutral} \leq$10 amu, even), (11 amu$\leq S_{neutral} \leq$23 amu) and ($S_{neutral}$=25 amu), respectively (we note that the events corresponding to $S_{neutral}$ =24 amu require further investigations to elucidate their source).

The left part of the histogram where events with even values of $S_{neutral}$ are mainly observed is connected to the H_2 molecular evaporation process. $\tau(S_{neutral})$ is found to decrease with increasing $S_{neutral}$ as observed in Fig. 4. Specific features such as the enhancement of $\tau(6)$ compared to $\tau(4)$ and $\tau(8)$ can be related to finite size effects such as "shell" effect on the cluster structure. Besides, in this part of the histogram, we note that $S_{neutral}$ events corresponding to odd $S_{neutral}$ values probably result from ionization and their production rates are one order of magnitude below the even ones.

The absolute electronic capture cross section of $H_{25}{}^+$ on helium ,$\tau(25)$, is 6.2 10^{-17} cm^2. This cross section should be compared to the absolute electronic capture cross section of $H_3{}^+$ ions on helium at 60 keV/amu. To our knowledge, this data is not available in the literature. Nevertheless, the electron capture cross sections for $D_3{}^+$ ions at 50 keV/amu on argon and hydrogen can be estimated from ref. 15. We notice that their values, 1.0 10^{-17} cm^2 and 0.9 10^{-17} cm^2, respectively, are much smaller than the electron capture cross section measured for $H_{25}{}^+$.

The main feature of Fig. 4 is the increase of $\tau(S_{neutral})$ for the odd $S_{neutral}$ values greater than 11 until they are comparable to the corresponding even ones. Moreover, $\tau(S_{neutral})$ exhibits a characteristic "hump" shape for the odd $S_{neutral}$ values as well as for the even ones. This effect is connected to the ionization of the cluster leading to the fragmentation of the unstable multicharged cluster resulting from the collision. The great number of neutral fragments detected in coincidence for large values of $S_{neutral}$ shows that ionization leads to fragmentation in small charged fragments. Nevertheless, as mentioned previously, the fragments are magnetically analysed 0.3 μs after the collision. Angular distribution of the neutral fragments should indicate

if the small charged fragments come directly from the transient multicharged cluster or result from evaporation of larger sized ionic fragments. The observation of neutral events with an odd $S_{neutral}$ value can be connected to the production of a H fragment. The importance of $\tau(S_{neutral})$ for odd $S_{neutral}$ values can be related to the H_2^+ ion dissociation ($(H_2^+)^* \rightarrow H^+ + H$) but also to the high efficiency of the reaction $H_2^+ + H_2 \rightarrow H_3^+ + H$ as discussed in our previous paper on multiple H_3^+ production [9].

From our data, we can evaluate the ionization rate, $R_{ionization}$, by calculating the ratio defined as follows

$$R_{ionization} = \sum_{S_{neutral}=11}^{23} \tau(S_{neutral}) \Big/ \sum_{S_{neutral}=2}^{25} \tau(S_{neutral}) \qquad (2)$$

$\tau(S_{neutral})$ values measured for odd values of $S_{neutral}$ in the range (2 amu$\leq S_{neutral} \leq$10 amu) should be taken into account in the calculation of the ionization rate. But their contribution to the ionization rate is equal to $((3.7\ 10^{-17}\ cm^2)/(28\ 10^{-16}\ cm^2)=1.3\%)$ and so is negligible. Indeed, $R_{ionization}$ has been found to be equal to 83%. This high value of the ionization rate confirms that multi-ionization processes can occur in the collision as already shown by the multiple H_3^+ production [9].

The first term in equation (2)

$$\sum_{S_n=11}^{23} \tau(S_{neutral}) = 23.3\ 10^{-16}\ cm^2 \qquad (3)$$

gives an order of magnitude of the ionization cross section. Approximating the H_{25}^+ cluster by a set of twelve H_2 molecules, this value divided by 12 ($\approx 1.9\ 10^{-16}\ cm^2$) is found to be of the same order of magnitude as the single ionization cross section ($1.5\ 10^{-16}\ cm^2$) of H_2 molecules by collision with helium at 60 keV/amu [16].

From the present results on neutral event yields, we can deduce the charge event production yields, $\tau(S_{charged})$. $\tau(S_{charged})$ corresponds to the sum of the mass numbers of all the charged fragments coming from one H_{25}^+ cluster ($S_{charged} = 25 - S_{neutral}$). From our previous results on the inclusive distributions of the charged fragments resulting from the fragmentation of

H_{25}^+ clusters colliding with helium at 60 keV/amu [6], we have deduce the production cross sections $\sigma_{25,p}$ of charged fragments of mass p. In Fig.5, we have reported $\tau(S_{charged})$ versus $S_{charged}$ and $\sigma_{25,p}$ versus p. We note that the fragments H^+ and H^0 have not been measured. From the neutral fragment measurements, the H^0 production is expected to be negligible, but this is not the case for the H^+ production.

From the inclusive distribution, one can calculate the ratio, R, of the sum of the masses of the charged fragments over the sum of the masses of the dissociated clusters, for the target thickness x used to obtain the spectrum presented in Fig.1a. R is given by

$$R = \frac{\sum_{p=2,3,5,\,odd}^{23} p\,\sigma_{25,p}\,x}{25\left(1 - e^{-\sigma_{25}^{dis} x}\right)} = 35.4\ \% \qquad (4)$$

This ratio can also be calculated from the sum of the of the fragment masses which are not detected in the neutral measurements :

$$R = \frac{\sum_{S_{charged}=2}^{24} S_{charged}\,\tau(S_{charged})\,x}{25\left(1 - e^{-\sigma_{25}^{dis} x}\right)} = 31.8\ \% \qquad (5)$$

These two values are in agreement if one takes into account the uncertainties of the measurements (± 10%). We notice that about 33.5 % of the mass of the dissociated cluster is found in charged fragments.

On the right side of the histogram (Fig.5), for $S_{charged}$>14 amu, the two distributions are similar except for even $S_{charged}$ values of $\tau(S_{charged})$ which have been assigned to ionization process. This part of the histogram reflects the process of molecular evaporation. The main feature of this histogram is the shape difference of the two distributions on the left side corresponding to the ionization process (except the yield $\tau(0)$) : The inclusive distribution of the charged fragment yield (with only odd mass fragment p) has a "U" shape and the charged event yield (with odd-even events) has a "hump' shape. If now we compare each $\tau(S_{charged})$ value with the production cross section $\sigma_{25,p}$ of the charged fragment of mass number $p=S_{charged}$, we see that, for (2 amu$\leq S_{charged} \leq$5 amu), the charged fragment production cross section is greater than

the $S_{charged}$ event production and for ($S_{charged}$>5 amu), the inverse is true. It appears that a great number of events correspond to the production of several small ionic fragments with p≤5. This shows the importance of multiple ionization of the incident cluster among the various excitation processes resulting from the collision.

4. Conclusion

Here, we have reported on measurements of the neutral fragment production yields resulting from the collision of 60-keV/amu-H_{25}^+ clusters with helium. The sum of the mass numbers of all the neutral fragments coming from one cluster is measured. The histogram obtained can be roughly divided into three $S_{neutral}$ value ranges corresponding to the various processes which result from the collision. Each cluster which dissociates is observed to give at least a neutral fragment. The evaporation process corresponds to the small values of $S_{neutral}$ (2 amu≤ $S_{neutral}$ ≤10 amu, even). The electron capture ($S_{neutral}$ =25 amu) is observed here for the first time in this high velocity range and the corresponding cross section is found to be larger than the electronic capture cross section of D_3^+ ions on argon or hydrogen at 50 keV/amu. The ionization process which corresponds to both odd and even values of $S_{neutral}$ appears to be involved in about 83% of the dissociation events. The comparison of these results with the inclusive ionic fragment distributions previously measured shows the importance of multi-ionization of the incident cluster resulting from the collision, leading to the production of several light ionic fragments (p≤5).

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Figure captions

Figure 1 : a) The mass spectrum of neutral fragments produced by collision of H_{25}^+ clusters with helium atoms under single-collision conditions. The number associated to each peak corresponds to the value of the sum of the mass numbers of all the neutral fragments coming from one cluster ($S_{neutral}$).

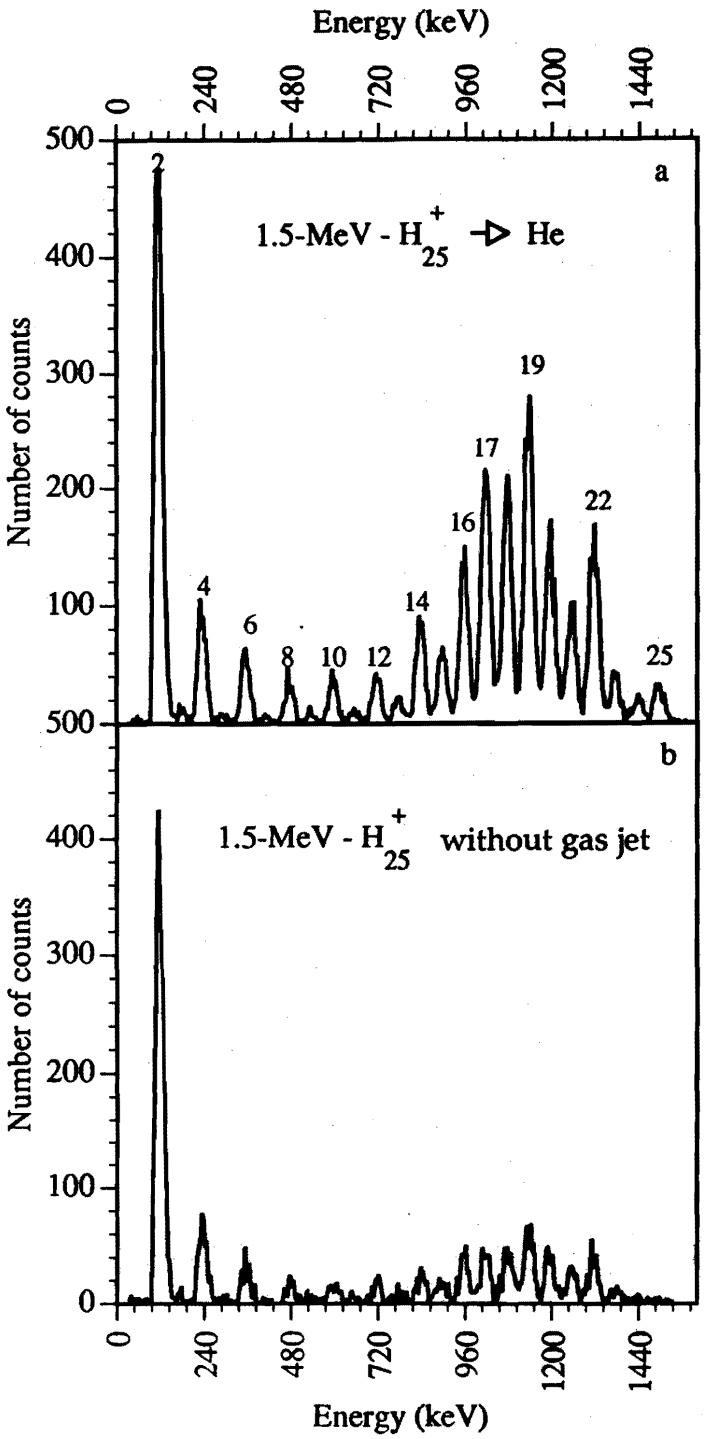
b) The same spectrum without a gas target.

Figure 2 : Fraction of the total number of neutral events as a function of the target thickness x. The slope gives the "neutral " cross section.

Figure 3 : H_{25}^+ transmitted fraction, F(x), measured as a function of the target thickness x. The slope gives the dissociation cross section.

Figure 4 : Histogram of the neutral event production yields, $\tau(S_{neutral})$, versus $S_{neutral}$.

Figure 5 : ▒ Histogram of the $S_{charged}$ event production yield, $\tau(S_{charged})$, versus $S_{charged}$ =(25-$S_{neutral}$), i.e. the sum of the mass numbers of all the charged fragments coming from one H_{25}^+ cluster. ■ Histogram of the production cross section of charged fragments resulting from the fragmentation of H_{25}^+ clusters colliding with helium at 60-keV/amu versus the charged fragment mass p [5].



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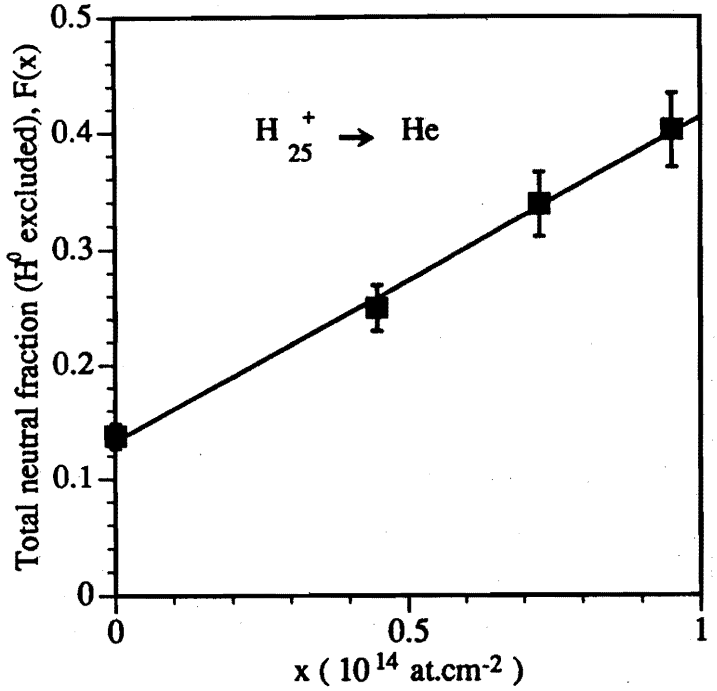

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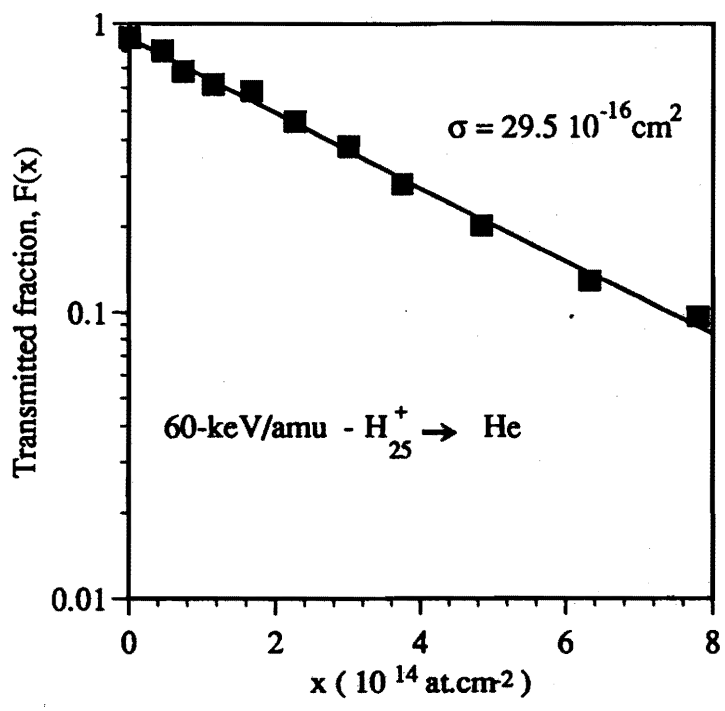

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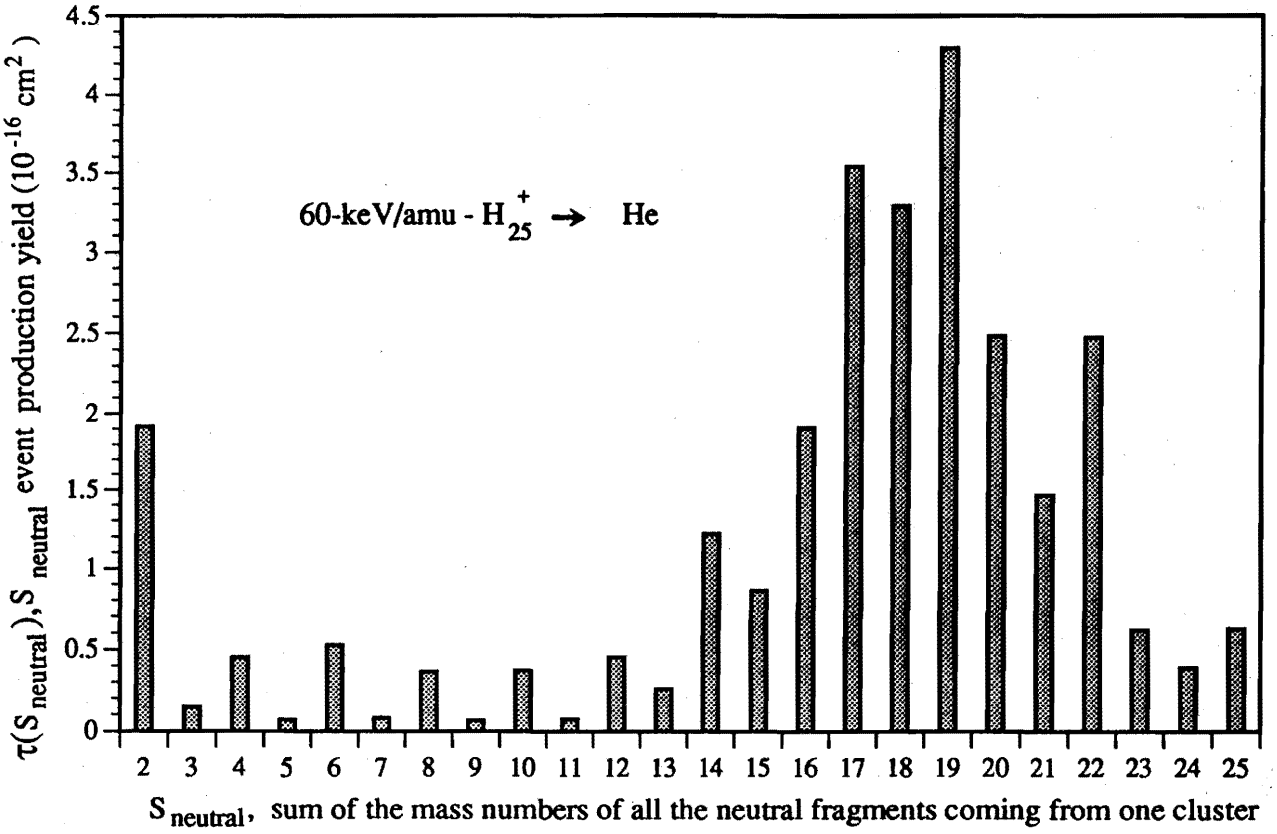

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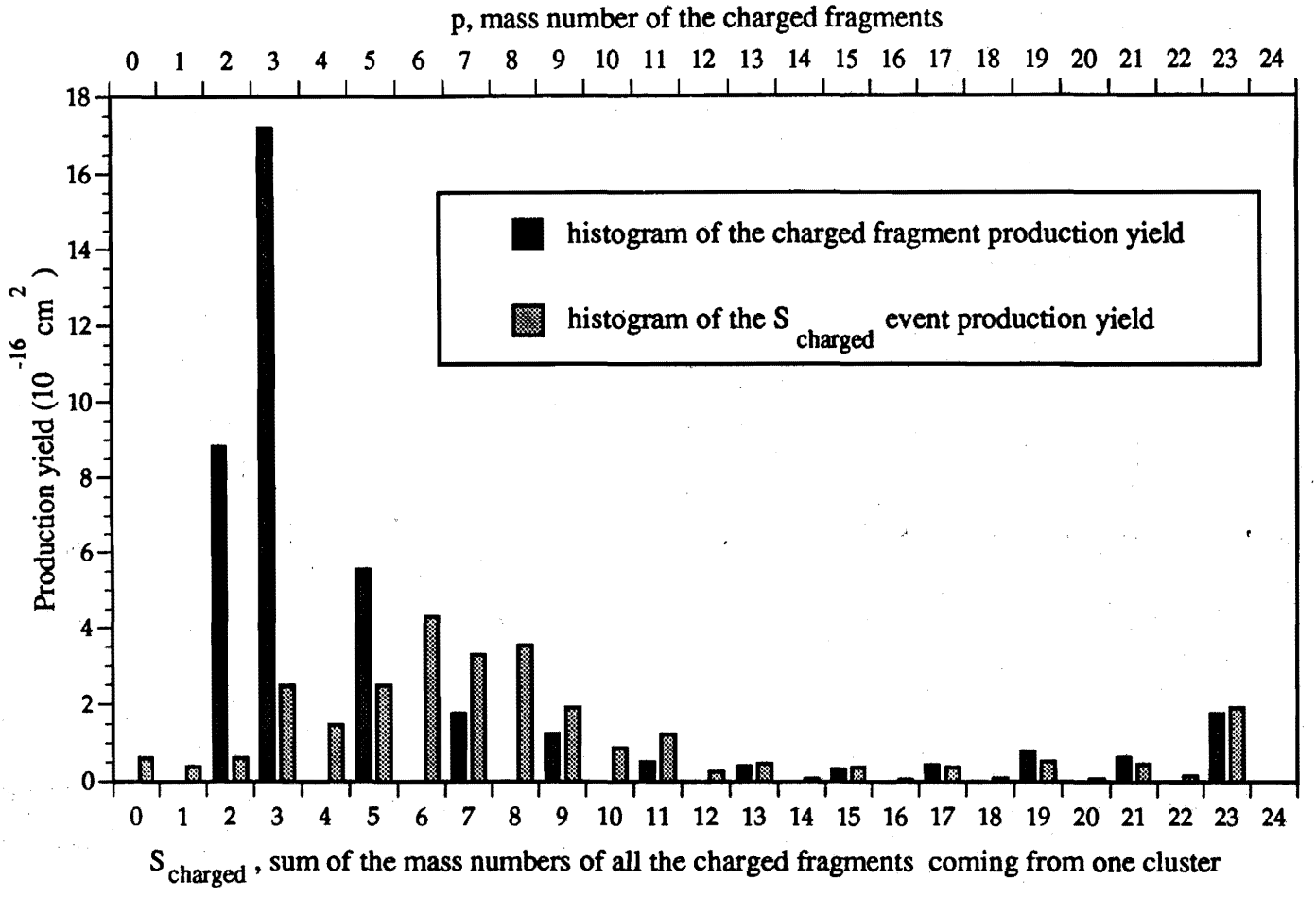

Fig 5 : B Farizon et al