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Strong inhibition effect on secondary-electron emission induced by fast hydrogen clusters

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We report on measurement of secondary-electron emission (SEE) induced by hydrogen clusters ($H_n{}^+$, $n \leq 19$, odd) from thin carbon foils for beam velocities above and around the Bohr velocity. The n dependence of the SEE yield shows a strong inhibition effect with respect to the atomic case. At a given velocity, the inhibition effect is observed first to increase with n and then to reach a saturation value ($n \geq 7$). These results are connected to the understanding of the inhibition effects observed with molecular beams.

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It is well known that the bombardment of a solid target with swift atomic particles can lead to ejection of electrons from the solid. The phenomenon is called secondary-electron emission (SEE), and the total secondary-electron yield γ is defined as the average number of electrons emitted per incoming projectile. Most of these emitted electrons have energies below 20 eV and originate mainly from a layer of the order of 10–20 Å below the solid surfaces. In the range of velocities studied here, the dominant production mechanism is the kinetic emission of electrons, generally considered as a three-step process. First, the projectile transfers kinetic energy to target electrons. Next, a fraction of these electrons moves from the bulk towards the target surfaces, and, finally, a fraction of the electrons reaching the surface passes through it. Thus, the kinetic emission is related to that fraction of the kinetic energy of the projectile which is communicated to target electrons (the electronic energy loss). Most SEE measurements have been performed with ionized atoms and few experiments [1–5] have been carried out with incoming molecular ions. Especially, it has been observed by several authors that γ, for various targets under bombardment by fast H^+, $H_2{}^+$, and $H_3{}^+$ ions of the same velocity, do not scale like the number of constituent protons of the projectiles. A reduced ratio R_n of the SEE yields for molecular ions $\gamma(H_n{}^+)$ and protons $\gamma(H_1{}^+)$ is defined as $R_n = \gamma(H_n{}^+)/(n\gamma(H_1{}^+))$ with $n=2$, 3 for $H_2{}^+$ and $H_3{}^+$, respectively. An observation of $R_n \neq 1$ is taken as an indication of a molecular effect. From the experimental data [1–5], it follows that R_n increases with increasing projectile velocity from values smaller than one to values greater than one in the energy range from about 10 keV/u to 1.2 MeV/u. Molecular effects and, recently, hydrogen-cluster effects have been investigated in other features related to atom-solid interactions such as charge-state distributions [6,7], beam-foil processes [8], ion desorption [9], and also in energy-loss measurements [7]. To our knowledge, the only experimental work on SEE measurements with cluster ions has been performed with $H_5{}^+$, $H_7{}^+$, and $H_9{}^+$ impinging on thick gold, molybden, and steel targets [10]. In this paper, we present the results on the total electron emission yield from thin carbon foils induced by hydrogen clusters $H_n{}^+$ ($n \leq 19$, odd) in the energy range 30–300 keV/u.

60–600 keV hydrogen-cluster beams are delivered by the Cockroft-Walton cluster accelerator of the Institut de Physique Nucléaire de Lyon. $H_n{}^+$ bursts of 80 ms duration are produced at a repetition rate of 0.5 Hz. After acceleration, the cluster beam is selected in energy and mass by electrostatic and magnetic analyzers. The beam is focused by an electrostatic triplet of quadrupole and a small aperture defines a cross section of 1.5 mm^2 on the target. The thin self-supporting carbon foils are produced by standard evaporation methods. The foils are held by circular frames with $\phi = 2.5$-mm holes, mounted on a target holder fixed on a goniometer. A negative voltage of 40 V applied to the target is enough for the electron emission yield to reach a saturation value [11]. The transmitted beam current (about 1 nA during the burst) is measured with a Faraday cup equipped with an electron suppressor ring. In order to protect the target from the electrostatic field due to the voltage of the suppressor ring, a metallic ring connected to the mass is set between the target and the suppressor ring. The angular aperture of the device is 20° to insure the full collection of the transmitted beam in the Faraday cup despite the multiple scattering and Coulomb explosion of the fragments. The thicknesses of the three targets used are 180 ± 18, 210 ± 21, and 400 ± 40 Å. They were determined by measuring the Rutherford scattering yield of α particles and by comparison with a calibrated target. Each fresh target is cleaned up by sputtering with $N_2{}^+$ beams delivered by the accelerator before use. The pressure in the beam line is less than 10^{-6} Torr.

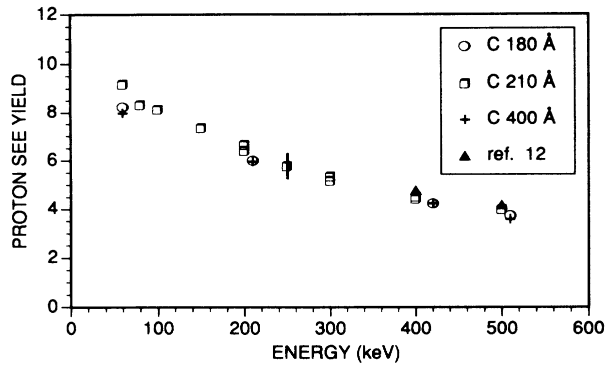

FIG. 1. Energy dependence of the total SEE yield for incident proton beams into various carbon foils.

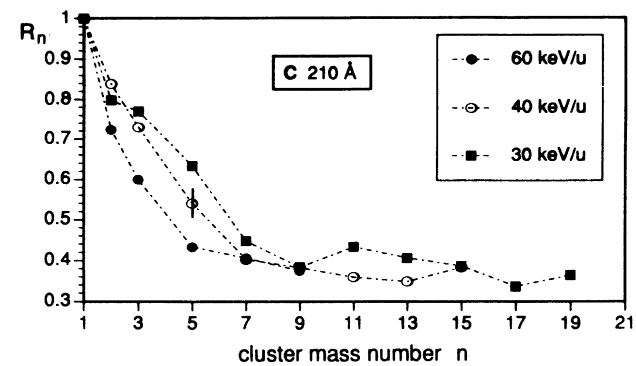

FIG. 3. The same as for Fig. 2 for 30–60 keV/u projectile velocities.

$\gamma(H_n{}^+)$ is obtained by calculating the charge balance at the target:

$$\gamma(H_n{}^+)=(Q_t/Q_{FC})q_n^f+(q_n^f-q_n^i)\,, \qquad (1)$$

where Q_t and Q_{FC} are the charges measured at the target and in the Faraday cup, respectively. The charge of the incident projectile, q_n^i is one for any cluster. q_n^f is the mean final charge state of the projectiles after leaving the foil exit surface. From data on atomic and cluster projectiles transmitted through thin carbon foils [6,7], we deduce the total mean charge q_n^f of the fragments emerging from the solid target. It must be noted that q_n^f depends slightly on the thickness of the foil. At emergence, the fragments are mainly atomic. In our velocity range, the negative-ion H^- fraction, the molecular and cluster transmitted fractions are negligible [6]. Moreover, the foils are thick enough to imply that the H^0 emergent species are protons having picked up a target electron. The expression (1) is valid only if other secondary processes are absent or negligible. Major disturbing effects are the production of positive or negative ions from the target. As mentioned in Refs. [1,2] and [4], and references therein, these effects are known to be small. They have been neglected in the present experiment.

Absolute errors in the SEE yields are in the order of ±10%, mainly due to the measurements with the Faraday cup and including the errors on q_n^f and on the thickness of the foil.

Before investigating cluster effects on the SEE yield, it is necessary to measure the SEE yield ($\gamma(H_1{}^+)$) induced by protons in the same conditions. The energy dependence of $\gamma(H_1{}^+)$ for 60–500 keV proton beams impinging on various carbon foils is presented on Fig. 1. These $\gamma(H_1{}^+)$ values are in very good agreement with previous results when data are available [12]. This experiment has been performed under standard high vacuum conditions. Thus, the value of $\gamma(H_1{}^+)$ is higher (about 25%) than the one measured by Meckbach, Braunstein, and Arista [13] at the same velocity under UHV conditions. Nevertheless, $\gamma(H_1{}^+)$ exhibits the same trend with velocity.

In Figs. 2–4 the cluster effect ratio R_n is plotted versus n for various projectile velocities and for a given target thickness (Figs. 2 and 3), and for various target thicknesses and for a given projectile velocity (Fig. 4). The $\gamma(H_1{}^+)$ values for 30 and 40 keV/u protons have been deduced from the data given in Ref. [13] by increasing these values of 25%.

The main experimental findings are the following:

(i) For all cluster mass number, velocity, and target thickness combinations, an inhibition effect with respect to the proton case is observed on the SEE yield ($R_n<1$) except in one case. Indeed, a ratio greater than one is obtained with 300 keV/u $H_2{}^+$ incident molecules as already shown for high incident velocities [5]. From our results,

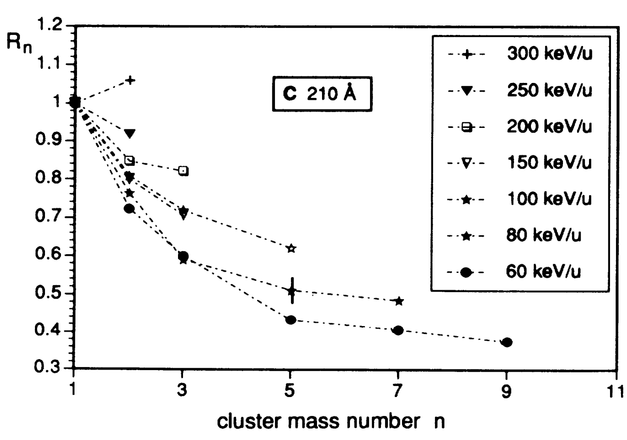

FIG. 2. Cluster mass number dependence of $R_n=\gamma(H_n{}^+)/(n\gamma(H_1{}^+))$ for various projectile velocities (60–300 keV/u) and for a 210-Å carbon foil. Typical error bar is included.

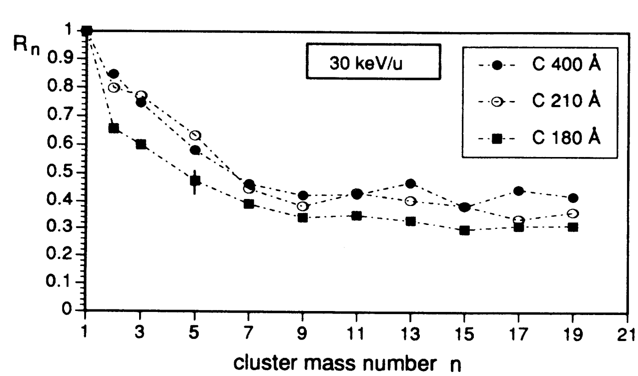

FIG. 4. The same as for Fig. 2 for 30 keV/u projectile velocity and for various target thicknesses (180, 210, and 400 Å).

R_2 equal to one is expected for incident energy between 250 and 300 keV/u. This is in agreement with the results of Svensson and Holmen [3] who have observed a R_2 value from copper equal to one for an incident energy of 200 keV/u. One has to notice that the rate of the inhibition effect $(1-R_n)$ observed with clusters (58% with H_7^+ at 60 keV/u) is much higher than the effect observed with molecular ions in the same conditions (28% with H_2^+ at 60 keV/u).

(ii) For a given velocity and a given target thickness, the inhibition effect is seen to increase first with n and then to reach a saturation value for $n=5$ or 7. The same trend is observed for any velocity and target thickness.

(iii) The inhibition effect increases with decreasing velocity down to 60 keV/u (for a given cluster mass number and a given target thickness). However, for velocities below 60 keV/u, the velocity dependence of R_n seems to change.

(iv) The inhibition effect seems to decrease slightly with increasing target thickness for various cluster mass numbers at a given projectile velocity (30 keV/u). The same trend has also been observed at 60 keV/u velocity.

The total SEE yield measured is the sum of the yields of electrons emitted from the entrance and exit surfaces. In the velocity range studied, the backward and the forward SEE induced by protons are of the same order of magnitude [13]. Thus, the strong inhibition effects observed with clusters on the total SEE yield ($\geq 50\%$) imply that both backward and forward SEE are involved. The slight dependence of R_n with target thickness at a given velocity (Fig. 4) has to be related to the forward SEE. Indeed, the internuclear separation between the fragments on their way through the solid increases due to the Coulomb explosion and the multiple scattering. Thus, at the exit of the foil, the cluster effects decrease with increasing target thickness. A great number of experimental studies have been carried out to test whether a possible proportionality between the SEE yield and the electronic stopping power exists. This proportionality was confirmed experimentally for proton bombardment in a wide energy range (10 keV–10 MeV) [12–14]. The forward SEE induced by H_2^+ ions (at high velocity) has also been observed proportional to the molecular electronic stopping power [5]. The hydrogen cluster energy loss [7,15,16] has been investigated in the velocity range studied here. The experimental results show an enhancement of the cluster energy loss with respect to the proton case for incident velocities higher than 60 keV/u and a decrease for smaller velocities. These effects are small (10% maximum) and exhibit a trend with velocity different from the one observed for the total SEE yield. These remarks seem to show that the most important part of the inhibition effect observed is not due to the forward emission.

Concerning the variation with n of the inhibition effect, it has to be connected to the structure of the incident projectile. The inhibition effect is observed to increase with increasing n up to $n \cong 5$–7 and then to reach a saturation value. It is known that a hydrogen cluster structure is a nucleation of H_2 molecules around an H_2^+ core [17]. The increase of R_n could be associated with the vicinity effect between the molecules of the cluster which are close together when reaching the foil. This cannot be the case since a saturation with n is observed for a small number of molecules in the cluster. Therefore, the saturation value of the inhibition effect would have to be compared to the inhibition effect that one could expect with a beam of fast neutral H_2 molecules.

About the incident electrons, they are lost by the projectile in the first layers of the foil. At high velocity, they lead to an enhancement of the SEE yield [5]. Nevertheless, this effect decreases with decreasing velocity and becomes negligible around the Bohr velocity since the incident electron energy is then too small to induce SEE. Screening effect of the projectile nucleus by their accompanying electrons during the liberation of target electrons has been proposed in order to explain the inhibition effects [1,2,5,18]. Such explanation has been notably suggested when studying SEE from gold surface under neutral-atom (H^0, He^0) bombardment in comparison with charged ions (H^+, H^-, He^+) of equal impact velocity [18]. $\gamma(H^0)$ has been found smaller than $\gamma(H^+)$. Moreover, $\gamma(H^-)$ has been obtained equal to $\gamma(H^0)$. These results connected to screening effects due to the different accompanying electrons have been explained in terms of charge-exchange processes. The inhibition effect observed on the SEE yield induced by He^0 atoms with respect to the He^+ case has led to the same conclusions. Turning to cluster results, the effect of the proximity of the protons at the entrance of the target has to be questioned. It is well known that the H_2^+ and H_3^+ molecular ion beams currently delivered by accelerators are excited in high vibration states and consequently the mean distances between the protons are higher than the distance calculated for a molecule in its fundamental state. The mean distance is about 1.3 Å for H_2^+ and 1.2 Å for H_3^+ [16]. Concerning H_n^+ clusters, they are weakly bound and no important vibrational excitation in the H_3^+ core and in the H_2 subunits can take place (cold molecules). The distance between the protons in the H_2 subunits of the projectile is close to the theoretical distance (0.74 Å) [16,17]. One has to notice that this distance is smaller than or of the same order of magnitude as the dynamic screening length due to the target electrons (0.7–1 Å in the carbon, in the velocity range studied here). It has been shown that charge-exchange processes are modified by such proton proximity [6] and, especially, the electronic-loss cross section is decreased with respect to the isolated proton case [19]. Therefore, if one compares the free H_2^+ molecular ions and the cold H_2 molecules in a cluster in the first layers of the target, an electron will stay bound to the protons of a cold H_2 projectile over a distance greater than in the H_2^+ case. This leads to a greater screening effect of the protons by an accompanying electron in the low-energy H_2 molecules case and could explain the strong inhibition effect observed on the total SEE yield induced by clusters.

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