

Populations of $2p$ and $3p$ terms in hydrogen excited by H^+ , H_2^+ , and H_3^+ ions passing through thin carbon foils

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Relative beam-foil populations of the $2p$ term in hydrogen have been measured as a function of the proton energy ($0.015 \leq E \leq 1.10$ MeV). For $E \geq 0.1$ MeV, these populations are found to be proportional to the equilibrium neutral fractions of protons emerging from a carbon foil. At lower energies ($E < 0.1$ MeV), the behavior of the excitation function of the $2p$ term is compatible with a decrease of the ratio of the $2p$ population to the ground-state population with respect to that ratio at higher energies. The first precise measurements of the dependence on the projectile energy ($0.1 < E < 1.8$ MeV) of the population of the $2p$ term excited by the passage of H_2^+ and H_3^+ ions through carbon foils of various thicknesses (2.5 – $22 \mu\text{g}/\text{cm}^2$) are reported. Only the long-dwell-time region ($t \geq 1.5$ fs) is considered in this work. The variation of $R = I^{\text{molec}}/I^{\text{atom}}$ (I^{molec} and I^{atom} are the Ly- α intensities per incident proton observed with molecular and atomic projectiles of the same velocity, respectively) with the projectile energy per nucleon (E/M) and the thickness (T) of the foil is well described by the following relation: $R = R_\infty [1 - C \exp(-kE/M)]$, where R_∞ , C , and k are parameters depending only on T , and on the number of protons in the cluster. A qualitative explanation of the $R(E/M, T)$ behavior is proposed. Values of R have also been measured for Ly- β radiation and are found to be significantly smaller than those obtained for Ly- α radiation, for foils of thickness $T \lesssim 6 \mu\text{g}/\text{cm}^2$.

I. INTRODUCTION

It is now well established that the fraction of neutral hydrogen atoms per incident proton, at the exit of thin carbon foils bombarded with H_2^+ and H_3^+ projectiles, is larger than that observed with protons of the same velocity.^{1,2} Several authors^{3–8} have also shown that the intensity of lines emitted by neutral hydrogen, after beam-foil interaction, depends on the projectile used: molecular projectiles (H_2^+ , H_3^+) leading to stronger intensities per proton than incident protons.

In the models proposed to describe the interaction of atomic clusters with solid targets, the internuclear separation in the emerging projectiles and consequently the dwell time t , i.e., the time spent by the projectile in the foil, plays a vital role (see, e.g., Refs. 2, 6, and 9–12). Three dwell-time regions are generally considered in the study of the overproduction of neutral atoms with molecular projectiles: (i) $t \geq 15$ fs, no cluster effect is observed; (ii) $2 \leq t \leq 15$ fs, an overproduction of H^0 is observed which decreases with the dwell time; and (iii) $t \leq 2$ fs, the overproduction of H^0 increases very rapidly when the dwell time decreases.² In this work the short-dwell-time region, $t < 2$ fs, is not studied.

First, we report measurements of the relative population of the $2p$ term in H^0 , excited after the passage of H^+ ions through carbon foils, as a function of the proton energy ($0.015 < E < 1.10$ MeV). These results are compared to the neutral fractions of proton beams emerging from carbon foils which have been measured by several workers.^{1,2,11,13–16}

Second, we present the first precise study of the dependence on the projectile velocity of the ratio

$R = I^{\text{molec}}/I^{\text{atom}}$, where I^{molec} and I^{atom} are Ly- α intensities per incident proton, obtained using molecular projectiles (H_2^+ , H_3^+) and protons of the same velocity, respectively. R values have been measured in the energy range 0.1 – 1.8 MeV with foils of thickness varying from 2.5 to $22 \mu\text{g}/\text{cm}^2$. Our R values are compared to R measurements of other authors and to $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ results² [ϕ_0^{molec} and ϕ_0^{atom} are the neutral fractions per incident proton measured using molecular projectiles (H_2^+ , H_3^+) and H^+ ions of the same velocity, respectively]. An analytical expression is proposed for fitting the dependence of R on the projectile energy per nucleon (E/M) and on the target thickness (T). A tentative explanation is given for the observed behavior of $R(E/M, T)$. Finally, we report R measurements for Ly- β radiation and compare them to those obtained under similar experimental conditions for Ly- α radiation.

II. EXPERIMENTAL PROCEDURE

Beams of H^+ , H_2^+ , and H_3^+ ions were produced by a 2-MV Van de Graaff accelerator and excited by passage through a carbon foil. The beam-foil spectroscopy arrangement described in detail previously^{17,18} has been slightly modified for this work. Beam currents of about 0.1 to $1 \mu\text{A}$ (3 mm in diameter) were used. A first additional carbon foil could be inserted into the beam in order to dissociate molecular projectiles into protons before they pass through the second carbon foil located about 10 mm after this additional removable foil. This well-known “double foil” technique was generally employed in this work for obtaining proton beams and provided low-energy protons which could not be directly produced by the ac-

celerator.

The radiation (Ly- α or Ly- β) emitted after the foil was monitored at right angles to the beam axis through a Seya-Namioka-type spectrometer equipped with three channeltron detectors. The number of photons was counted for a predetermined number of ions passing through the foil. Repeated measurements of intensity in different experimental conditions (beam intensity, source pressure, etc.) indicate that errors are typically about 10%.

Experimental problems encountered in this work are the following: (i) the production of uncontaminated beams of H^+ , H_2^+ , and H_3^+ ions; (ii) the precise measurement of the number of ions passing through the foil; and (iii) the knowledge of the foil thickness which increases during the irradiation. These problems will be now examined in more detail.

A. Verification of the purity of H^+ , H_2^+ , and H_3^+ beams

The beam purity was verified using two metallic plates parallel to the beam axis between which an adjustable voltage (0–3 kV) was applied. These plates were located at the entrance of the excitation chamber. The deflected ions in different charge states were visualized on a quartz window located in the excitation chamber. It was observed that, for pressure lower than a few 10^{-6} Torr in the whole beam transport system and in the excitation chamber, the incident beam contained essentially singly charged ions. By counting the number of photons emitted after the foil, with and without applying the voltage to the plates, the contribution of the neutral component of the incident beam to the Ly- α intensity was found to be less than 5%, at any energy, in our vacuum conditions described above.

B. Measurement of the number of incident ions

In our setup, the excitation chamber which is electrically isolated is in fact a Faraday cup; the collimated ion beam enters the chamber and passes through the foil. If all the ions and secondary electrons emerging from the foil are collected by the whole chamber and if the beam contains only singly charged ions, the charge recorded is proportional to the number of incident ions. In order to collect all the secondary electrons which could escape (principally through the pumping system), a voltage of +90 V was applied to the chamber. We used beams of about 3 mm in diameter to avoid scattering of the beam by the 5-mm foil holder. We verified that the ion currents measured with and without the foil interposed into the beam were the same, which indicates that all the secondary electrons emitted by the foil electrically connected to the chamber were collected by the chamber. Under these conditions, the collected charge is proportional to the number of incident ions since the ion beam is practically pure as discussed in Sec. II A.

C. Measurement of the foil thickness

The foil thickness could be measured at any time during the irradiation using the following method. The protons backscattered at a given angle by the carbon target were

detected by a silicon surface-barrier detector located inside the excitation chamber. The numbers of protons backscattered by the different foils, for a given collected charge, are proportional to the foil thicknesses. The proportionality coefficient was obtained from the number of protons scattered by several foils of known thickness which had not yet been irradiated. Since the relative increase of foil thickness during the irradiation could be important for the thinnest foils ($\approx 2 \mu\text{g}/\text{cm}^2$),¹⁹ it was necessary to monitor the thickness of the foils during the measurements.

III. EQUILIBRIUM BEAM-FOIL POPULATION OF THE $2p$ TERM

A. Results

We have measured the relative population of the $2p$ term in hydrogen, excited by the passage of protons through carbon foils, as a function of the beam energy at the exit of the foil. These populations have been obtained by multiplying the observed intensity of the Ly- α radiation by the proton velocity after the foil. We verified that they are independent of the foil thickness even for the thinnest foils employed (equilibrium populations). The energy loss of protons in carbon was obtained from Ref. 20 using $1.65 \text{ g}/\text{cm}^3$ for the volume density of carbon.² Our results are given in Fig. 1. Different proton beams were used: (i) protons produced directly by the accelerator and (ii) protons obtained by dissociating H_2^+ and H_3^+ ions in a first carbon foil (see Sec. II). Results obtained with these different beams are in good agreement (see Fig. 1). Let us also note that our measurements agree well with the data of Gabrielse⁵ who measured beam-foil excitation of the $2p$ term in H^0 between 0.16 and 0.70 MeV.

B. Comparison with the neutral fraction

In Fig. 1 we have also plotted the equilibrium neutral fraction of proton (or deuteron) beams emerging from carbon targets and measured by several authors.^{1,2,11,13–16} The recent data of Kreussler and Sizmann¹⁶ for $0.02 < E < 0.2 \text{ MeV}$ are not plotted in Fig. 1 but are in good agreement with the other data. Our relative populations are normalized to the mean value of the neutral fraction observed at 0.3 MeV. For $E \geq 0.1 \text{ MeV}$, the excitation function of the $2p$ term closely follows the neutral fraction variation, but for $E < 0.1 \text{ MeV}$ these two sets of data are no longer proportional. This difference of behavior means that, at low energies, the ratio of the total number of H^0 to the number of H^0 in the $2p$ state is larger than that ratio at higher energies. This seems to indicate that the number of H^0 in the ground state relative to the number of H^0 in the $2p$ state is larger at low energies than at high energies since most of the atoms emerging from a foil are in their ground state.

The values predicted for the neutral fractions in a semi-quantitative model proposed by Brandt and Sizmann²¹ are also plotted in Fig. 1 (solid curve) for comparison. These

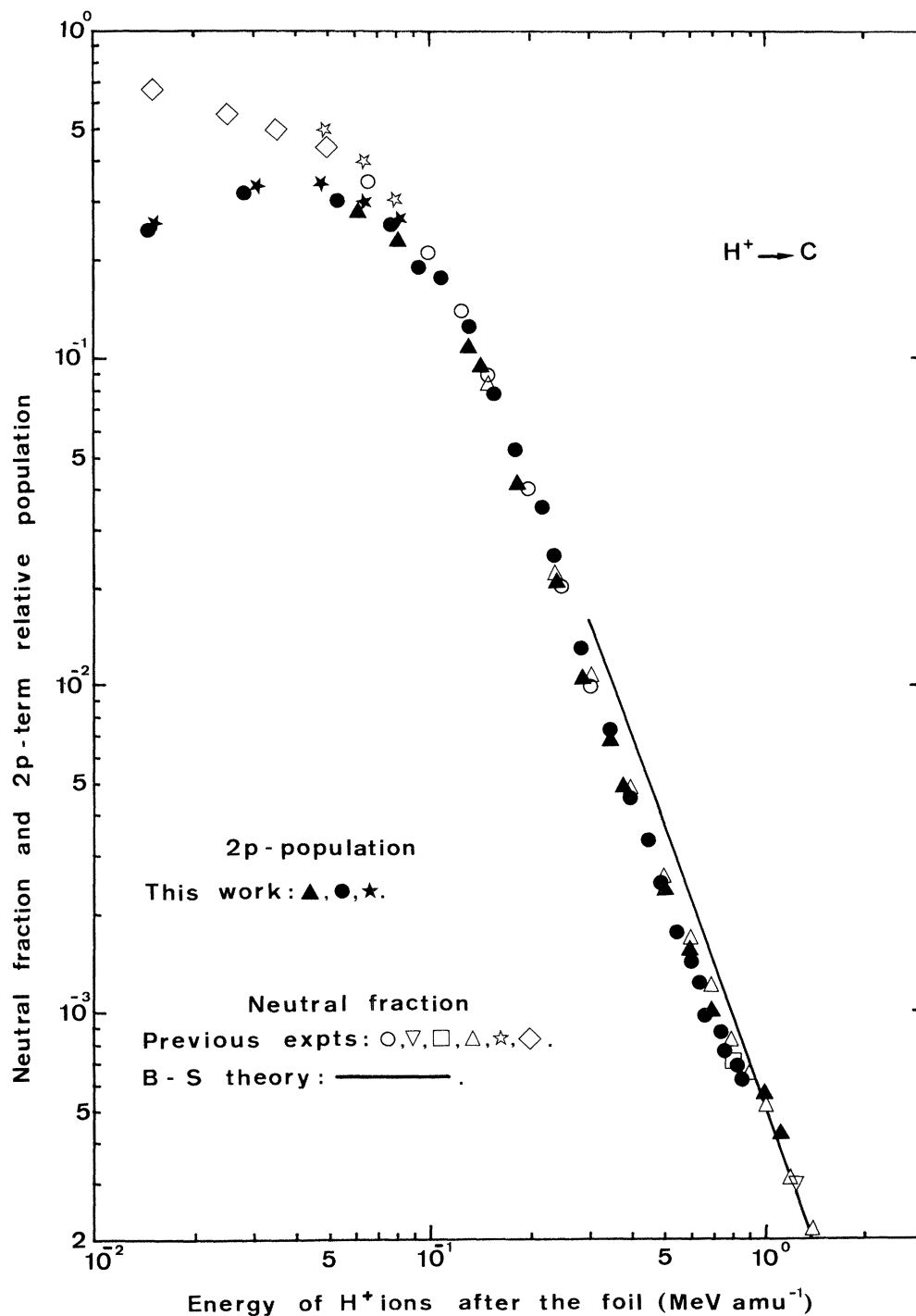


FIG. 1. Equilibrium neutral fraction and relative population of the $2p$ term for proton beams emerging from carbon foils as a function of the proton energy. Neutral fraction data are from Meggitt *et al.* (Ref. 1), \circ ; Gaillard *et al.* (Ref. 2), ∇ ; Cue *et al.* (Ref. 11), \square ; Chateau-Thierry and Gladieux (Ref. 13), \triangle ; Berry *et al.* (Ref. 14), \star ; and Berkner *et al.* (Ref. 15), \diamond . Data of Ref. 15 are obtained with deuterium beams. Solid curve shows theoretical predictions of Brandt and Sizmann (BS, Ref. 21). Our population data are obtained using proton beams directly produced by the accelerator (\blacktriangle) or obtained by dissociating H_2^+ (\bullet) and H_3^+ (\star) ions in a carbon foil; they are normalized to the neutral fraction observed at 0.3 MeV.

values have been calculated using Eqs. (1)–(3) given in Ref. 22, which are valid for $E > 0.3$ MeV. These theoretical values are in close agreement with the experimental neutral fractions and with the $2p$ -population data for

$E > 0.9$ MeV, as already shown by Chateau-Thierry *et al.*²² for the neutral fraction data at energies up to 16 MeV. However, for $E < 0.9$ MeV, the experimental values are significantly smaller than the theoretical results.

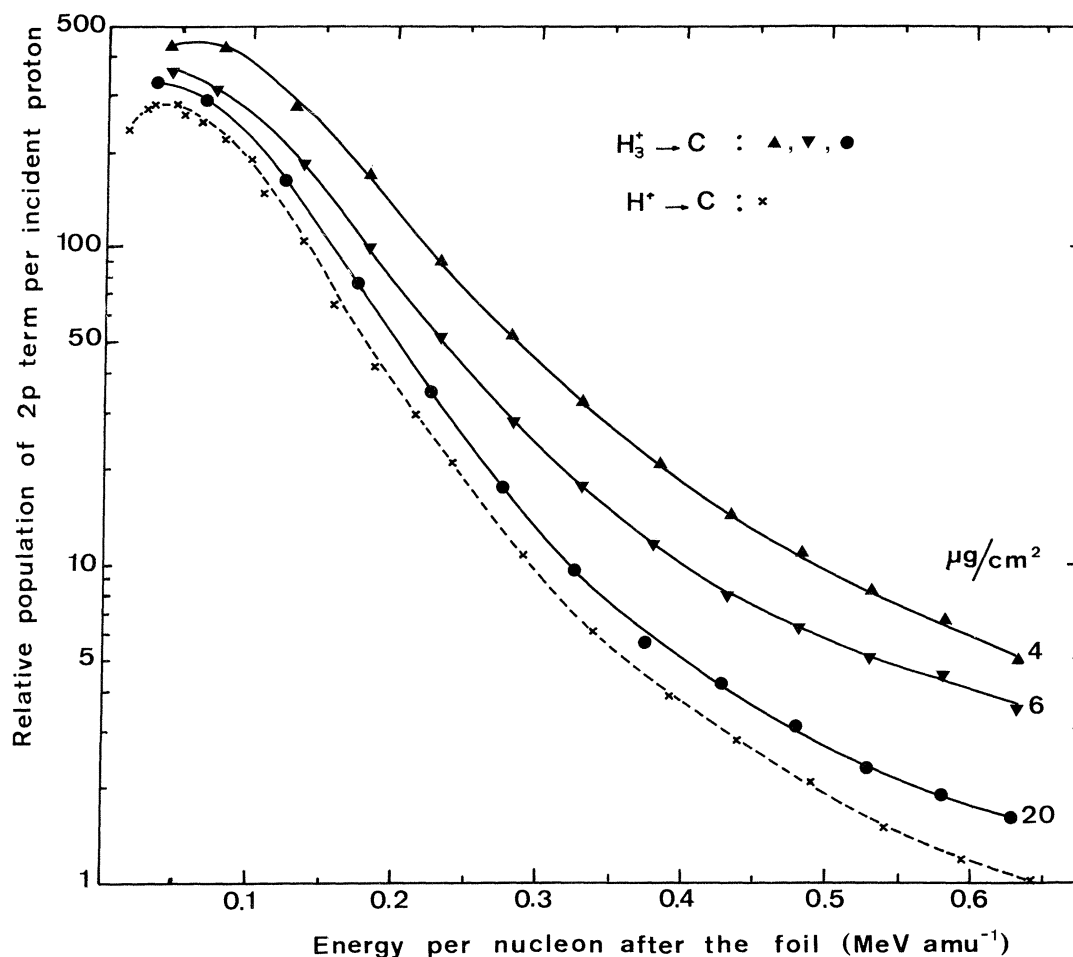


FIG. 2. Relative populations of the $2p$ term excited by the passage of H_3^+ and H^+ ions through carbon foils of various thicknesses as a function of the projectile energy per nucleon after the foil. Solid curves and dashed curve are drawn to guide the eye.

IV. MOLECULAR EFFECTS IN BEAM-FOIL POPULATIONS OF THE $2p$ TERM

A. Experimental results

We have measured relative populations per incident proton for the $2p$ term excited after the passage of H^+ , H_2^+ , and H_3^+ ions in carbon foils of various thicknesses (2.5 – $22 \mu\text{g}/\text{cm}^2$) and projectile energies ranging from 0.1 to 1.8 MeV . Relative populations are determined as explained in Sec. III A; the proton beams were obtained by breaking up H_2^+ and H_3^+ projectiles in a thin carbon foil (see Sec. II).

In Fig. 2 relative populations per incident proton of the $2p$ term are plotted as a function of the energy per nucleon after the foil for H^+ and H_3^+ projectiles. As already mentioned in Sec. III A, populations of terms excited by the passage of protons through carbon foils are independent of the foil thickness T (at least for values of $T \gtrsim 2.5 \mu\text{g}/\text{cm}^2$ considered in this work). Excitation functions obtained with H_3^+ projectiles are given in Fig. 2 for carbon foils of 4 , 6 , and $20 \mu\text{g}/\text{cm}^2$. We see that the $2p$ population per incident proton is larger for H_3^+ projectiles than

for protons of the same velocity; the enhancement of the population being greater for thinner foils. These results are in agreement with previous observations.^{5,6}

In Figs. 3(a) and 3(b), ratios $R = I^{\text{molec}}/I^{\text{atom}}$ (I^{molec} and I^{atom} have been defined in Sec. I) are plotted as a function of E/M . Data are given for different foil thicknesses. R increases with E/M and reaches a saturation value R_∞ for a value of E/M which increases with the thickness T . For a foil of a given T , R_∞ is larger for H_3^+ ions than for H_2^+ ions and for a given molecular projectile, R_∞ increases when T decreases.

B. Analytical expression for $R(E/M, T)$

The dependence of R on E/M can be expressed by the simple relation

$$R = R_\infty [1 - C \exp(-kE/M)], \quad (1)$$

where R_∞ , C , and k are parameters depending on T and on the number of particles in the cluster. For H_2^+ projectiles, the experimental data are well fitted using Eq. (1) with the parameters given by the following relations:

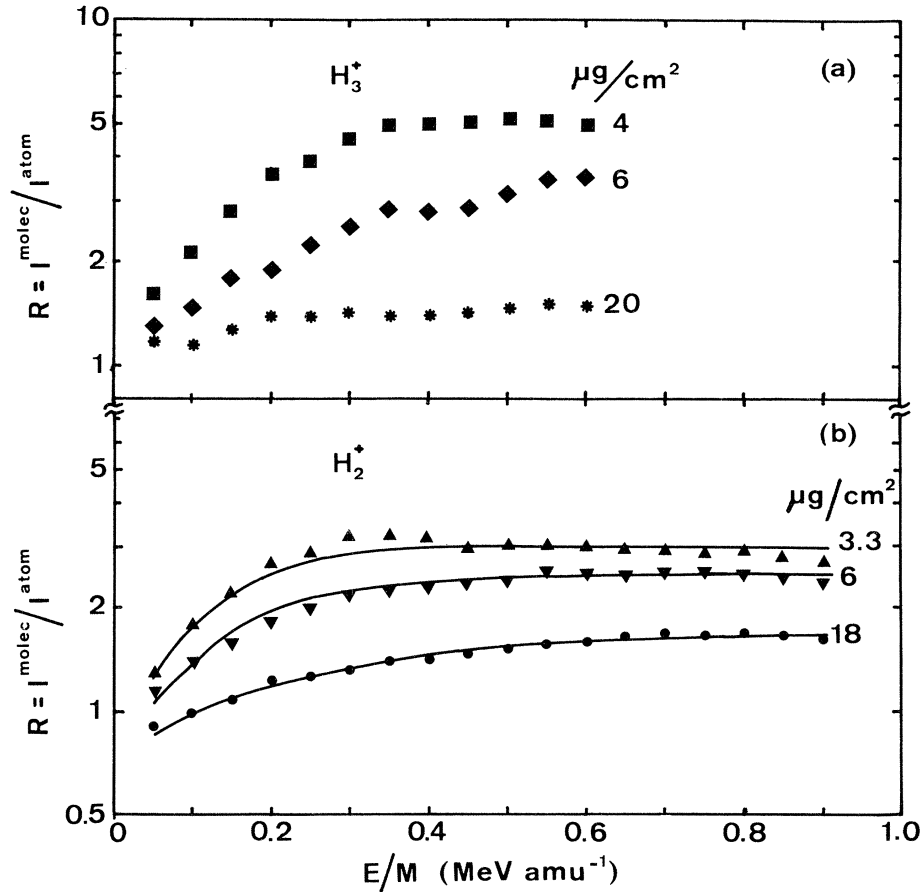


FIG. 3. Ratios $R = I^{\text{molec}}/I^{\text{atom}}$ as a function of the energy per nucleon of the projectile after the foil, for foils of various thicknesses. I^{molec} and I^{atom} are Ly- α intensities per incident proton obtained (a) with H_3^+ ions and protons of the same velocity, respectively, and (b) with H_2^+ ions and protons of the same velocity, respectively. Solid curves represent fits to the data obtained using Eqs. (1)–(4) (see text).

$$R_{\infty} = 3.9 - 0.77 \ln T, \quad (2)$$

$$C = 6.17 T^{-0.63} / R_{\infty}, \quad (3)$$

$$k = 18.0 T^{-0.573}, \quad (4)$$

where E/M and T are expressed in MeV/amu and in $\mu\text{g/cm}^2$, respectively. The solid curves in Fig. 3(b) give the R values calculated using Eqs. (1)–(4).

C. Comparison with other results

By plotting our R values as a function of the dwell time t , we can compare our results to the R values obtained by Brooks and Berry⁶ for Ly- α radiation and to the $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ values (ϕ_0^{molec} and ϕ_0^{atom} have been defined in Sec. I) determined by Gaillard *et al.*² Our R results are in agreement with the R values given in Ref. 6, but are larger than the $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ values given in Ref. 2. For a dwell time $t \approx 3$ fs, $R \approx 2.5$ and 4.5 for H_2^+ and H_3^+ projectiles, respectively, i.e., a factor about 2 larger than the $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ values.² Kobayashi and Oda⁸ have also observed that the $I^{\text{molec}}/I^{\text{atom}}$ values for the Balmer lines ($n \geq 3$) are much larger than the $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ values. Since it is expected that most ions are in the ground state at the

exit of a foil, all these results seem to indicate that the molecular enhancement is smaller for the ground-state population than for excited-state populations.

D. Discussion of the results

In previous works,^{2–8} no detailed study of the behavior of R and $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ on E/M and T has been made. It was generally supposed, in first approximation, that the dwell time t is the relevant parameter, R and $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ depending only on the dwell time.^{2,5,6,8} Gaillard *et al.*² have suggested the following “surface effect” for the overproduction of neutrals observed with molecular projectiles in the long-dwell-time region ($t \gtrsim 2$ fs). When a proton emerges from the foil there is a probability P_1 , independent of the proximity of other protons, that it captures an electron, and a probability P_2 that it captures an electron correlated with a proton of the same cluster. Then one can write the following:

$$\phi_0^{\text{molec}}/\phi_0^{\text{atom}} = 1 + P_2/P_1. \quad (5)$$

In the model of Ref. 2, P_2/P_1 depends only on the distance S separating the protons in the cluster as it emerges from the foil, i.e., essentially on the dwell time t . This

means that, in the electron-capture probability of clusters leading to H^0 formation, one can isolate the velocity dependence, which is the same as that of protons, from the influence of the cluster separation (or dwell time) and thus write the following:

$$P_2(v, t) = P_1(v)F(t), \quad (6)$$

$$\phi_0^{\text{molec}}/\phi_0^{\text{atom}} = 1 + F(t), \quad (7)$$

$F(t)$ being a decreasing function of t .

This model could be applied to our R results by replacing P_1 , P_2 , and $F(t)$ by P_1^* , P_2^* , and $F^*(t)$ where the asterisk refers to the capture of an electron in the $2p$ state only. This would lead to the following relation:

$$R = 1 + P_2^*/P_1^*,$$

where

$$P_2^*(v, t)/P_1^*(v) = F^*(t). \quad (8)$$

If our R values given in Fig. 3(b) are plotted as a function of t , it appears that Eq. (8) is not rigorously valid in the dwell-time region $t \lesssim 3$ fs where, for a given t , R decreases when v or T increases. For $t > 3$ fs, Eq. (8) seems valid within experimental errors.

For a given foil thickness, R tends, as t decreases, toward a maximum value R_∞ which is reached with H_2^+ projectiles for $t \simeq 2.5$ fs and 3.5 fs for the 3.3- and 6- $\mu\text{g}/\text{cm}^2$ foils, respectively [see Fig. 3(b)]. These t values correspond to an internuclear separation S of the dicluster at the exit of the foil about 2.5 Å.²³ This result could be explained by assuming that $P_2^*(v, t)/P_1^*(v)$ reaches a saturation value as t decreases (v increases) for $S \simeq r_{2p}$, where $r_{2p} = 2.6$ Å is the radius of the $2p$ electron orbit in hydrogen. Indeed, when $S < r_{2p}$ some electrons correlated with the cluster may miss the capture in the $2p$ orbit of one proton and be captured preferentially in the $1s$ orbit. (Let us recall that this work is not concerned with the short-dwell-time region where the R behavior should be due to the role played by electrons incident with the molecule that maintain some correlation with the nuclei while in the target.^{2,6,11}) Examination of Fig. 3 shows that, with H_3^+ projectiles and a foil of given thickness, R_∞ is reached for higher velocities (shorter t values) than those required with H_2^+ projectiles. This situation confirms the above explanation since the dwell time required to reach a given S value is shorter for H_3^+ than for H_2^+ projectiles.²

Let us note also that at internuclear distances S smaller than those corresponding to R_∞ , an increase of $\phi_0^{\text{molec}}/\phi_0^{\text{atom}}$ when S decreases could be observed because the major part of hydrogen atoms are in the ground state ($r_{1s} = 0.5$ Å) at the exit of the foil.

For foils of different thicknesses, the increase of R when T or v decreases, at a given dwell time ($t \lesssim 3$ fs), cannot be accounted for in the model of Gaillard *et al.*² This R behavior may be tentatively explained by one of the following assumptions. (i) In the probability that a proton captures in the $2p$ state an electron correlated with a proton of the same cluster, one cannot isolate the velocity dependence, which is not the same as that of protons, from the influence of the cluster separation (or dwell time). For a given t , $P_2^*(v, t)/P_1^*(v)$ decreases when v or T

increases. (ii) In addition to the surface effect² there is a "solid effect" leading to an increase of the number of electrons correlated to the protons in the cluster when T decreases.

V. MOLECULAR EFFECTS IN BEAM-FOIL POPULATIONS OF THE $3p$ TERM

We have measured $I^{\text{molec}}/I^{\text{atom}}$ for Ly- β radiation ($R_\beta = I^{\text{molec}}/I^{\text{atom}}$) using foils of various thicknesses and H_2^+ beams of different energies. The curves giving R_β as a function of E/M have a trend similar to that observed for R_α given in Fig. 3(b).

We have also determined R_α/R_β for several foils of different thicknesses and H_2^+ projectiles in the energy range 0.3–1.8 MeV. The errors which could arise from the change of experimental conditions during the measurements (structure of the foil, characteristics of the beam, etc.) were suppressed by measuring Ly- α and Ly- β intensities successively, the only modified parameter being the wavelength. No dependence on the beam energy has been found for R_α/R_β , within experimental errors. For foil thicknesses $T \gtrsim 10$ $\mu\text{g}/\text{cm}^2$ $R_\alpha/R_\beta \simeq 1$, but for thinner foils R_α/R_β becomes significantly larger than one. For a 2.6- $\mu\text{g}/\text{cm}^2$ foil we obtained

$$R_\alpha/R_\beta = 1.3 \pm 0.1,$$

the error indicated represents twice the standard deviation of the mean value of several measurements at different beam energies. Let us note that Kobayashi and Oda⁸ have also found a decrease of $I^{\text{molec}}/I^{\text{atom}}$ for the Balmer lines when the principal quantum number n increases.

VI. SUMMARY AND CONCLUSIONS

A. Equilibrium population of the $2p$ term

We have measured the relative population of the $2p$ term in neutral hydrogen, excited after the passage of protons through a carbon foil, as a function of the proton energy. This excitation function has been compared to the neutral fraction data of the literature plotted as a function of the proton energy (Fig. 1). Except at low energies ($E \lesssim 0.1$ MeV), the $2p$ population is proportional to the neutral fraction. At energies lower than about 0.04 MeV, a decrease of the $2p$ population as E decreases is observed. This indicates that, at low energies, there is an enhancement of the number of H^0 in the ground state relative to the number of H^0 in the $2p$ state with respect to that ratio at higher energies.

B. Molecular effects for the $2p$ -term population

First, we have measured the precise dependence on the projectile energy ($0.1 < E < 1.8$ MeV) of $R = I^{\text{molec}}/I^{\text{atom}}$ for Ly- α radiation in the dwell-time region $t \gtrsim 1.5$ fs using H_2^+ and H_3^+ ions passing through carbon foils of different thicknesses. We have observed for the first time the existence of a saturation value R_∞ as E/M increases for a foil of given thickness. The R data measured are very well fitted by the relation $R = R_\infty [1 - C \exp(-kE/M)]$

where R_∞ , C , and k are parameters depending only on the target thickness T and on the number of protons in the projectile. For H_2^+ projectiles, the simple relations $R_\infty = a - b \ln T$, $C = cT^{-d}/R_\infty$, and $k = fT^{-g}$ (a , b , c , d , f , and g being positive numerical constants) have been found.

Second, we have interpreted the existence of a saturation value R_∞ , for a given foil thickness, in the simple model of Gaillard *et al.*² by assuming that P_2^*/P_1^* (see Sec. IV D), which increases when the separation S of the protons in the cluster decreases, reaches a saturation value when S is approximately equal to the radius of the 2p orbit of H^0 .

Finally, we have observed an increase of R when v or T decreases, t being constant ($t \lesssim 3$ fs) which cannot be accounted for in the simple model of Ref. 2. This R behavior was tentatively explained either by a surface effect where the dependence of P_2^* on v would not be similar to that of P_1^* on v , or by a solid effect added to the surface effect and leading to a variation with the thickness of the foil of the number of electrons correlated to the cluster.

Let us note, however, that a detailed description of molecular effects in beam-foil populations would have to take into account the formation of quasimolecular states^{6,11} and wake-riding phenomena.⁹

C. Molecular effects for the 3p-term population

We have measured $I^{\text{molec}}/I^{\text{atom}}$ for Ly- β radiation (R_β) using H_2^+ projectiles and foils of various thicknesses. These values have been compared to those obtained for Ly- α radiation (R_α) in the same experimental conditions. It was found that, for thin carbon foils ($T \lesssim 6 \mu\text{g}/\text{cm}^2$), R_α/R_β is significantly larger than 1 and does not seem to depend on the projectile energy ($0.15 < E/M < 0.9$ MeV amu⁻¹).

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