

Experimental study of the $2s$ and $2p$ populations of hydrogen atoms resulting from the interaction of 0.8-MeV/amu H^+ , H^0 , and H_2^+ projectiles with thin carbon foils

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We present measurements of the Stark mixing of the $n=2$ excited states of hydrogen atoms resulting from the interaction of 800-keV/amu hydrogen projectiles with thin carbon foils. Various excitation mechanisms have been studied in which the projectiles are atomic or molecular species and the electron associated with the excited state originates from the incident projectiles or results from a capture in the target. From these measurements we deduce the relative fractions of the $2p$ and $2s$ sublevels in the various cases. We show in particular that the $2s$ sublevel is rather insensitive to the excitation process.

I. INTRODUCTION

The interaction of fast atomic and molecular ion beams in the MeV/amu velocity range with solid targets has been the subject of extensive interest in recent years. Measurements of the yield of neutral hydrogen atoms produced by fast atomic (H^+ , H^0) and molecular (H_2^+ , H_3^+) beams traversing thin carbon foils are used to investigate the mechanisms of interaction of fast hydrogen ions with solids. From previous transmission experiments^{1,2} it seems well established that atomic and molecular hydrogen species can survive the passage through thin carbon foils. In the case of atomic projectiles, these experiments have been analyzed in terms of capture and loss cross sections, just as in the case of a gas target, and are compatible with the idea that a bound system such as H^0 can survive inside a solid target. In the case of molecular incident projectiles, most of them are observed to dissociate, and electron capture and loss processes must be considered in the framework of repulsive molecular states.

In a recent paper,³ we described experiments in which beam-foil spectroscopy is combined with charge-state measurements for atomic and molecular hydrogen projectiles. For H^0 , H_2^+ , and H_3^+ incident projectiles, respectively, the foil thickness dependences of the neutral fraction and the $2p$ state population of H^0 have been measured simultaneously. Other experimental studies⁴⁻⁹ have also revealed molecular effects in the formation of excited states of transmitted atomic fragments.

In order to interpret the measurements of the Lyman- α emission in beam-foil excitation of H^+ , H^0 , and H_2^+ projectiles, the ratio of the $2p$ and $2s$ populations $R = P_{2p}/P_{2s}$ must be known. The experimental determination of this ratio has been done by Gabrielse¹⁰ in the case of incident proton beams. In this paper, we present measurements of the ratio R for excited H atoms produced either by target electron capture (H^+ beam) or by

direct atomic excitation (H^0 beam), by dissociation of original or reconstituted molecules (H_2^+ beam).

II. DESCRIPTION OF THE VARIOUS METHODS USED FOR PRODUCING EXCITED HYDROGEN ATOMS

We have done experimental work on the $n=2$ excited hydrogen atoms in the four cases described below. Two of them involve atomic incident projectiles, the two others involve molecular incident projectiles.

The first case, (a), is the most usual one: protons are sent into a carbon foil of any thickness; then all the emergent hydrogen atoms, excited or not, result from the capture of a target electron.

In the second case, (b), a neutral hydrogen atom is sent into a very thin carbon target. From previous studies¹⁻³ we know how the neutral fraction in the transmitted beam depends on the dwell time t_d , the time spent by the projectile in the target: it is characterized by a simple exponential decay which levels off to a constant value corresponding to the equilibrium neutral fraction. In the exponential decay regime (for dwell times typically lower than 10^{-15} s) the projectiles are transmitted with their original electron, in contrast with the equilibrium regime where they are reconstituted after successive electron losses and target electron captures. If the target is sufficiently thin, the excited hydrogen atom may result from the direct excitation of the incident neutral projectile.

In the third, (c), and fourth, (d), cases the incident projectiles are H_2^+ molecular ions. All of them are broken up in the foil, except a very small fraction, always less than 10^{-2} , which survives passage through the foil. Most of the emergent species, in the MeV energy range, are two-proton clusters. Most of the H atoms detected in the transmitted beam are associated with a proton and then the emergent ($H^+ + H$) clusters must be described as H_2^+

molecules in repulsive states. The study^{1,2} of the dependence of the neutral fraction on target thickness has then revealed two regimes: for short dwell times ($\leq 10^{-15}$ s) the exponential decay of the neutral fraction is interpreted as the transmission of the original molecule after excitation in a repulsive state [case (c)]. For longer dwell times ($1-15 \times 10^{-15}$ s) the original electron has been lost and the emergence of a neutral atom results from the capture of a target electron by the two-proton cluster into a repulsive molecular state [case (d)]. In both cases, the emergent H_2^+ molecule, either transmitted or reconstituted, can also be in a bound state. This is what is called the molecular transmission and is mentioned here just for the sake of completeness. If the dwell time is more than 15×10^{-15} s the separation distance between the protons is such that they behave as independent particles.

The potential energy diagram of the H_2^+ molecule shows that six molecular orbitals converge into the state $H^+ + H$ ($n=2$) for infinite internuclear separation. Any of these six molecular states can contribute to the emission of Lyman- α radiation by a hydrogen atom originating from an incident H_2^+ ion, either by direct excitation of the H_2^+ $1s \sigma_g$ ground state [case (c)], or by capture of a target electron by a two-proton cluster [case (d)]. It is clear that the comparison between the four excitation processes must be made for a given projectile velocity, which is 0.8 MeV/amu. In each case the target thickness has been chosen to give the desired dwell time.

III. EXPERIMENTAL

A. Principle of the experiment

In order to determine the ratio between the $2s$ and $2p$ level populations at the exit surface of the foil, we apply an electric field parallel to the beam axis to induce the Stark mixing of the $2s$ (metastable) and $2p$ (unstable) levels. Under these conditions, the light emission from the Lyman- α transition depends on the field intensity and on the relative $2s$ and $2p$ level populations in the transmitted beam. Therefore, the ratio R can be deduced from the study of this dependence. As the need for very thin targets limits the beam intensity, the light is collected over a significant length of the beam path, in contrast with the time-resolved quantum-beam technique.^{11,12}

B. Experimental procedure

Mass-analyzed beams of H^+ and H_2^+ ions were obtained from the 2.5-MV Van de Graaff Accelerator of the University Claude Bernard Lyon-1. Two experimental arrangements have been used for the three incident beams of H^+ , H^0 , and H_2^+ projectiles. The carbon foils were mounted on a target holder that allowed target translation. The method used for the determination of the target thicknesses and the measurement of the neutral fraction is described in Ref. 3. The Stark mixing between the $2p$ and $2s$ levels is induced by a uniform longitudinal field produced between two parallel plates, 9 mm apart. The first plate, located 6 mm downstream of the foil, is grounded. The fields were varied from 0 to 1.7 kV/cm. A solar

blind G26E315 photomultiplier (PM) tube closed by a MgF_2 window detects the Lyman- α photons emitted between the two electrodes at 90° with respect to the beam direction. Three collimators placed in front of the PM tube are used to define the region of observation and also to avoid edge effects of the holes drilled through the plates for the passage of the beam.

The H^0 transmission experiment requires an incident H^0 beam intense enough to allow beam-foil-type measurements. We found that the best way of preparing such a beam is to dissociate an H_2^+ beam through a gas target with a gas pressure adjusted to maximize the H^0 production. Downstream from the gas target a permanent magnet deviates the charged component of the beam which is used to normalize the intensity of the H^0 beam. One can assume that the H^0 atoms are in the $1s$ state when they enter the target since their time of flight is very long and the $2s$ level is quenched by the magnetic field. The intensity of neutral atoms at the carbon target was about 10^9 particles per second.

For the H^+ and H_2^+ measurements the incident beam intensity is measured by a beam chopper located at the entrance of the target chamber. Preliminary experiments with an H^+ beam traversing foils of various thicknesses and with an H_2^+ beam traversing a "double-foil" target

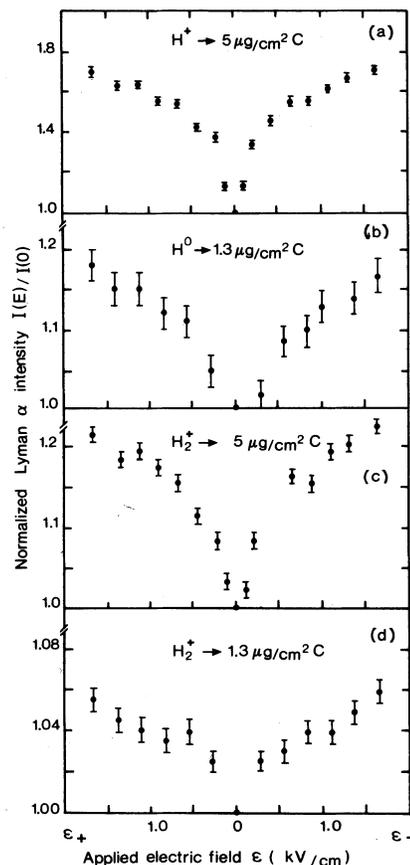


FIG. 1. Dependence of the Lyman- α intensity on the applied electric field, observed in the four combinations of projectiles and target thicknesses (see text). Intensities are normalized to unity when no field is applied.

have shown that the charge state and the light emission measurements are not sensitive to the angular spread caused by multiple scattering and molecular breakup.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Experimental data

The dependence of the Lyman- α intensity on the applied electric field is shown in Fig. 1 for the four cases previously described: (a) H^+ beam on a $5\text{-}\mu\text{g}/\text{cm}^2$ foil; (b) H^0 , $1.3\ \mu\text{g}/\text{cm}^2$; (c) H_2^+ , $5\ \mu\text{g}/\text{cm}^2$; (d) H_2^+ , $1.3\ \mu\text{g}/\text{cm}^2$. The intensity of the light has been normalized in such a way that it is equal to unity when no field is applied. In all cases we observe rather symmetrical enhancement of the light emission when the applied fields are successively parallel and antiparallel to the beam direction. Since the amplitudes and the patterns are quite different, one can expect that a careful analysis of the data should give a different value of R for each case.

B. Analysis of the measurements

For studying the evolution of the atomic system with the electric field we use the formalism of the density matrix to describe the atom ensemble at the exit surface of the carbon foil. The free evolution between the target and the first field plate is then written as a function of the lifetimes and energies of upper levels $2s$ and $2p$. Finally introducing the Stark Hamiltonian leads to the time evolution of the density operator $\sigma(t)$ within the electric field domain. The intensity of the emitted light is given as usu-

al by $\text{Tr}(\sigma \cdot D)$, where D is the detection operator. By integrating this expression of light intensity along the observation path, the populations of upper levels $2s$ and $2p$ are deduced using simulation procedures, the results of which are used for comparison with experimental data in the four cases. The primary purpose of this analysis is to extract R , the ratio of the $2p$ and $2s$ populations. However our calculation will also show that coherent effects are negligible in this experiment. In the following section, we use the phenomenological theory of radiative damping and we neglect the hyperfine structure.

1. Excitation matrix

Due to the purely electrostatic nature of the collision and the cylindric symmetry of the excitation,¹³ the excitation matrix $\sigma(0)$ is written in a particularly simple manner in the $|l, |m_l| \rangle$ basis set. The only nonzero matrix elements are

$$\langle 1, 1 | \sigma | 1, 1 \rangle = \sigma_{p1},$$

$$\langle 1, 0 | \sigma | 1, 0 \rangle = \sigma_{p0},$$

$$\langle 0, 0 | \sigma | 0, 0 \rangle = \sigma_{s0},$$

and

$$\langle 1, 0 | \sigma | 0, 0 \rangle = \langle 0, 0 | \sigma | 1, 0 \rangle^* = \sigma_{sp},$$

where σ_{p1} , σ_{p0} , and σ_{s0} are the populations of the $p \pm 1$, $p0$, and $s0$ levels and σ_{sp} is the coherence term.

In the coupling basis set $|l, s, j, m_j \rangle$ the matrix is transformed into

	$ 1, \frac{1}{2}, \frac{3}{2}, \frac{3}{2}\rangle$	$ 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2}\rangle$	$ 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	$ 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$
$ 1, \frac{1}{2}, \frac{3}{2}, \frac{3}{2}\rangle$	$\frac{1}{2}\sigma_{p1}$	0	0	0
$ 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2}\rangle$	0	$\frac{1}{3}\sigma_{p0} + \frac{1}{6}\sigma_{p1}$	$\frac{\sqrt{2}}{6}(\sigma_{p1} - \sigma_{p0})$	$\frac{1}{\sqrt{6}}\sigma_{sp}$
$ 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	0	$\frac{\sqrt{2}}{6}(\sigma_{p1} - \sigma_{p0})$	$\frac{1}{6}\sigma_{p0} + \frac{1}{3}\sigma_{p1}$	$\frac{-1}{2\sqrt{3}}\sigma_{sp}$
$ 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	0	$\frac{1}{\sqrt{6}}\sigma_{sp}^*$	$\frac{-1}{2\sqrt{3}}\sigma_{sp}^*$	$\frac{1}{2}\sigma_{s0}$

2. Free evolution

The free evolution of the density matrix before the electric field is applied is given by the equation

$$\frac{d\sigma}{dt} = -\frac{i}{\hbar}[H_a, \sigma],$$

where H_a is the free atomic Hamiltonian. Therefore, the matrix element evolution is described by the following set of equations:

$$\sigma(lj, l'j', t) = \sigma(lj, l'j', t=0) e^{-i(E_{lj} - E_{l'j'})t/\hbar} \times e^{-(\Gamma_l + \Gamma_{l'})t/2},$$

where E_{lj} is the energy of the lj level, Γ_l the decay constant of the l level, and t the time of flight from the target. This equation system allows us to calculate the den-

ty matrix elements at the time t_0 when the projectile enters the electric field region.

3. Evolution in the electric field

The Stark Hamiltonian H_S is written in the basis $|l, s, j, |m_j| \rangle$. Its four diagonal terms are given by

$$\langle l, s, j, m_j | H_S | l, s, j, m_j \rangle = E_{lj} - i\hbar \frac{\Gamma_l}{2}$$

and the four nonzero extra diagonal terms are

$$\langle 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2} | H_S | 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \rangle = A\epsilon$$

$$= \langle 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} | H_S | 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2} \rangle,$$

$$\begin{aligned} \langle 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} | H_S | 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \rangle &= B\epsilon \\ &= \langle 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} | H_S | 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \rangle, \end{aligned}$$

where ϵ is the applied electric field strength, and A and B are the coefficients of the first-order Stark perturbation.¹⁴

Let S be the matrix of the components of the eigenstates of H_S in the basis $|j, |m_j| \rangle$. The evolution of the density operator is written as

$$\begin{aligned} \sigma(t) &= \sum_{\alpha, \beta, l, m} S_{\alpha l}^{-1} \sigma_{lm}(t_0) (S_{m\beta}^{-1})^\dagger \\ &\times e^{-i(\omega_\alpha - \omega_\beta^*)(t-t_0)} |\alpha\rangle \langle \beta|, \end{aligned}$$

	$ 1, \frac{1}{2}, \frac{3}{2}, \frac{3}{2}\rangle$	$ 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2}\rangle$	$ 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	$ 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$
$ 1, \frac{1}{2}, \frac{3}{2}, \frac{3}{2}\rangle$	$\frac{1}{2} A_{2p-1s}$	0	0	0
$ 1, \frac{1}{2}, \frac{3}{2}, \frac{1}{2}\rangle$	0	$\frac{5}{6} A_{2p-1s}$	$-\frac{\sqrt{2}}{6} A_{2p-1s}$	0
$ 1, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	0	$-\frac{\sqrt{2}}{6} A_{2p-1s}$	$\frac{2}{3} A_{2p-1s}$	0
$ 0, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}\rangle$	0	0	0	0

where A_{2p-1s} is the transition probability of the Lyman- α transition. Finally, the intensity of the emitted light is given by $I = \text{Tr}(\sigma \cdot D)$. I_+ is the emission intensity integrated over the observation region when ϵ is parallel to the beam direction and I_- is the corresponding intensity when ϵ is antiparallel to the beam direction. It has been shown^{15,16} that $(I_+ + I_-)$ depends only on the populations and that $(I_+ - I_-)$ depends only on the coherences. Therefore, we need measurements of both I_+ and I_- intensities.

5. Determination of the populations

Let $I_{p1}(\epsilon)$, $I_{p0}(\epsilon)$, $I_{s0}(\epsilon)$ be the simulated sums $(I_+ + I_-)$ corresponding to initial conditions where only one sublevel $p1$, $p0$, or $s0$, respectively, is populated. The experimental intensity $(I_+ + I_-)_{\text{expt}}$ depends linearly on these elementary intensities

$$(I_+ + I_-)_{\text{expt}} = \sigma_{p1} I_{p1} + \sigma_{p0} I_{p0} + \sigma_{s0} I_{s0},$$

where the coefficients σ_{p1} , σ_{p0} , and σ_{s0} are the initial populations. Therefore a linear three parameter fit gives these populations. In fact, the two curves $I_{p1}(\epsilon)$ and $I_{p0}(\epsilon)$ are so close that the fit does not allow accurate individual determination of σ_{p1} and σ_{p0} , but gives only their sum.

6. Coherence sp

Let $I_{sp}(\epsilon)$ be the simulated difference $(I_+ - I_-)$ for a coherence normalized to unity. The coherence σ_{sp} can be deduced from the experimental difference $(I_+ - I_-)_{\text{expt}}$ through the following expression: $(I_+ - I_-)_{\text{expt}} = \sigma_{sp} I_{sp}$.

Since the difference $(I_+ - I_-)$ is found to be very small in our data the coherence term σ_{sp} appears to be negligible

where the states $|j, |m_j| \rangle$ are written in Latin letters and the eigenstates of H_S in Greek letters. ω_α and ω_β are the complex eigenvalues of the Hamiltonian.

4. Intensity of the emitted light

The detection operator D associated with the measurement of the emitted light in a given direction and with a given polarization, is defined by

$$D = (\vec{\epsilon}_\lambda, \vec{d}) P_0 (\vec{\epsilon}_\lambda, \vec{d})^\dagger,$$

where \vec{d} is the dipole electric operator, $\vec{\epsilon}_\lambda$ the polarization operator, and P_0 the projector of the lower level. As our observation is made at 90° with respect to the beam, and without the polarizer, the detection matrix is

in the four experimental conditions. In conclusion this theoretical treatment leads to the direct determination of the relative populations of the 2s and 2p levels.

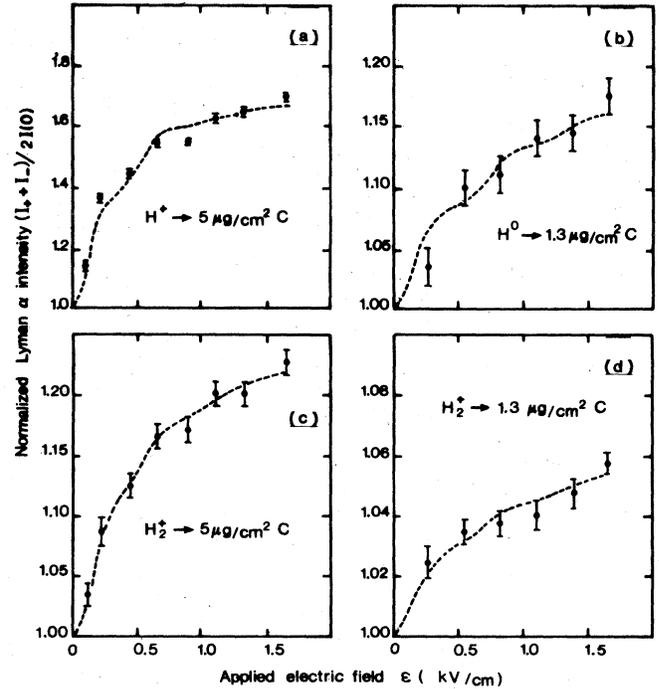


FIG. 2. Dependence of the normalized sum $(I_+ + I_-)/2I(0)$ on the electric field in the four studied cases and computer fits (dashed lines) from which the values of R are deduced.

TABLE I. Ratio R of the $2p$ and $2s$ populations for the following excitation mechanisms: (a) capture of a target electron by H^+ projectiles; (b) direct excitation of incident H^0 projectiles; (c) production of excited hydrogen atoms by dissociation of reconstituted molecular ions; (d) production of excited hydrogen atoms from dissociation of original H_2^+ projectiles. The variations of the $2s$ population are also given (in parentheses), starting from the value 1 in the case (a).

Target thickness ($\mu\text{g}/\text{cm}^2$)	Incident projectile	
	Atomic	Molecular
5	(a) 1.22 ± 0.05 (1)	(c) 2.7 ± 0.1 (0.89 ± 0.07)
1.3	(b) 3.3 ± 0.4 (0.74 ± 0.1)	(d) 4.3 ± 0.2 (1 ± 0.09)

C. Results and discussion

In Fig. 2 the dependence of the normalized sum $(I_+ + I_-)/2I(0)$ on the electric field is shown, along with the best computer fits (dashed lines) from which the values of R (the ratio of the $2p$ and $2s$ populations) can be deduced.

In Table I the R values corresponding to our four experimental conditions are given. They range from a value close to 1 in the case of electron capture by an emergent proton to more than 4 when the hydrogen atom results from the excitation of an incident H_2^+ molecule.

We first discuss the values obtained in (a) and (b), where the incident projectiles are protons and hydrogen atoms, respectively. The only experimental study available for comparison is work by Gabrielse¹⁰ who has measured the energy dependence of R for incident protons by means of an interference beat technique. A value of 1.1 can be extracted from his data for our velocity, in good agreement with our value of 1.22.

A theoretical study of projectile excitation in a binary collision has been done by Yager and Lane¹⁷ but unfortunately for He^+ ions. The calculated value, obtained for projectiles of 0.8 MeV/amu, is 4.4 ± 0.5 . This would seem to imply that excitation favors the $2p$ term, in qualitative agreement with what we find with H_0 of the same velocity.

On the other hand, the experimental fact that cases (a) and (b) do not lead to the same values of the $2p$ and $2s$ populations shows that the strong surface field ($\sim 10^8$ V/cm) experienced by the projectiles on emergence^{18,19} does not produce the complete mixing of the atomic levels. Since we know from our previous experimental studies³ that the $2p$ population in the emergent neutral atoms is enhanced by a factor of 2 when electron capture is replaced by direct excitation of an incident H^0 , we can deduce the corresponding change of the $2s$ population

from our knowledge of R . Going from (a) to (b) R is increased by 2.7: it follows that the $2s$ population is reduced by 1.35, thus much less than the $2p$ increase.

In the cases (c) and (d), where the incident projectile is an H_2^+ molecule, the values of R are 2.7 and 4.3, respectively. A comparison of these figures with the R value 1.22 obtained in the case (a) (electron capture by a single proton) shows that the relative increases of R when passing from (a) to (c) and (d) are 2.2 and 3.5, respectively. Again we recall the results of our previous studies³ in which the enhancement of the Lyman- α emission by molecular effects has been measured very precisely: the $2p$ fraction among the neutral atoms was found to have increased by a factor 1.95 in case (c) and by a factor 3.5 in case (d) with respect to the proton beam case (a). The relative $2s$ populations are given in parentheses in Table I.

We are led to the rather remarkable conclusion that the increase of the $n=2$ population by the molecular effects—on either capture or transmission—involves uniquely the $2p$ sublevel; the $2s$ term is not affected. The interpretation of this effect is rather difficult, particularly because six molecular orbitals of H_2^+ lead to the formation of a hydrogen atom in the $n=2$ level at infinite separation. The fact that four of them ($2s\sigma_g$, $3p\sigma_u$, $4f\sigma_u$, $3d\sigma_g$) lead to both $2s$ and $2p$ sublevels whereas the two others ($2p\pi_u$ and $3d\pi_g$) lead to the $2p$ sublevel only is a very crude qualitative explanation of the observed enhancement of the $2p$ population.

In conclusion, the variety of the values obtained for the relative populations of the two sublevels of $n=2$ hydrogen atoms produced by the passage through thin foils of atomic and molecular projectiles shows that very different excitation mechanisms can be involved, and that the $2p$ population is much more sensitive to the nature of the excitation process than the $2s$ population. This study also shows that our understanding of the interaction of very simple projectiles with a solid target is far from being complete.

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