

*FORMATION OF HIGHLY EXCITED HYDROGEN ATOMS BY PROTON CHARGE  
EXCHANGE IN GASES*

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A study was made of the formation of highly excited hydrogen atoms, having principal quantum numbers  $n \geq 8$ , by the charge exchange of 60–180 keV protons in the molecular gases  $H_2$ ,  $N_2$ , and  $CO_2$ . The relative yield of highly excited atoms was determined by ionizing them in a strong electric field (the Lorentz ionization). An estimate of the cross sections for capture into the state with  $n = 10$  gave values of  $10^{-19}$ – $10^{-20}$   $cm^2$  for the cases investigated here.

## INTRODUCTION

RECENTLY Sweetman<sup>[1]</sup> and Hiskes<sup>[2]</sup> have shown that the so-called Lorentz ionization of highly excited hydrogen atoms is important in the injection of fast atomic hydrogen beams into some types of plasma trap employing magnetic mirrors, in the initial stage of plasma accumulation. The essence of this ionization is as follows. When an electric field  $\mathbf{E}$  acts on an atom it reduces the atom's potential barrier on one side only. The same occurs under the action of an equivalent electric field  $\mathbf{E}' = c^{-1}\mathbf{v} \times \mathbf{B}$ , which appears when the atom moves at a velocity  $\mathbf{v}$  in a magnetic field whose induction is  $\mathbf{B}$ . In fields  $\mathbf{E}$  and  $\mathbf{E}'$  of practical interest (of the order of  $1 \times 10^5$  V/cm), the barrier is reduced by an amount close in value to the ionization potentials of highly excited hydrogen atoms (with a principal quantum number  $n$  close to 10). Because of this, it is possible to produce ionization by under- and over-the-barrier transitions. The electric field which causes the ionization of a hydrogen atom in a quantum state, whose principal quantum number is  $n$ , is approximately proportional to  $n^{-4}$ . In the case of under the barrier transitions, the value of the field which destroys such a state also depends on the residence time of the atom in the given field.<sup>[3,4]</sup> In practice, all atoms with  $n \geq 10$  should be ionized in fields  $E \geq 1 \times 10^5$  V/cm in a time not longer than  $1 \times 10^{-10}$  sec. In the present work, we investigated highly excited atoms with  $n \geq 8$ .

Various data on the properties of highly excited atoms can be found in Bethe and Salpeter's monograph<sup>[3]</sup> and in the work of Rice and Good.<sup>[4]</sup> Interesting practical data on the de-excitation life-

time of highly excited atoms are given in the theoretical work of Hiskes et al.<sup>[6]</sup>

The formation of fast highly excited hydrogen atoms is possible in proton charge exchange and the dissociation of molecular hydrogen ions during their passage through gases,<sup>[1,7,8]</sup> vapors<sup>[9]</sup> and gas-discharge plasma.<sup>[10]</sup> For velocities  $v \geq 1 \times 10^8$  cm/sec, it is assumed<sup>[2]</sup> that the probability of charge exchange to levels with sufficiently high values of  $n$  is proportional to  $n^{-3}$ .

The present work was carried out in order to study the efficiency of proton charge exchange to highly excited states of H in molecular gases, for which there were practically no data. The proton energy was 60–180 keV, which corresponded approximately to energies of particles injected into plasma traps.

## MEASUREMENT METHOD

The method we employed for measuring the relative number of highly excited atoms in an atomic beam, using dissociating electric fields, was suggested by Riviere and Sweetman.<sup>[1,11]</sup> This method makes it possible to determine the percentage content of highly excited atoms in the beam, dissociated by an electric field  $E$ , as a function of the field intensity:  $I(E)$ . The dependence  $I(E)$ , which gives the total number of atoms dissociated by an electric field, is known as the integral dependence. The differential dependence  $dI(E)/dE$  represents the population of various highly excited states of atoms in the beam.

The experimental arrangement is shown in Fig. 1. A monokinetic proton beam, produced by means of apparatus described earlier,<sup>[12]</sup> entered

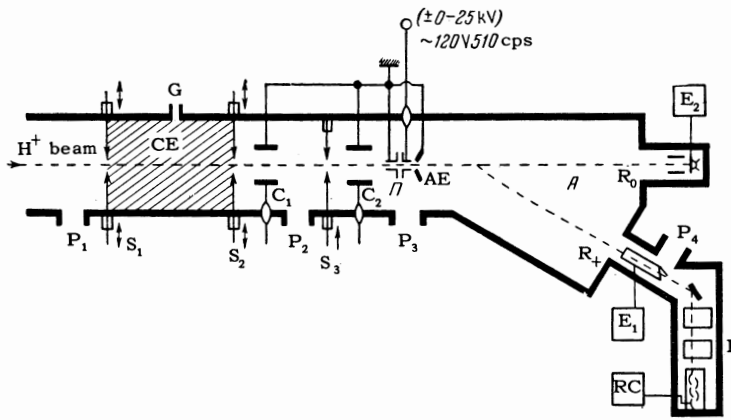


FIG. 1. Layout of apparatus: CE is the charge-exchange chamber; G is the gas input to this chamber;  $C_1$ ,  $C_2$  are the filtering capacitors; D is the dissociation gap; AE is the auxiliary electrode; A is the magnetic analyzer;  $R_+$  and  $R_0$  are, respectively, the proton and atom receivers; I is the ion counter;  $E_1$  and  $E_2$  are the electrometers; RC is the phase-sensitive recording circuit;  $S_1$ ,  $S_2$  and  $S_3$  are the collimating slits;  $P_1$ ,  $P_2$ ,  $P_3$  and  $P_4$  are the vacuum pumps.

a charge-exchange chamber CE. The pressure in the chamber was such that in most of the experiments only single collisions took place (the gas target thickness was of the order of  $10^{-2}$  mm Hg  $\times$  cm). Next, the atomic hydrogen beam, formed in the chamber CE, was passed through capacitors  $C_1$  and  $C_2$  in which a weak electric field (of the order of 1 kV/cm) extracted the charged particles. The system of slits  $S_1$ ,  $S_2$ , and  $S_3$  facilitated differential pumping and improved the vacuum conditions in the analyzer part of the apparatus. The pressure in the analyzer did not exceed  $5 \times 10^{-6}$  mm Hg and was almost independent of the pressure in the chamber CE. Next, the atomic beam entered the dissociation (ionization) gap D, across which a strong electric field (up to  $2 \times 10^5$  V/cm) was applied. The gap consisted of two stainless steel disks of 10 mm diameter with an aperture of 1 mm diameter for the passage of the beam. The gap between the disks was 1 mm. Having entered the gap, the beam was restricted by a diaphragm of 0.35 mm diameter. The electrode AE was to reduce the defocusing action of the gap. The field along the axis of the gap was calculated, at the request of the present authors, in the Mathematical Physics division of the Physico-technical Institute of the U.S.S.R. Academy of Sciences. The results of the calculation were in good agreement with measurements carried out in an electrolytic tank. The field distribution in the gap is shown in Fig. 2.

Protons, formed when highly excited hydrogen atoms are dissociated (ionized) by the electric field, were directed by the magnetic field of the analyzer A either into the Faraday type receiver  $R_+$  (then the current was measured with the electrometer  $E_1$ ) or to the ion counter I. The construction of the counter is described in detail in [13]. Control tests showed that the width of the line in the receiver  $R_+$  was less than the dimensions of the entry window of the receiver. The line could

broaden due to the defocusing action of the gap G and the scatter of the secondary proton energy associated with the formation of the protons in the gap field.

The atomic flux was recorded with the receiver  $R_0$ , using secondary electron emission and the electrometer  $E_2$ . The construction of the receiver  $R_0$  is described in [12]. The receiver was calibrated by the thermal effect of a beam. The total atomic hydrogen flux through the gap G represented a current of  $2 \times 10^{-11}$ – $5 \times 10^{-10}$  A and the proton current formed in the gap ( $i_+$ ) was  $2 \times 10^{-13}$ – $5 \times 10^{-12}$  A.

The integral dependence  $I(E)$  was determined from the proton current and the atomic flux  $i_0$  using the formula

$$I(E) = i_+(E) / i_0 - i_+(0) / i_0, \quad (1)$$

where the current  $i_+(0)$  is associated with the stripping of hydrogen atoms in the residual gas, and  $i_+(E)$  is the proton current in a field  $E$ . The differential dependence was measured directly by superimposing a weak alternating field  $\Delta E$  (of 1.3 kV/cm amplitude and 510 cps frequency) on a con-

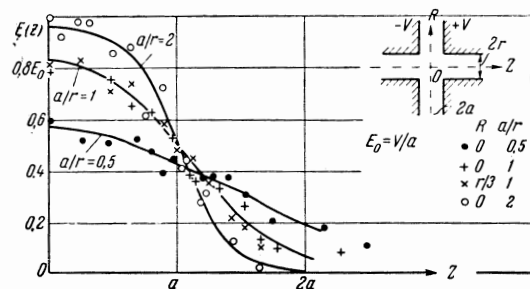


FIG. 2. The distribution of fields in the dissociation gap for various distances between the electrodes. The continuous curves represent the calculations, while the points represent the results of measurements in an electrolytic tank. The configuration and dimensions of the electrodes are shown on the right.

stant ionizing field  $E$ . The alternating component of the proton current  $\Delta i_+$ , measured with the counting system of  $I$ , was recorded with a resonance amplifier and a phase-sensitive recording circuit RC. The value of  $\Delta i_+/\Delta E$ , referred to the atomic flux  $i_0$ , was proportional to  $dI/dE$ . The absolute value of  $dI/dE$  could be determined from the dependence  $I(E)$ .

The experimentally determined population of states may differ from the initial population if a considerable number of highly excited atoms becomes de-excited between the charge-exchange chamber and the gap. To check this, a special control test was carried out in which the  $I(E)$  dependence was recorded twice: once with a space of 40 cm between the charge-exchange chamber and the gap (this distance was used in all other measurements), and a second time with a separation of 80 cm. The dependences  $I(E)$  were in both cases identical within the experimental error. This indicated that the lifetime of highly excited hydrogen atoms in the presence of de-excitation was considerably longer than the time of flight and the experimental dependences  $I(E)$  represented the initial conditions.

Random errors in the determination of  $I$  did not exceed  $\pm 15\%$ .

## RESULTS AND DISCUSSION

We measured the integral dependences  $I(E)$  for highly excited hydrogen atoms formed by charge exchange of 60, 120 and 180 keV protons in molecular gases  $H_2$ ,  $N_2$  and  $CO_2$  under single-collision conditions. The results of the measurements are given in Fig. 3. Measurements were also carried out at 60 keV for charge exchange in Ne, Ar, and He and at 120 keV in He (the results are given below in Table II).

It follows from Fig. 3 that in a constant ionizing field the values of  $I$  for all gases, except molecular hydrogen, do not differ by more than 15%. Hydrogen gives values of  $I$  about 30% higher than nitrogen and carbon dioxide. On increase of the atomic beam energy, the yield of highly excited atoms becomes greater. The threshold in the integral dependence  $I(E)$  is due to the fact that in the calculation of  $I(E)$  it was not possible to include some of the excited atoms with the highest quantum number  $n$ , ionized by the magnetic field on entering the analyzer A (equivalent to an electric field  $E'$  of the order of 10 kV/cm). The current of protons formed in this way was included in the value of  $i_+(0)$ .

Sweetman's results<sup>[1]</sup> for  $I(E)$ , obtained at 200

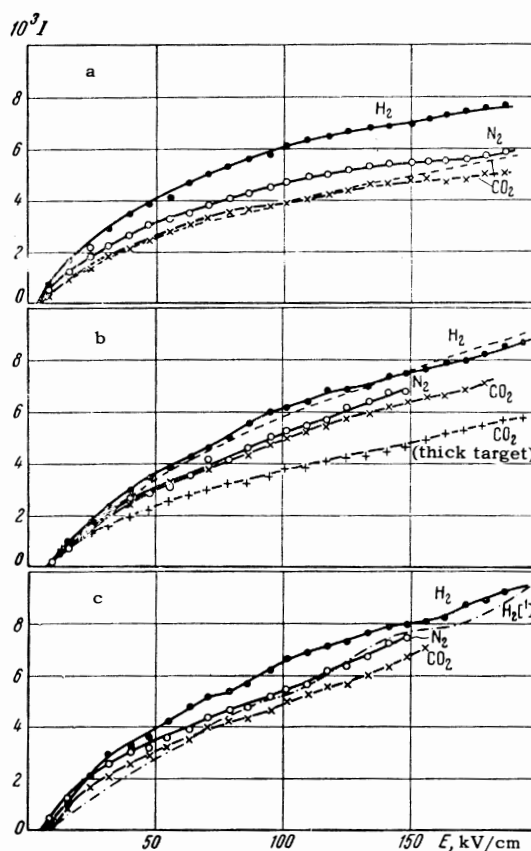


FIG. 3. Integral dependences  $I(E)$  for various molecular gases at the energies: a) 60 keV; b) 120 keV; c) 180 keV. The continuous curves represent the results of the present work; the dashed curves are the results calculated using Eq.(3); the chain curves give the data of Sweetman<sup>[1]</sup> for hydrogen at 200 keV.

keV with molecular hydrogen, agreed within the experimental error with the analogous data obtained by us for 180 keV (Fig. 3c).

The differential dependence  $dI(E)/dE$ , measured for the charge exchange of 120 keV protons in hydrogen, is shown in Fig. 4. The presence of a threshold in weak fields is due to the fact that highly excited atoms with  $n \gtrsim 20$  are ionized in the electric fields of  $C_1$  and  $C_2$  (Fig. 1) before reach-

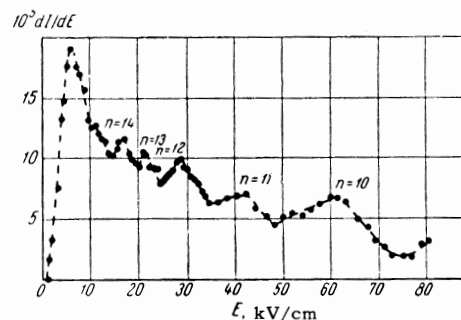


FIG. 4. Differential dependence  $dI/dE$  for the charge exchange of 120 keV protons in molecular hydrogen.

ing the gap D. The maxima in the dependence  $dI/dE$  represent the quantum states with  $n = 10-14$  (the states with  $n > 15$  were not resolved in our experiments). The positions of the maxima in these curves agreed with the results of Riviere and Sweetman<sup>[7]</sup> and those of Futch and Damm.<sup>[9]</sup>

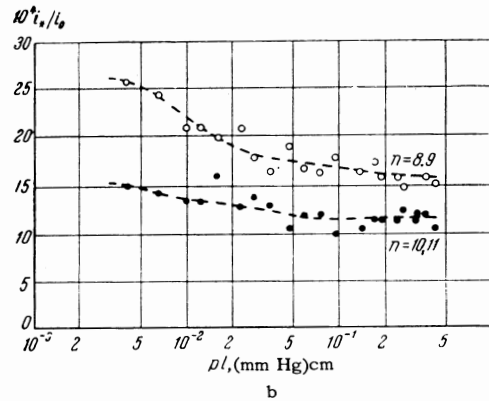
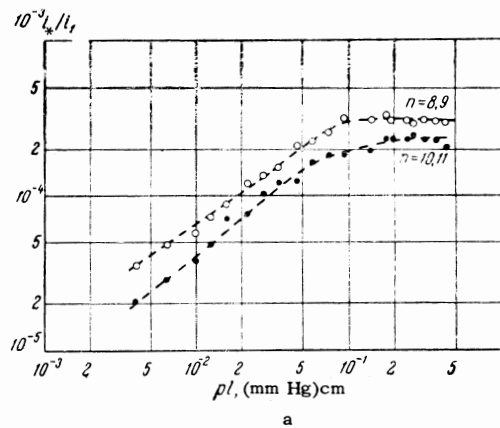
The relative population of the states with different values of  $n$  could be found approximately from the areas under the corresponding maxima in the differential curve. The values of the absolute population could be obtained from the integral dependence, as a difference  $I(E_1) - I(E_2)$ , where  $E_1$  and  $E_2$  are found from the positions of the minima in the  $dI/dE$  curve, which are on either side of a peak corresponding to a given value of  $n$ . In both cases, the population of a state with a given  $n$  was found to be approximately proportional to  $n^{-3}$ . The accuracy of the determination of the power exponent of this dependence was not great: it did not exceed  $1/2$ .

The information obtained on the population of various highly excited states of hydrogen could be used to estimate the cross section for the charge exchange of  $H^+$  to these states. The cross section for the charge exchange to a state with a given  $n$  could be found as the product of the population of a given state and the total cross section for the charge exchange of  $H^+$ . The estimated values of the cross sections for the charge exchange to the state with  $n = 10$  are listed in Table I, the total charge-exchange cross sections being taken from<sup>[14]</sup> and our preliminary experiments.

In practice, we are interested not only in the relative content of highly excited atoms in an atomic beam, but also in the number of highly excited hydrogen atoms compared with the primary proton beam. In both instances, it is important to know the relative yield of highly excited atoms when the thickness of the gas target is varied. To this end, we measured the dependence of the yield of highly excited hydrogen atoms on the pressure in the case of the charge exchange of 120 keV protons in  $CO_2$ . In determining the dependence of the

**Table I.** Cross sections for charge exchange to a state with  $n = 10$  (in  $10^{-20} \text{ cm}^2$  per molecule)

Gas	$H^+$ energy, keV		
	60	120	180
$H_2$	11	1.8	0.4
$N_2$	25	8	2.5
$CO_2$	38		
He	6	1.8	



**FIG. 5.** Dependence of the yield of highly excited hydrogen atoms on pressure: a) relative to the primary  $H^+$  beam; b) relative to the atomic beam;  $i_*$  is the flux of atoms in the indicated states,  $i_0$  is the total flux of atoms,  $i_1$  is the primary proton beam current,  $pl$  is the gas target thickness.

yield, compared with the primary proton beam, the intensity of the latter was found past the gap by adding the proton and neutral components of the beam.

The dependence of the yield of highly excited hydrogen atoms on the target thickness, relative to the primary  $H^+$  beam, is shown in Fig. 5a, and relative to the atomic beam in Fig. 5b. The curves show the relative yield of highly excited hydrogen atoms for the sum of states with  $n = 8$  and  $9$ ,<sup>1)</sup> and for  $n = 10$  and  $11$  (the ionizing fields were 80–160 and 40–80 kV/cm). The yield of highly excited atoms, relative to the primary proton beam, increased with increase of the target thickness right up to 0.1 mm Hg  $\times$  cm, at which a charge equilibrium was established in the beam. The yield of highly excited atoms, relative to the atomic beam, was of a different nature. With increase of

<sup>1)</sup>The region of dissociating (ionizing) fields for  $n = 8$  or  $9$ , was obtained by extrapolation of the results in Fig. 4, allowing for the fact that the dissociating field is proportional to  $n^{-4}$ .

Table II. Values of the parameter a from various sources

Gas	H <sup>+</sup> energy, keV												
	18	25	37.5	42	50	60	62,5	75	87,5	100	120	180	200
He					(0,65)	0,7							
Ne					(0,65)	0,7							
Ar					(0,85)	0,7							
Kr					(0,75)								
Xe					(1,0)								
H <sub>2</sub>		{0,72 (0,3)	1,1 (0,6)		1,7 (0,9)	1,1	1,7 (1,1)	1,8 (1,3)	1,8 (1,2)	1,9 (1,3)	1,3	1,3	1,3
N <sub>2</sub>						0,85					1,1	1,1	
CO <sub>2</sub>						0,75				1,1(0,6)			
H <sub>2</sub> O	(0,6)			(0,7)									
Li	1,7												
n*	9-13	14	14	9-13	14	8-11	9-14	14	14	14	14	9-14	9-14
Source	[7]	[*]	[*]	[*]	[*]	[7]	**	[*]	[*]	[*]	[*]	**	**

\*Gives the range of n in calculations  
 \*\*Present work

the target thickness, there was some depletion of highly excited states, in agreement with the results of [5,8].

As mentioned earlier, the population of a level with a principal quantum number n may be expressed by a dependence a/n<sup>3</sup>, where a is a parameter which depends on the nature of the gas and on the energy, and is therefore the principal characteristic of the efficiency of a charge-exchange target. Table II lists the values of a obtained in the present work and also those found from the published data. This table gives the values of a both for the case of single collisions and for the case of a "thick" target, which ensures charge equilibrium in the charge beam. The values for the latter case are given in parentheses. By and large, the value of a was determined from a part of the I(E) dependence covering several values of n. The exception in this respect was the work of Riviere and Sweetman, [8] who listed only the data for n = 14; therefore, their value of a was less exact. The table gives the proton energy; the results for deuterons [7,9] are included in the column corresponding to half their energy.

It is evident from Table II that the highest yield of highly excited atoms at 25-200 keV is obtained for hydrogen. For lithium vapor, the efficiency at 18 keV is comparable with the efficiency for hydrogen at optimum energies. Unfortunately, there are no other data on alkali metal vapors, which are efficient charge-exchange targets.

For practical calculations, it is convenient to express the dependence I(E) in the analytic form. The semi-empirical formula, giving this dependence for the case of charge-exchange of protons to highly excited states, has the form

$$I(E) = (a / 2\sqrt{b})\sqrt{E}, \tag{2}$$

where I(E) is a dimensionless quantity which represents the fraction of excited hydrogen atoms in

an atomic beam. This formula is based on the assumption that the relative population of a state with a given n is a/n<sup>3</sup> and the electric field destroying the state is b/n<sup>4</sup>, where n has a continuous range of values. The values of the dimensionless parameter a are listed in Table II. The parameter b is found from the positions of the maxima of the dependence dI/dE. According to the data in Fig. 4, the value of this parameter is b = 6.2 × 10<sup>5</sup> kV/cm. Similar values of b may be deduced from the data given in [7,9]. The value of b decreases slowly with increase of the residence time of highly excited atoms in the dissociating field (for an increase in this time by three orders of magnitude, i.e., up to approximately 1 × 10<sup>-7</sup> sec, the value of b varies no more than 25% [4]).

Substituting the value of b thus obtained into Eq. (2), we find that

$$I(E) = 6.4 \cdot 10^{-4} a\sqrt{E}, \tag{3}$$

where E is expressed in kV/cm. The dependences calculated using Eq. (3) were in good agreement with the experimental dependences for E > 2 × 10<sup>4</sup> V/cm (Fig. 3a and 3b). The calculated curves were fitted to the experimental ones at E of the order of 1 × 10<sup>5</sup> V/cm because, as mentioned previously, it was not possible to measure I(E) experimentally at E < 1 × 10<sup>4</sup> V/cm. Equation (3) allows us to estimate the relative number of protons captured in the active zone of a trap.

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