

*PRODUCTION OF HIGHLY EXCITED HYDROGEN ATOMS BY CHARGE EXCHANGE  
OF PROTONS IN METAL VAPORS*

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We determine the relative population of the highly excited states of hydrogen atoms with principal quantum numbers  $9 \leq n \leq 16$  in charge exchange of protons of energy 10-180 keV in vapor of Mg, Ca, Zn, and Cd. For Mg and Cd we investigate also the yield of highly-excited atoms relative to the primary proton beam as a function of the vapor pressure. A comparison is made of the efficiency of production of highly excited hydrogen atoms for targets made of alkali-metal vapors, vapors of metals of group II of the periodic table, and gases. It is established that Mg is the most effective target at proton energies below 50 keV. The features of the mechanism whereby the electron is captured in a highly excited state of the hydrogen atom are discussed. Estimates are presented of the cross sections for the stripping of fast hydrogen atoms in the ground and excited states.

## 1. INTRODUCTION

THIS investigation is a continuation of a cycle of studies<sup>[1-3]</sup> of the efficiency of charge exchange of protons in highly-excited states of the hydrogen atom in gases and metal vapors. These studies were undertaken in connection with the possible use of Lorentz ionization of highly-excited hydrogen atoms to accumulate plasma in magnetic traps.<sup>[4-6]</sup> The charge-exchange targets investigated in<sup>[1-3]</sup> were molecular and inert gases and alkali-metal vapors. The studies revealed certain essential features of the processes of charge exchange of protons in alkali-metal vapors with production of highly-excited hydrogen atoms. At low energies (<30 keV), the efficiency of production of highly-excited hydrogen atoms is much larger than at high energies (> 60 keV), and exceeds by almost one order of magnitude the efficiency of gas targets. The reason is that at low energies the electron participating in the charge exchange is the outer, weakly-bound electron of the alkali-metal atom, whereas inner electrons take part at high energies.

It is of interest in this connection to continue the investigations of the influence of the structure of the target atoms and the electron binding energy on the formation of highly-excited hydrogen atoms in proton charge exchange. In the present investigation the targets chosen were metals of group II of the periodic table: Mg, Ca, Zn, and Cd, whose

atoms, like the atoms of the alkali-metals, have a small number of electrons in the outer shell. It is also known that vapors of group II metals are more convenient in practice, owing to their lower chemical reactivity.

## 2. MEASUREMENT PROCEDURE

The experimental setup and the method used to measure the relative number of highly-excited atoms in the atomic beam, based on their destruction by a strong electric field, were described in our earlier papers.<sup>[1,2]</sup> Just as in our earlier work, the primary-proton energy  $T$  ranged from 10 to 180 keV.

In the present work we measured the relative number of highly-excited atoms,  $I$ , ionized by the field  $E$ . If the population of the states with principal quantum number  $n$  is  $a/n^3$ , in agreement with experiment<sup>[1-3,7,8]</sup> for  $n \gg 1$ , then

$$I(E) = 6.4 \cdot 10^{-4} a \sqrt{E}, \quad (1)$$

where  $E$  is in kilovolts per centimeter.

The dimensionless quantity  $a$  depends on the type of target and is the principal measure of the efficiency of the charge-exchange target for the production of highly-excited hydrogen atoms. Knowing the value of  $a$  in the case of single collisions (thin target) and the total proton charge exchange cross section  $\sigma_0$ , we can determine the cross section  $\sigma_c^n$  for the production of highly-

excited atoms with principal quantum number  $n$  by charge exchange:

$$n^3\sigma_c^n = a\sigma_0. \quad (2)$$

However, the method of determining the vapor pressure from the chamber temperature, which we use in the present work, does not make it possible to measure the absolute value of the cross sections  $\sigma_0$  and  $\sigma_c^n$  with acceptable accuracy.<sup>1)</sup> Nonetheless this method makes it possible to estimate the indicated cross sections and to carry out a comparative analysis of the relative dependences of  $\sigma_c^n(T)$  for different charge-exchange targets. To this end we determined, on the basis of (1) and (2) the relative cross section

$$Q^n = \sigma_c^n / \sigma_{c \max}^n, \quad (3)$$

where  $\sigma_{c \max}^n$  is the maximum value of the cross section  $\sigma_c^n$ .

For practical problems, interest attaches to the so-called "thick target," in which the fast particles experience multiple collisions. In this case, measurements of the ratio of the total number of fast atoms produced in the charge exchange to the number of particles in the beam  $\Phi_0$  (neutral fraction), are important, as well as measurements of  $\Phi_n$ —the fraction of the highly-excited atoms with principal quantum number  $n$  as functions of the pressure in the charge-exchange chamber. For a thick target, just as for a thin target (this is shown in [3]), the values of  $\Phi_n$  can be determined on the basis of relations similar to (1) and (2).

Random errors in the measurement of the individual quantities, namely  $a$  and  $Q^n$  for the thin target and  $\Phi_0$  and  $\Phi_n$  for the thick target, as estimated from the reproducibility of the results, did not exceed in our investigation  $\pm (15-20)\%$ .

### 3. RESULTS AND DISCUSSION

#### A. Thin Target

1. Relative population of highly-excited states of hydrogen atoms. Figure 1 shows the values of  $a$ , which characterize the population of the highly-excited hydrogen atoms, as function of the proton energy  $T$ . The values of the principal quantum number  $n$ , for which the values of  $a$  were determined, lie in the range  $9 \leq n \leq 16$ .

<sup>1)</sup>The cross sections  $\sigma_0$  and  $\sigma_c^n$  quoted for Mg in [2] were measured less accurately than in Alkali-metal vapors. Nonetheless, the values of  $\sigma_0$  for Mg obtained in [9], are very close to our data in [2].

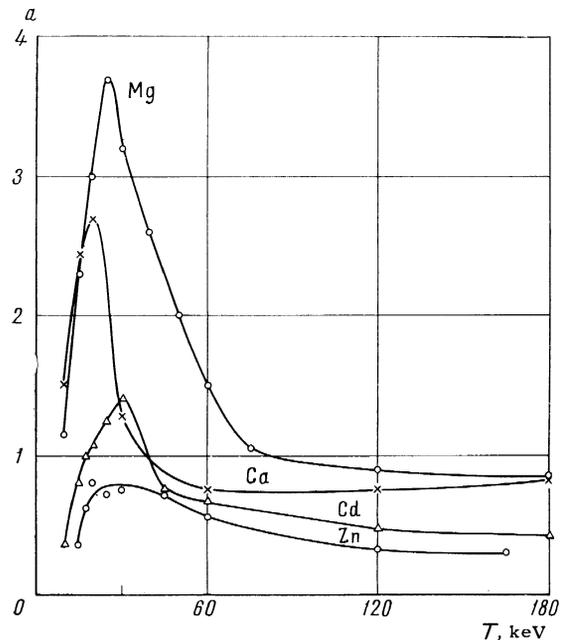


FIG. 1. Quantity  $a$  (thin target) as a function of the proton energy. The charge-exchange targets are marked on the corresponding curves.

It is seen from the figure that the  $a(T)$  plots for all the metals have a clearly pronounced maximum in the 25–30 keV region, and change little at energies above 60 keV. One can also note that the energy at which  $a$  is a maximum increases with increasing ionization potential of the metal atom, and the maximum on the  $a(T)$  curve becomes broader. Such tendencies are exhibited also by the alkali-metals investigated by us in [3]. This agrees with the results of calculations by Hiskes,<sup>[10]</sup> who showed that in the case of charge exchange of protons in hydrogenlike targets with formation of highly-excited hydrogen atoms, the maximum of  $a$  shifts toward higher energies with decreasing principal quantum number of the outer electron of the target (i.e., with increasing ionization potential).

We note also that the maximum of the quantity  $a$  is much smaller for Zn and Cd than for Mg and Ca.

2. Cross sections for the production of highly-excited hydrogen atoms in proton charge exchange. To compare the  $\sigma_c^n(T)$  dependences for different targets, Fig. 2 shows the corresponding values of  $Q^n$  for Mg, Ca, Zn, and Cd, obtained in our work, and for Na and K obtained from the earlier work.<sup>[3]</sup> It is seen from the figure that the  $Q^n(T)$  curves for Mg and Ca have a kink, just as for the alkali metals, and that prior to the kink the values of  $Q^n$  depend strongly on the proton energy, and after the kink they change little. This gives grounds for as-

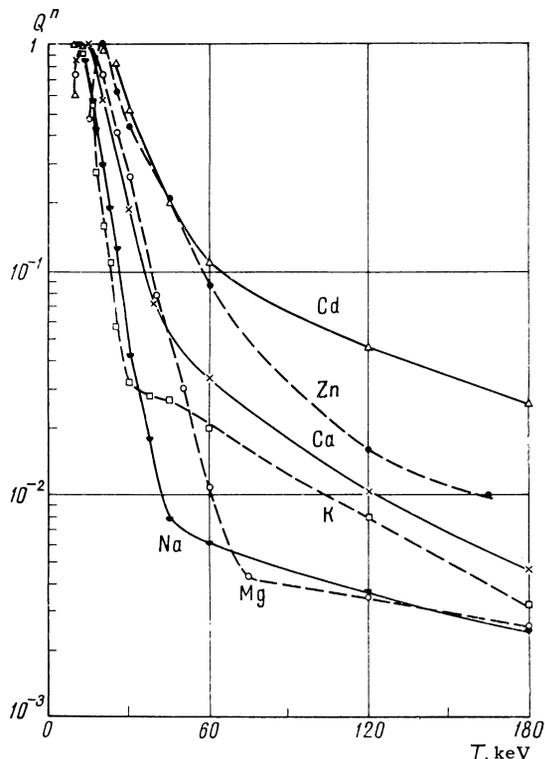


FIG. 2. Relative cross section  $Q^n$  vs. proton energy. The charge-exchange targets are indicated on the corresponding curves.

suming that in the case of alkali-earth metals at low energies, the charge exchange is with the outer electrons, and at high energies with the inner ones.

It is interesting to trace the dependence of the position of the maximum on the  $\sigma_C^n(T)$  curves on the binding energy of the outer electron in the target atom. Figure 3 shows, on the basis of the present data and those of our preceding work,<sup>[1-3]</sup> the proton energy  $T_{\max}$  corresponding to the maximum cross section  $n^3 \sigma_C^n$ , as a function of the binding energy  $V_1$  of the outer electron of the tar-

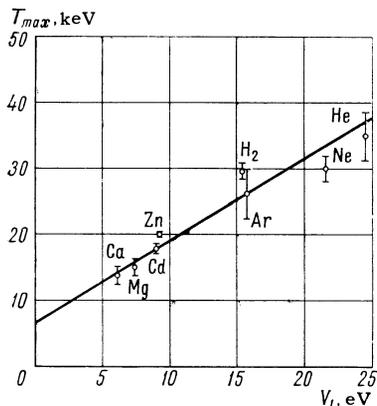


FIG. 3. Proton energy  $T_{\max}$  corresponding to the maximum cross section  $\sigma_C^n$  vs. the first ionization potential of the target atoms.

get atom (first ionization potential). It can be seen on the figure that the function  $T_{\max}(V_1)$  can be approximated by a direct proportionality. On the other hand, the proton velocity corresponding to  $T_{\max}$  turns out to be close to  $\sqrt{2V_1/m}$  (where  $m$  is the electron mass), which characterizes the velocity of the electron on the outer orbit of the target atom. It thus turns out that the greatest probability of electron capture in a highly-excited state of the hydrogen atom (when the electron goes over from the discrete state to a state close to the continuum), occurs at a proton velocity close to the velocity of the outer electron in the target atom. This agrees with quasiclassical notions concerning atomic collisions.<sup>[11]</sup>

Let us consider the question of the applicability of the adiabatic hypothesis<sup>[12]</sup> to the capture of an electron in a highly-excited state of the hydrogen atom. According to this hypothesis, the greatest charge-exchange probability occurs at a proton velocity

$$v_m \approx \alpha |\Delta E| / \hbar, \quad (4)$$

where  $\Delta E$  is the change of the internal energy upon collision, and  $\alpha$  is a quantity of the order of the atomic dimensions. In a review by Fogel,<sup>[13]</sup> and the book by Hasted,<sup>[14]</sup> it is noted that for the capture of one or two electrons by an atom and a singly-charged ion, in which the transition occurs between discrete states, the value of  $\alpha$  turns out to depend on the type of the process and does not depend on the nature of the colliding particles. If this were to be valid also for charge exchange with production of a highly-excited hydrogen atom, when  $\Delta E \approx V_1$ , then the proportionality  $v_m \propto V_1$  should remain. Our experimental data (Fig. 3) do not confirm such a dependence. In our case the relation (4) can be satisfied by assuming that

$$\alpha \propto V_1^{-1/2}. \quad (5)$$

It follows therefore that for the capture of an electron in a highly-excited state of a hydrogen atom, we obviously cannot regard the quantity  $\alpha$  in the adiabatic criterion as a constant for different targets. A similar conclusion for the loss of an electron by a fast ion (when the electron goes over from the discrete state to the continuous spectrum) is contained in a paper by Nikolaev.<sup>[15]</sup>

The relation  $v_m \propto V_1^{1/2}$ , which follows from our experimental data, can also be obtained theoretically from an analysis of the Bohr formula. This question was considered by Drukarev.<sup>[16]</sup>

We note also that the position of the kink on the  $Q^n(T)$  and  $\sigma_C^n(T)$  plots for alkali metals and for Mg and Ca corresponds to proton velocities close

to  $(2V_j/m)^{1/2}$ , where  $V_j$  is the binding energy of the electrons of the nearest internal subshell, i.e., close to the velocity of these electrons. This circumstance is an additional confirmation of our assumption that at high energies the charge exchange of protons with atoms of alkali and alkali-earth metals is realized essentially with the internal electrons. In the case of Zn and Cd, the binding energies of the outer electrons are close to the binding energies of the electrons of the next subshell (d-electrons). This may be the cause of the smooth decrease of  $Q^n(T)$  and the small values of  $a$  for the indicated metals.

Certain interest attaches also to the question of the factors influencing the maximum cross section for the capture of an electron in a highly-excited state of the hydrogen atom ( $\sigma_c^n$ ). A similar question for the cross section of nonresonant single-electron charge exchange in the ground state of the atom was considered by Fedorenko and Belyaev;<sup>[17]</sup> it was noted that  $\sigma_{max}$  is influenced essentially by the defect of the resonance of the reaction  $\Delta E$  and by the mass of the target atom. When the electron is captured in a highly-excited state of the hydrogen atom, the maximum cross section  $\sigma_c^n$  turns out to depend only on the first ionization potential of the target atom. The foregoing is illustrated by Fig. 4, which shows data on the function  $\sigma_c^n(V_1)$  for all the cases investigated by us. The values of  $\sigma_c^n$  for the alkali metals<sup>[3]</sup> were taken for a proton energy  $T = 10$  keV, whereas the maximum values of the cross sections should be expected at somewhat

lower energies. The values of  $\sigma_c^n$  for group-II metals obtained in the present paper can be regarded, as already indicated, as estimates only.

As seen from Fig. 4, the data presented for  $\sigma_c^n$  correspond to the same dependence on the ionization potential for all the targets. Exceptions are the points for Ca and Zn, for which the estimates of the absolute cross sections are least reliable. An empirical formula expressing this dependence is

$$n^3 \sigma_c^n = \frac{5.4 \cdot 10^{-13}}{V_1^{5/2}} \text{ cm}^2, \quad (6)$$

where  $V_1$  is in electron volts.

A similar  $\sigma_c^n(V_1)$  dependence can be obtained by simple reasoning, used by Bohr<sup>[11]</sup> to estimate the cross section for the capture of an electron by fast  $\alpha$  particles. According to Bohr, the capture cross section  $\sigma_c$  is presented in the form of a product  $\sigma_c = \sigma_1 f k$ , where  $\sigma_1$  is the cross section for a collision at which an energy of the order of  $mv^2/2$  ( $m =$  electron mass) is transferred to an electron having an orbital velocity comparable with  $v$ ;  $f$  is the probability that capture will take place after the collision;  $k$  is the number of atomic electrons with orbital velocity  $v_e \approx v$ . By examining the motion of the proton in the field of an atom with effective charge  $Z^* = Z/n_1$  (where  $Z$  is the charge of the atomic nucleus and  $n_1$  is the principal quantum number of the captured electron), we obtain an impact parameter  $b = 2Z^2 e^2 / n_1^2 m v^2$  and a collision cross section  $\sigma \approx 4 a_0^2 (Z^2 / n_1^2) (v_0/v)^4$ , where  $a_0 = \hbar^2 / me^2$  and  $v_0 = e^2 / \hbar$ .

The probability of electron capture in a state with principal quantum number  $n_2$  will be expressed in terms of  $f \approx n_2^{-3} (v_0/v)^3$ . Assuming  $k \approx 1$ , we obtain the cross section for the capture of an electron in a state  $n_2$ :

$$\sigma_c^{n_2} \approx 4\pi a_0^2 \frac{Z^2}{n_1^2} \frac{1}{n_2^3} \left(\frac{v_0}{v}\right)^7. \quad (7)$$

If the capture has the greatest probability when  $v \approx v_e = v_0(V_1/V)^{1/2}$  (where  $V_0$  is the ionization potential of the hydrogen atom), then we get from (7), recognizing that  $Z^2/n_1^2 = V_0/V_1$ ,

$$n_2^3 \sigma_c^{n_2} \approx 4\pi a_0^2 (V_0/V_1)^{5/2}. \quad (8)$$

The obtained expression reflects correctly not only the  $\sigma_c^n(V_1)$  dependence, but also gives maximum cross section values which are close to those obtained in the experiments.

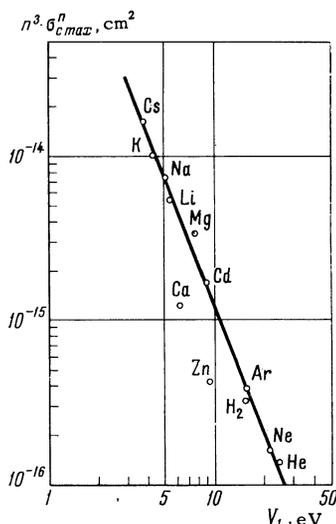


FIG. 4. Dependence of maximum cross section for the capture of an electron in a highly-excited state of the hydrogen atom,  $\sigma_c^n$ , on the first ionization potential of the target atoms.

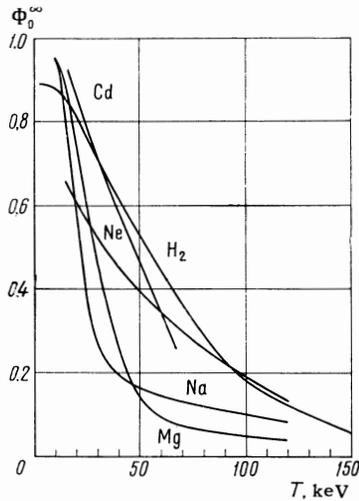


FIG. 5. Equilibrium neutral fraction  $\Phi_0^\infty$  vs. primary-proton energy. The charge-exchange targets are indicated on the corresponding curves.

## B. Charge Exchange of Protons in a Thick Target with Formation of Highly Excited Hydrogen Atoms

1. Composition of the beam. An investigation of the charge exchange of protons with production of highly excited hydrogen atoms in a thick target was carried out in Mg and Cd vapor up to pressures that ensured charge equilibrium in the beam.

Figure 5 shows the dependences of the equilibrium neutral fraction  $\Phi_0^\infty$  for Mg and Cd, obtained

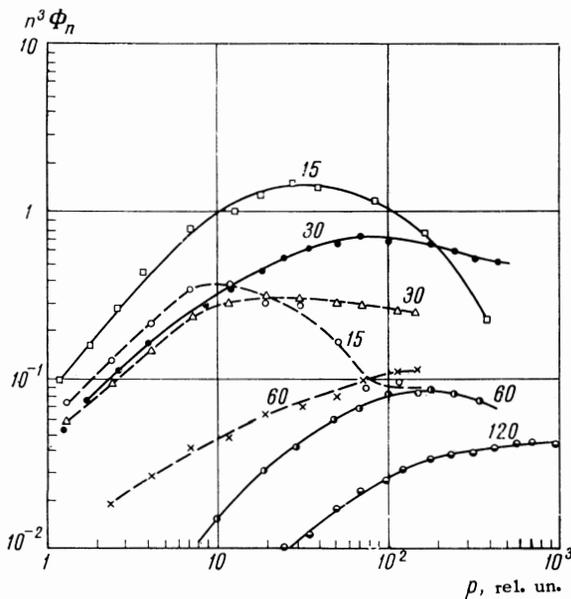


FIG. 6. Fraction  $\Phi_n$  of n-th state of hydrogen atom vs. vapor pressure. Solid curves—charge exchange in Mg, dashed—charge exchange in Cd, the primary-proton energies are shown on the corresponding curves in keV.

in the present investigation, and a comparison with the values of  $\Phi_0^\infty$  for Na and Ne from our earlier paper<sup>[3]</sup> and for H<sub>2</sub> from Allison's paper.<sup>[18]</sup> It can be seen that for Cd, Mg, and Na, at proton energies lower than 15 keV, the yield of neutral atoms during charge exchange is approximately 90% of the number of protons entering the chamber.

Figure 6 shows the obtained  $n^3\Phi_n(p)$  plots, which characterize the yield of the highly excited atoms relative to the primary proton beam for Mg and Cd. It is seen from the figure that at low energies, when charge exchange with the outer electron apparently predominates, the  $n^3\Phi_n(p)$  have a maximum and do not yet reach equilibrium values at pressures at which charge equilibrium has already set in in the beam. At high energies the  $n^3\Phi_n$  tend monotonically to equilibrium values, which are appreciably lower than the maximum of  $n^3\Phi_n$  at low energies. These singularities of  $n^3\Phi_n(p)$  were observed by us also for alkali metals.<sup>[3]</sup>

The equilibrium and maximal values of  $n^3\Phi_n$  as functions of the proton energy are shown in Fig. 7. In addition to the data for Mg and Cd, the same figure shows for comparison data for Na and Ne, obtained by us earlier,<sup>[3]</sup> and for H<sub>2</sub> from data of [7, 18]. As follows from the figure, for Mg the yield of highly excited atoms, referred to the primary proton beam, is larger than for Ne, and appreciably larger than for Cd. At proton energies above 50 keV, the values of  $n^3\Phi_n$  are large for Ne and H<sub>2</sub>.

2. Stripping cross section of fast hydrogen atoms. The available information on the equilibrium fractions and electron capture cross sections enable us to estimate the stripping cross sections of the hydrogen atoms.

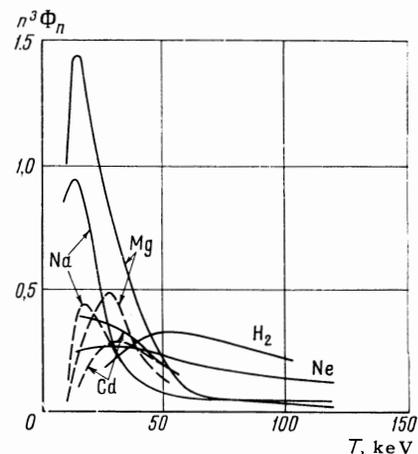


FIG. 7. Fraction  $\Phi_n$  of n-th of hydrogen atom vs. primary-proton energy. Continuous curves—maximum yield, dashed—equilibrium value. The charge-exchange targets are indicated on the corresponding curves.

Stripping cross sections of fast hydrogen atoms in the total electron scattering cross section (in units of  $10^{-16} \text{ cm}^2$ )

	Target											
	Na			Mg			Cd			Ne		
Atom energy, keV	15	30	60	15	30	60	15	30	60	15	30	60
Stripping cross section $\sigma_l^n$ of atoms in highly excited states	20	10	4	$\geq 13$	10	5	13	12	10	1.7	3	2.5
Total electron scattering cross section $\sigma_s$	140	90	60	—	—	—	50	38	37	2.5	2.9	3.4
Total stripping cross section $\sigma_l$	2.4	3.8	3.7	1.7	3.3	2.8	1.5	3.6	5.5	1.7	2.1	2.1
Stripping cross section $\sigma_l^1$ of atoms in ground state	1.3	2.9	3.2	—	—	—	—	—	—	1.5	1.9	2.0

It is of interest to estimate first the stripping cross section of highly excited atoms. It is indicated in [19] that the cross section for the stripping of highly excited hydrogen atoms should not depend on the principal quantum number  $n$  when  $n \gg 1$ , and if we neglect cascade processes that lead to additional excitation, it can be determined from the following formula:

$$\sigma_l^n = \sigma_c^n \Phi_{+^\infty} / \Phi_{n^\infty}, \quad (9)$$

where  $\Phi_{+^\infty}$  is the equilibrium fraction of the protons. It is noted in the same paper that the cross section  $\sigma_l^n$  should be close to the total cross section  $\sigma_s$  for the scattering of electrons having a velocity equal to that of the hydrogen atoms.

We estimated the cross sections  $\sigma_l^n$  by means of (9). The results of the estimates of  $\sigma_l^n$  for different targets, together with the data for  $\sigma_s$  from [20], are listed in the table. As can be seen from the table, in the case of metal-vapor targets the cross section  $\sigma_l^n$  is noticeably smaller than  $\sigma_s$ , whereas for Ne these cross sections have close values. Comparison of  $\sigma_l^n$  and  $\sigma_s$  for other gases from data of [8] shows that they are also close. This is evidence that the mechanism of stripping of highly excited hydrogen atoms in metal vapor is a more complicated process than assumed in [19].

We have also estimated the stripping cross sections of the hydrogen atoms in the ground state  $\sigma_l^1$  and the total stripping cross section  $\sigma_l$ , which are listed in the table.

The cross section  $\sigma_l^1$  was measured in a separate experiment. Two collision chambers were used. In the first, filled with Ne, the proton beam was neutralized, and in the second the fast hydrogen atoms were stripped. The pressure of the gas or of the metal vapor in the second chamber corresponded to the case of single collisions. The sufficiently large distance between chambers and the presence of an electric field in that section made it possible for most excited atoms to go over

into the ground state during the time of flight. On the other hand, the number of long-lived highly excited atoms in the beam was smaller than 1%. [3] This made it possible to regard the stripping cross section measured in this manner as the stripping cross section of the ground state  $\sigma_l^1$ .

The total stripping cross section  $\sigma_l$  was determined in an experiment with a single collision chamber for an equilibrium target. According to [18], this cross section is given by

$$\sigma_l = \sigma_0 \Phi_{+^\infty} / \Phi_0^\infty. \quad (10)$$

In the case of an equilibrium target of metal vapor, the neutral beam contains a large number of excited atoms, and the average time between collisions is comparable with the lifetime of the low-excited states. The cross section for the stripping of the atoms in the excited states in targets made of metal vapor is much higher than the ground-state stripping cross section (as seen from the table), and we find that for these targets the total stripping cross section is appreciably larger than the stripping cross section of the ground state, especially at low energies.

It must be borne in mind, however, that the total stripping cross section  $\sigma_l$  is averaged over all the excited states. Inasmuch as the distribution of the atoms over the low-excited states is unknown to us, the identification of the total stripping cross section  $\sigma_l$  with the stripping cross section of any definite state is impossible in this case.

In the case of gas targets (as seen from the table with Ne as an example), the difference between the cross sections  $\sigma_l$  and  $\sigma_l^1$  is small, since the cross sections  $\sigma_l^n$  are close to  $\sigma_l^1$ , and also since the production of excited hydrogen atoms in gases is less probable.

#### 4. CONCLUSIONS

On the basis of the present results and the earlier papers, [1-3] dealing with charge exchange of

protons with production of highly excited hydrogen atoms, we can draw the following conclusions.

1. In the case of alkali metals and group-II metals, at low energies (<30 keV) the principal role is apparently played by charge exchange with the outer weakly-bound electrons of the metal atom. In this energy region, the cross section for the production of highly-excited hydrogen atoms has a maximum and greatly exceeds the cross section  $\sigma_C^n$  in inert gases, for which the charge exchange takes place on the filled shell. At high energies (>60 keV), the charge exchange is on the internal electrons with relatively high binding energy, and the efficiency of production of highly exciting atoms is much lower than at low energies.

2. The maximum probability of capture of an electron in a highly excited state of the hydrogen atom takes place when the velocity of the proton is close to the velocity of the electron in the target atom. Therefore the region effective for production of highly excited atoms shifts towards higher energies with increasing target ionization potential.

3. For targets of alkali-metal vapor and vapors of group-II metals the pressure dependence of the yield  $\Phi_n(p)$  of the highly excited atoms of hydrogen has a maximum at low proton energies, and equilibrium with respect to excitation sets in at higher pressures than required to establish charge equilibrium.

4. From a comparison of the investigated targets it follows that a target of practical interest in the production of highly excited hydrogen atoms by charge exchange is one of Mg vapor in the energy region below 50 keV, whereas at higher energies preference should be given to gases.

In conclusion, the authors are grateful to Professor N. V. Fedorenko for valuable remarks and constant interest in the work, and are deeply grateful to G. F. Drukarev for a discussion of the results.

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