

an approximately correct value for the cross section would be required. We note that when the cross section is in this region, condition (17) required for the validity of the method of distorted waves is approximately satisfied. Moreover, the short-range contribution to the radial matrix element  $F_2$  must be about the same or less than the approximate long-range contribution, the contribution from  $r > r_0$  given by Eq. (34). Thus, use of the method of distorted waves, or first-order perturbation theory with  $V_0$  included in  $H_0$ , and only fairly approximately correct values for  $V_0(r)$  and  $V_2(r)$  when  $r < r_0$  should lead to a sufficiently accurate determination of

the cross section if  $\alpha_2$  and  $Q$  are known to indicate whether it is  $\gtrsim 0.3\pi/k_i^2$ , and hence of practical significance, or  $\lesssim 0.3\pi/k_i^2$ , and hence of little practical significance. We think that the latter, is more likely the true situation in most cases. It almost certainly is when  $Q$  is negative.<sup>17</sup>

<sup>17</sup> Additional discussion of the short-range region is given in II and III. In the latter it is also shown that within the framework of Stabler's theory the interaction of the electron with the permanent dipole of polar molecular ions such as  $H_2O^+$  or  $NH_3^+$  gives no contribution to the rotational cross section. This results because the angular part of the matrix element yields the selection rule  $|l-l'|=1$ , while the radial part yields  $l=l'$ .

## Production of Highly Excited Neutral Atoms for Injection into Plasma Devices

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We have considered the production of highly excited states of atomic hydrogen by charge-exchange reactions, discussing first the reaction  $H^+ + H(1s) \rightarrow H(\text{excited}) + H^+$ ; we have then used the results for this reaction as a basis with which to compare excited-state production in hydrogen atoms by charge exchange in gases other than atomic hydrogen. We have expressed our results in terms of the equilibrium ratio  $i_n/i_e$ , that is, the ratio of the flux of excited states with principal quantum number  $n$  to the (constant) flux of protons incident on the neutralizer. We find that with atomic hydrogen as neutralizer the ratio  $i_n/i_e$  has a maximum of about  $0.75/n^3$  at 20 keV. For alkali atoms such as lithium and sodium, the maximum value of  $i_n/i_e$  occurs at about 10 keV, being of similar magnitude to, but probably smaller than, the value for atomic hydrogen. In the case of the inert gases helium, neon, and argon, the maximum value of  $i_n/i_e$  occurs in the vicinity of 50–70 keV and again is of similar magnitude to, but probably slightly larger than, the maximum value for atomic hydrogen. The inert gases thus offer a number of advantages over alkali gases as neutralizers: they probably yield as large if not larger values of  $i_n/i_e$  at maximum; the maximum occurs at higher energies, so that the natural decay lengths of the excited atoms are correspondingly longer; they may be very much more easily handled experimentally.

### 1. INTRODUCTION

THE formation of highly excited atomic states from charge-exchange reactions of the type

$$H^+ + H(1s) \rightarrow H(\text{excited}) + H^+$$

is of considerable interest as a source of readily ionizable neutral particles for injection into plasma devices.<sup>1</sup>

In previous publications<sup>2-4</sup> we have calculated the cross section, say  $\sigma_n^e$ , for formation of highly excited atoms with principal quantum number  $n$  from the above exchange reaction. This has the form

$$\sigma_n^e = \frac{256}{5n^3 p^2 \beta^5} \pi a_0^2, \quad (1)$$

<sup>1</sup> D. R. Sweetman, Nucl. Fusion Suppl. 279 (1962), Part I.

<sup>2</sup> S. T. Butler, R. M. May, and I. D. S. Johnston, Phys. Letters 10, 281 (1964).

<sup>3</sup> S. T. Butler and I. D. S. Johnston, Nucl. Fusion 4 (to be published).

<sup>4</sup> R. M. May, Phys. Rev. 136, A669 (1964), and Nucl. Fusion 4, 111 (1964).

where  $a_0$  is the Bohr radius;  $p = (mv/\hbar)a_0$ , where  $m$  is the electron mass and  $v$  is the speed of the incident ions, and

$$\beta = (p^2 + 1)^2 / 4p^2. \quad (2)$$

Within the framework of first-order perturbation theory, Eq. (1) is accurate to order  $1/n^2$ . The cross section (1) shows a marked resonance at  $p=1$  ( $\beta=1$ ) which corresponds to an incident proton energy of 25 keV. This resonance point corresponds to the condition  $\hbar v/e^2 = 1$ , that is, for an incident proton speed equal to the average speed of the electron in its initial 1s state.

Moreover, as we have commented previously,<sup>2</sup> this first-order perturbation result should be reasonably accurate even at  $p \approx 1$ ; this is indicated by an impact parameter analysis along the lines of that performed by May.<sup>5</sup>

<sup>5</sup> R. M. May, Phys. Letters 11, 26 (1964).

It is well known that the first-order perturbation result for the  $1s-1s$  exchange reaction overestimates the cross section in the vicinity of  $p=1$  by a factor of 2 to 3; the impact parameter analysis of Ref. 5, leading to a "corrected Born" result, is required to yield good agreement with experiment at all energies. The impact parameters responsible for the formation of highly excited states are, however, on the average somewhat larger than for the  $1s-1s$  exchange, and the result of the straightforward Born approximation is more reliable in these cases. For detailed accuracy we are in the process of computing a "corrected Born" result to improve Eq. (1), but this equation is completely adequate for the purposes of the present paper.

In this paper we employ the above cross section to predict the actual concentration of highly excited neutrals in the beam emerging from a hydrogen atom exchange medium. For this purpose we need to know the ionization cross section, say  $\sigma_n^i$ , of a highly excited hydrogen atom moving through a medium of ground-state hydrogen atoms. It may readily be seen that, for large  $n$ , this can be identified (to order  $1/n^2$ ) with the total scattering cross section of an electron, moving with the same speed as the high  $n$  state, by a ground-state hydrogen atom. The cross section  $\sigma_n^i$  is thus independent of  $n$  for large  $n$ , and is known from electron scattering data.<sup>6</sup>

In Sec. 2 we evaluate the flux ratio  $i_n/i_c$  in terms of the relevant cross sections, where  $i_c$  is the (constant) flux of protons incident on the exchange medium and  $i_n$  is the emerging flux of neutrals with principal number  $n$ .

The results indicate that the maximum value of the flux ratio  $i_n/i_c$  occurs in the region of 20 keV, where its value is approximately  $0.75/n^3$ . The measurements of Riviere and Sweetman<sup>7</sup> would appear to be roughly consistent with such a predicted value of  $i_n/i_c$ , although they have employed molecular hydrogen as the exchange medium so that no detailed comparison is possible.

In Sec. 3 we discuss the significance of spontaneous radiative transitions of the highly excited atoms on their population in the emerging beam. It is found that the range of  $s$  states against radiative decay is  $(0.42) n^3 p$  cm, of  $p$  states is  $(0.040) n^3 p$  cm, and of  $d$  states  $(0.12) n^3 p$  cm. In the energy range for which  $i_n/i_c$  is a maximum the  $s$  and  $p$  states form the main constituents of the highly excited atoms in the emerging beam.<sup>2</sup> These ranges are such that it should be possible for the highly excited neutrals to be used in experimental devices.

In Sec. 4 we comment on the manner in which the equilibrium ratio  $i_n/i_c$  is dependent on the nature of the neutralizer gas. It has been suggested in the literature that alkali atoms may offer enhancement of the

high- $n$  excited-state population, and Futch and Damm<sup>8</sup> have indeed found such enhancement by the use of lithium as compared to water vapor. However, due to the large values of  $\sigma_n^i$  for both lithium and water vapor, it appears to us that neither of these gases will form an optimum neutralizer. Our results indicate instead that the inert gases helium, neon, and argon may well provide the best possible neutralizing media.

## 2. EQUILIBRIUM FRACTIONS OF HIGHLY EXCITED STATES

In this section we discuss the equilibrium fractions of highly excited states in a beam emerging from a gas of ground-state hydrogen atoms. We will for the moment ignore spontaneous radiative transitions, as we shall see in the next section that, except for very low densities of the neutralizer gas, they do not play a role in determining the equilibrium beam.

Let the equilibrium flux of protons be  $i$  and let  $\rho$  be the number density of neutral atoms in the exchange gas. Then, as a function of distance of travel  $x$ , the flux of excited atoms with principal quantum number  $n$  is given by the equation

$$\frac{di_n}{dx} = i\rho\sigma_n^e - i_n\rho\sigma_n^i + \rho \sum_{n' \neq n} \{i_{n'}\sigma(n' \rightarrow n) - i_n\sigma(n \rightarrow n')\}. \quad (3)$$

Here,  $\sigma_n^i$  is the ionization cross section of states with principal quantum number  $n$ , while  $\sigma(n' \rightarrow n)$  and  $\sigma(n \rightarrow n')$  are the inelastic scattering cross sections whereby the principal quantum numbers of the highly excited neutrals are changed by scattering.

It can readily be shown that the final term on the right-hand side in Eq. (3) depends on  $n$  to a higher inverse power than the other two. Greatest contribution to the sum comes from  $n'$  close to  $n$  and cancellation of the two terms  $i_{n'}\sigma(n' \rightarrow n)$  and  $i_n\sigma(n \rightarrow n')$  ensures that their difference is smaller than each of the first two terms by a factor involving inverse powers of  $n$ .

Thus, in equilibrium we have

$$i_n/i_c = \sigma_n^e/\sigma_n^i. \quad (4)$$

This ratio, however, does not give us an absolute assessment of the number of high-energy neutrals in the emerging beam as  $i$  itself is not automatically known and is energy-dependent. Let  $i_c$  be the flux of protons incident on the neutral gas. Then

$$i = i_c - \sum_n i_n,$$

and hence

$$i_n/i_c = \frac{\sigma_n^e/\sigma_n^i}{1 + \sum_n (\sigma_n^e/\sigma_n^i)}. \quad (5)$$

<sup>8</sup> A. H. Futch and C. C. Damm, Nucl. Fusion 3, 124 (1963).

<sup>6</sup> H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, New York, 1952).

<sup>7</sup> A. C. Riviere and D. R. Sweetman, in *Proceedings of the Sixth International Conference of Ionization Phenomena in Gases* (Published by the conference, Paris, 1963), Vol. 1b.

An alternative way of writing this equation is also

$$i_n/i_c = (\sigma_n^e/\sigma_n^i) \left\{ 1 - \frac{1}{i_c} \sum_n i_n \right\}. \quad (5')$$

Equation (5') would be the more favorable expression as the efficiency fraction  $(1/i_c)\sum_n i_n$  is one which is often directly measured in experimental observations; if measurements of this fraction were available as a function of energy, our ratio  $i_n/i_c$  would once and for all be expressed simply in terms of the cross sections  $\sigma_n^e$  and  $\sigma_n^i$ .

Such measurements are, however, apparently not available in the literature, and we therefore use Eq. (5). Here we note that in the summation in the denominator the ground-state term ( $n=1$ ) predominates; the exchange cross section  $\sigma_n^e$  is a maximum for  $n=1$  and the ionization cross section  $\sigma_n^i$  is a minimum. Thus we may simplify Eq. (5) to

$$i_n/i_c = (\sigma_n^e/\sigma_n^i) \frac{1}{1 + \sigma_1^e/\sigma_1^i}. \quad (6)$$

In this expression we may use the corrected Born results of May<sup>5</sup> for  $\sigma_1^e$  which are in close agreement with the experimental results of Fite *et al.*,<sup>9</sup> in the range of observation up to 40 keV, and which become even more accurate at higher energies. For  $\sigma_1^i$  we employ the computed cross sections of Bates & Griffing.<sup>10</sup>

For a complete evaluation of  $i_n/i_c$  we are thus left with a determination of  $\sigma_n^i$  for large  $n$ . Any direct calculation of this cross section leads immediately to a very simple result. The momentum space wave function of the electron in a moving high- $n$  hydrogen atom is equivalent to a sharp wave packet whose speed is that of the moving atom, and for which the spread in electron momenta is  $\lesssim \hbar/na_0$ .

Any scattering process in which the electron momentum is changed by an amount  $\gg \hbar/na_0$  will automatically free it from its high  $n$  state; yet, in the total electron scattering cross section momentum recoils,  $\lesssim \hbar/na_0$  can readily be seen to make a small contribution of order  $1/n^2$ .

Thus,  $\sigma_n^i$  can automatically be identified, with great precision, with the total scattering cross section for an electron moving at the same speed as the high- $n$  state. Such data is available, for example, in Ref. 6.

It is of interest that the cross section  $\sigma_n^i$  is thus, for large  $n$ , independent of  $n$ ; at the energies of interest to us, it is a few times the geometric cross section of the hydrogen atom ground state. This means that as a very highly excited hydrogen atom moves through a gas of ground-state atoms a large number of these will pass through the orbital of the high- $n$  state before it is

TABLE I. The ratio  $i_n/i_c$  as a function of energy for protons passing through atomic hydrogen.

Energy (keV)	$i_n/i_c$ in units of $1/n^3$	
	$\sigma_n^e$ from Eq. 1	Lower limit to $\sigma_n^e$
10	0.20	0.05
20	0.75	0.32
30	0.58	0.40
40	0.34	0.29
50	0.22	0.20
60	0.17	0.17
70	0.14	0.14
100	0.07	0.07

finally disrupted. In other words, one may consider the high- $n$  state to exist even though the electron "orbit" may embrace a large number of neutral 1s atoms in addition to the central proton.

Use of Eq. (1) for  $\sigma_n^e$  and electron scattering data for  $\sigma_n^i$  yields the values for  $i_n/i_c$  given in the first column of the right-hand side of Table I.

It is seen that there is a maximum in  $i_n/i_c$  of approximately  $0.75/n^3$  in the vicinity of 20 keV.

As mentioned in Sec. 1, we believe Eq. (1) to be reasonably accurate in view of the increased accuracy of the first-order perturbation method for high- $n$  states. In order to obtain *lower limits* to the values of Table I, we may, however, adopt the following procedure: Write

$$\sigma_n^e = \sigma_1^e (\sigma_n^e/\sigma_1^e), \quad (7)$$

and for the ratio  $\sigma_n^e/\sigma_1^e$  use the expression

$$\frac{\sigma_n^e}{\sigma_1^e} = \frac{1}{n^3} \left\{ \frac{p^2(p^2+4)}{(p^2+1)^2} \right\}^5. \quad (8)$$

Equation (8) is the ratio for  $\sigma_n^e/\sigma_1^e$  obtained by using first Born approximation for both  $\sigma_n^e$  and  $\sigma_1^e$ . We know that the Born approximation overestimates  $\sigma_1^e$  by about a factor of more than 2 at  $p=1$ , and more seriously for  $p<1$ , yet should be more accurate for  $\sigma_n^e$ . Thus, when the corrected Born results are used for  $\sigma_1^e$  in Eq. (7), and Eq. (8) is employed for the ratio  $(\sigma_n^e/\sigma_1^e)$ , the result should yield a lower limit to  $\sigma_n^e$ , and thus to  $i_n/i_c$ .

This lower limit to  $i_n/i_c$  is given in the second column of Table I.

It should be remarked that the value of the factor  $(1 + \sigma_1^e/\sigma_1^i)^{-1}$ —see Eq. (6)—is approximately  $1/30$  at 10 keV and  $1/10$  at 20 keV. At these energies, therefore, the total efficiency for conversion to neutrals is high, although, of course, most of the neutral population is in the ground state. The above factor gradually decreases with increasing energy so that at high energies, a larger fraction of all neutrals goes into high- $n$  states, although the over-all efficiency for neutral production has dropped.

One may wonder whether we have somewhat underestimated the factor  $(1 + \sigma_1^e/\sigma_1^i)^{-1}$ , and thus under-

<sup>9</sup> W. L. Fite, R. Stebbings, D. Hummer, and R. Brackmann, *Phys. Rev.* **119**, 663 (1960).

<sup>10</sup> D. R. Bates and G. Griffing, *Proc. Phys. Soc. (London)* **66**, 961 (1953).

estimated  $i_n/i_c$  at its maximum near 20 keV, by the use of the values calculated in Ref. 10 for  $\sigma_1^i$ . The calculations of Bates and Griffing pertain to the ionization of one ground-state hydrogen atom due to collision with a second atom without excitation of this second atom. One may expect the total ionization cross section  $\sigma_1^i$  to be somewhat larger due to added contributions from excitation, and thus that our values for  $i_n/i_c$  are systematically low. This may actually be true, although there is evidence that the effect must be small. The total ionization cross section of hydrogen atoms against hydrogen molecules, as a function of energy, has been measured by numerous workers.<sup>11</sup> One may expect this cross section to be roughly twice that for ionization against  $H$  atoms. If we use the results for  $H_2$  molecules of Ref. 11, divided by 2, we obtain values of  $i_n/i_c$  almost identical to those given in Table I.

Finally, it is interesting to enquire as to the conditions under which equilibrium is established, as the equilibrium beam will maximize the flux of highly excited states. If we wish to ensure that equilibrium is achieved for all states up to a certain principal quantum number  $N$ , the path length  $\lambda$  required is given by the condition  $\sigma_N^e \rho \lambda \gg 1$ , as the cross section  $\sigma_N^e$  is the smallest one involved in establishing equilibrium. If we substitute for  $\sigma_N^e$  from Eq. (1), we find

$$\lambda \gg N^3 p^2 \beta^5 (2 \times 10^{14} / \rho) \text{ cm}, \quad (9)$$

with  $\rho$  in units of  $\text{cm}^{-3}$ . Hence, the density  $\rho$  must be chosen sufficiently large to ensure that the equilibrium length  $\lambda$  is satisfactorily small.

### 3. SPONTANEOUS TRANSITIONS

Once the highly excited atoms emerge from the charge-exchange target region so that they no longer undergo collisions, their lifetime in the absence of external fields is determined by their lifetime for spontaneous decay.

These lifetimes have been numerically calculated (for all  $l$ ) for<sup>12</sup>  $n \leq 25$ : a simpler and more general result for  $n \gg 1$  gives<sup>13</sup>

$$\begin{aligned} \tau(n,0) &= (0.19)n^3 \times 10^{-8} \text{ sec}, \\ \tau(n,1) &= (0.018)n^3 \times 10^{-8} \text{ sec}, \\ \tau(n,2) &= (0.054)n^3 \times 10^{-8} \text{ sec}, \\ \tau(n,3) &= (0.12)n^3 \times 10^{-8} \text{ sec}. \end{aligned} \quad (10)$$

The qualitative features of these results are easily explained by two observations<sup>14</sup>: (a) transitions with  $l \rightarrow l-1$  are significantly more probable than those with  $l \rightarrow l+1$ ; (b) the larger the energy difference

TABLE II. The fractional contributions of different orbital angular-momentum states among the highly excited atomic hydrogen states formed by exchange.

Incident proton energy (keV)	Fractional contribution to $\sigma_n^e$ from $l$ th state: $K_l$			
	$l=0$	1	2	3
25	0.29	0.54	0.15	0.02
50	0.34	0.52	0.13	0.01
100	0.46	0.45	0.08	0.01
200	0.62	0.34	0.04	...
500	0.81	0.18	0.01	...

between initial and final states, the more probable the transition. The initial  $p$  state is the only one with direct access to the ground state, and is thus the shortest lived; the lifetimes for the  $d$  and  $f$  states increase regularly for the reason (b); the  $s$  state lives significantly longer than the  $d$  state (although they have a common largest energy transition) because of reason (a).

For a beam of  $H$  atoms produced by charge exchange, the distribution among the various  $l$  states for a given  $n \gg 1$  has been determined in our earlier work.<sup>2-4</sup> The distribution is independent of  $n$  and depends only on the incident proton energy; the fractional contribution of the  $l$ th state to the total cross section for  $n \gg 1$  is called  $K_l$  and is displayed for various energies in Table II.

It is to be noticed that for large incident proton energies ( $\gg 25$  keV) the excited states are essentially all  $s$  states; near the resonance for formation of excited states ( $\sim 25$  keV) more than half the states are  $p$  states and the shorter lifetime  $\tau(n,1)$  is relevant. Almost all (70%) of this decay process is directly into the ground state: All the decay processes are dominated by transitions into the lowest available states.

The characteristic lifetimes (10) may be converted into characteristic decay lengths for the highly excited atoms in the beam by multiplying by the velocity of the beam which, by the definition of  $p$ , is  $v = pe^2/\hbar$ :

$$\begin{aligned} \lambda(n,0) &= (0.42)n^3 p \text{ cm}, \\ \lambda(n,1) &= (0.040)n^3 p \text{ cm}, \\ \lambda(n,2) &= (0.12)n^3 p \text{ cm}, \\ \lambda(n,3) &= (0.26)n^3 p \text{ cm}. \end{aligned} \quad (11)$$

Thus, for  $n=10$  and at the resonance (proton energy = 25 keV,  $p=1$ ), we may say that 54% of the highly excited atoms decay after 40 cm, a further 17% between 120 and 260 cm, and the remaining 29% after 420 cm. These dimensions, particularly the 40 cm one, could well be of the same order as the dimensions of the experimental apparatus.

To follow the spontaneous decay more closely, we observe that the distribution in time for the decay is Poissonian, so that the number of atoms surviving after a time interval  $t$  is

$$n(t) = n(0)e^{-t/\tau}. \quad (12)$$

<sup>11</sup> S. K. Allison, Rev. Mod. Phys. 30, 1137 (1958).

<sup>12</sup> J. R. Hiskes and C. B. Tarter, University of California Radiation Laboratory Report No. UCRL-7088 Rev. I, 1964 (unpublished).

<sup>13</sup> R. M. May, Nucl. Fusion 4, 111 (1964).

<sup>14</sup> H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer-Verlag, Berlin, 1957), p. 268.

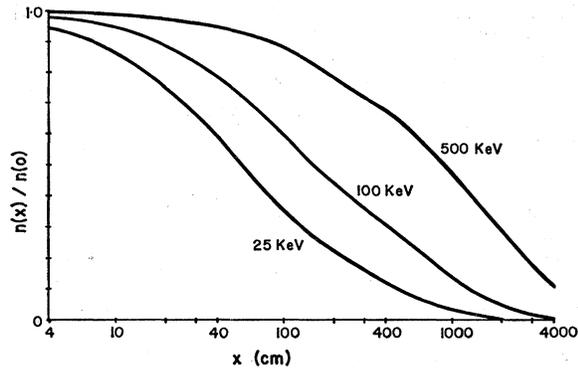


FIG. 1. Attenuation by spontaneous transitions for the beam of excited H atoms in the state  $n=10$ , for incident proton energies of 25, 100, and 500 keV. This distance scale is logarithmic.

This relation can be translated into a survival law for highly excited atoms formed by charge-exchange: The fraction of the beam surviving a distance  $x$  after leaving the target is

$$\frac{n(x)}{n(0)} = \sum_{l=0}^{n-1} K_l \exp\{-x/\lambda(n,l)\}. \quad (13)$$

The characteristic distances  $\lambda(n,l)$  are given by (11) and the  $K_l$  factors by Table II.

The fraction of the beam surviving after a distance  $x$  is displayed in Fig. 1 for  $n=10$  and for several values of the incident proton energy. To obtain the fraction for any other value of  $n$ , one simply multiplies the distance scale by  $(n/10)^3$ .

A comparison between Eqs. (9) and (11) yields the condition on the density for equilibrium to be established well within the spontaneous decay length for the  $p$  states. We need

$$\rho \gg (5 \times 10^{15}) p \beta^5 \text{ cm}^{-3}, \quad (14)$$

where, by "much greater than," we mean at least two orders of magnitude.

#### 4. NEUTRALIZER GASES OTHER THAN ATOMIC HYDROGEN

The question as to whether the population of highly excited states can be enhanced by the use of other neutralizer gases is of considerable interest. In this section, we investigate the effects obtained with two opposite extremes for the neutralizer gas, alkali atoms and inert gases.

(a) Alkali gases. It has been suggested<sup>15</sup> that enhancement of the highly excited state population may be produced by the use of an alkali gas as neutralizer, on the grounds that the outer electron is similar to a hydrogen atom electron in an excited state. On the basis of this suggestion Futch and Damm<sup>8</sup> have

<sup>15</sup> J. R. Hiskes and M. H. Mittleman, U. S. Atomic Energy Commission Report UCRL-9969, 1962 (unpublished), p. 128.

explored the use of lithium vapor and have reported an enhanced excited-state population as compared to the use of water vapor.

We therefore consider both lithium and sodium vapor from a theoretical point of view in order to explore the suggestion of Hiskes and Mittleman further. It is true that for a lithium and a sodium atom, the outer electron has a wave function which is well represented by a hydrogen-like wave function for an effective central charge.<sup>16</sup>

Thus the high  $n$  exchange cross-section  $\sigma_n^e$  for lithium and sodium may be estimated by employing the general formula of May<sup>17</sup> for charge-exchange from hydrogen-like excited states. For electronic pickup from a  $2s$  state, for example, with effective charge  $z$ , we have the result

$$\sigma_n^e = \frac{\pi a_0^2}{z^5 n^3 p^2} \frac{32\gamma^5}{5} \left( 1 - \frac{5\gamma}{6} + \frac{5\gamma^2}{28} \right), \quad (15)$$

where

$$\gamma = 4(p/z)^2 \left\{ (p/z)^2 + \frac{1}{4} \right\}^{-2}.$$

This is therefore appropriate for lithium, for which  $z^2=1.6$ .

A similar, but slightly more lengthy, formula is applied for pickup from a  $3s$  state<sup>17</sup> such as in the case of sodium, for which  $z^2=3.4$ .

The ionization cross section  $\sigma_n^i$  is known from electron scattering data<sup>18</sup> for both lithium and sodium. Thus, we may readily plot the ratio  $i_n/i = \sigma_n^e/\sigma_n^i$  as a function of energy for both cases. The conversion of this ratio to  $i_n/i_c$  offers considerable difficulties at present; we could perform a "corrected Born" calculation for  $\sigma_1^e$ , but any calculation of  $\sigma_1^i$  along the lines of Ref. 10 would require tedious numerical computation and would be of dubious accuracy.

We choose therefore to compare the ratios  $i_n/i$  for lithium and sodium with the corresponding values for hydrogen atoms. From Eq. (15), and the data of Ref. 18, we obtain the values for  $i_n/i$  given in Table III.

It is to be observed that the maxima in  $i_n/i$  for both lithium and sodium occur in the vicinity of 10 keV. This is due to the fact that the exchange cross section—Eq. (15)—has its maxima in these cases in the vicinity of 10 keV; the maximum occurs, of course, when the speed of the proton is equal to the average speed of the electron in its initial atomic state, and this speed is lower in the cases of lithium and sodium than for hydrogen.

It is true that the exchange cross section  $\sigma_n^e$  at maximum is vastly greater for lithium and sodium than for hydrogen; in units of  $\pi a_0^2/n^3$ , the maximum for

<sup>16</sup> John C. Slater, *Quantum Theory of Atomic Structure* (McGraw-Hill Book Company, Inc., New York, 1960), Vol. 1.

<sup>17</sup> R. M. May, University of Sydney, School of Physics, Report TR. 10 (unpublished); R. M. May and J. G. Lodge, Phys. Rev. (to be published).

<sup>18</sup> J. Perel, P. Englander, and B. Bederson, Phys. Rev. **128**, 1148 (1962).

lithium is 2960, and for sodium, 4200 as compared with hydrogen for which the value of  $\sigma_n^e$  at maximum is 62. However, of equal importance in determining the equilibrium ratio  $i_n/i$  is the ionization cross section  $\sigma_n^i$ , which is very much larger for lithium and sodium than for hydrogen.

It is to be observed that the maximum value of  $i_n/i$  for lithium and sodium is 15 and 21, respectively, in units of  $1/n^3$ , while the corresponding maximum for hydrogen, occurring at 20 keV, is 11.

It is significant that these maximum values of  $i_n/i$  for lithium and sodium are very little greater than for hydrogen, and that they occur at a lower energy. In the case of hydrogen, the passage from  $i_n/i$  to  $i_n/i_c$  at the 20-keV maximum has a reduction factor of about  $\frac{1}{10}$ . This factor decreases quite rapidly with decreasing energy, and is approximately 1/30 at 10 keV. This is due to two reasons:

(i) The value of  $\sigma_1^e$  for hydrogen at 20 keV is a factor of 4 less than its maximum value at zero energy. (The value at 20 keV is  $9\pi a_0^2$  increasing to about  $40\pi a_0^2$  at zero energy.)

(ii) The ionization cross section  $\sigma_1^i$  has a maximum near 25 keV, and decreases with decreasing energy.

If the lithium and sodium values for  $i/i_c$  were the same as hydrogen at 10 keV, i.e., 1/30, the maxima in  $i_n/i_c$  would be very appreciably less than the hydrogen maximum. Actually, it is even likely that the appropriate factor is less than 1/30, as the cross section  $\sigma_1^e$  for lithium and sodium itself has a maximum near 10 keV. It is here that the relative speed between proton and electron is equal to the average speed of the captured electron in its hydrogen ground state.

We thus consider it quite likely that the 10-keV maxima in  $i_n/i$  for lithium and sodium will be reduced by a factor greater than 30 in the passage to  $i_n/i_c$ . It should be recognized also that the very large cross sections yielded by Eq. (15), for the lithium and sodium cases tend to indicate that the Born approximation in these cases may be yielding a more serious overestimate of the cross sections than in the case of hydrogen.

TABLE III. A comparison of  $i_n/i$  for the different neutralizing media H, Li, and Na as a function of incident proton energy.

Energy (keV)	$i_n/i$ in units of $1/n^3$		
	H	Li	Na
4	0.5	2.9	4.1
7	2.2	9.7	17
10	4.6	15	21
13	7.0	11	11
16	8.7	6.8	4.3
20	11	4.1	2.3
25	9.0	1.2	1.2
30	6.9	0.6	0.7
40	3.3	...	...
50	1.8	...	...

TABLE IV. A comparison of the quantity  $(\sigma_1^e/\sigma_n^e) i_n/i_c$  for the different exchange media H, He, Ne, and A.

Energy (keV)	$(\sigma_1^e/\sigma_n^e) i_n/i_c$			
	H	He	Ne	A
10	0.05	0.12	0.30	0.05
20	0.10	0.21	0.32	0.10
30	0.08	0.25	0.29	0.12
40	0.06	0.27	0.25	0.15
50	0.05	0.27	0.22	0.16
60	0.04	0.27	0.20	0.14
70	0.03	0.23	0.19	0.16
80	0.03	0.21	0.17	0.16
90	0.02	0.18	0.15	0.14
100	0.02	0.15	0.14	0.11
150	0.007	0.07	0.09	0.06

It would appear therefore that both lithium and sodium are probably significantly less effective as neutralizer media than atomic hydrogen. It is very unlikely that they are any better, and there would seem no strong reason at all why these substances, with their associated experimental difficulties, should be employed.

It should be mentioned that water vapor would also appear to be a rather poor neutralizer. The water-vapor molecule has two donor electrons which can most easily be picked up, but its three-atom molecule presents a large cross section for electron scattering, thus yielding large values of  $\sigma_n^i$ . The fact that lithium vapor yields an enhancement over water vapor is therefore not inconsistent with our conclusions above.

(b) Inert gases. In view of the results pertaining to alkali gases, it seems natural that we should investigate gases which represent the other extreme. Instead of choosing substances for which  $\sigma_n^e$  is large, we might look at gases for which  $\sigma_n^i$  is small. These are, of course, the inert gases.

Naturally, we expect the cross sections for electron exchange to be very much smaller for inert gases than for the alkalis, but in the equilibrium beam it is the ratio  $\sigma_n^e/\sigma_n^i$  which is of fundamental importance. We therefore attempt a comparison between helium, neon, and argon with the hydrogen results.

In the case of the inert gases, the cross sections  $\sigma_1^e$  and  $\sigma_1^i$  are available from direct measurement—Ref. 11—as are also the values of  $\sigma_n^i$  from electron scattering data—Ref. 6. Thus, a reliable comparison between the inert gases and hydrogen, based entirely on experimental results, may be made in terms of the quantity

$$\left(\frac{\sigma_1^e}{\sigma_n^e}\right) \frac{i_n}{i_c} = \left(\frac{\sigma_1^e}{\sigma_n^i}\right) \frac{1}{1 + \sum_n (\sigma_n^e/\sigma_n^i)}. \quad (16)$$

This comparison is shown in Table IV.

The values in Table IV should form a quite significant comparison basis between the inert gases and hydrogen in that the increased difficulty of picking up an electron from the tightly bound outer shell is already represented in the cross section  $\sigma_1^e$ .

TABLE V. Estimates of  $i_n/i_c$  for He, Ne, and A, as neutralizer media, respectively.

Energy (keV)	$i_n/i_c$ in units of $1/n^3$		
	He	Ne	A
10	0.03	0.04	0.04
20	0.10	0.18	0.15
30	0.22	0.34	0.29
40	0.34	0.41	0.44
50	0.43	0.45	0.53
60	0.50	0.44	0.48
70	0.44	0.42	0.54
80	0.41	0.40	0.51
90	0.37	0.37	0.42
100	0.31	0.33	0.34
150	0.13	0.21	0.15

In the case of hydrogen the ratio  $\sigma_n^e/\sigma_1^e$ , when Born approximation is used for both cross sections, is given by Eq. (8), which shows this ratio to have a maximum value at  $p^2=2$  of  $4.2/n^3$ . It is the use of this formula when applied to the hydrogen figures of Table IV which gives us our "lower limit" column for  $i_n/i_c$  in Table I.

It is clear from Table IV that, provided the ratio  $\sigma_n^e/\sigma_1^e$  is of comparable magnitude for the inert gases as for hydrogen, these gases could well show some enhancement in excited-state production over hydrogen.

To obtain a rough idea of the final results for the inert gases, we may derive a Born approximation ratio similar to Eq. (8) using an independent particle picture for the electron closed shells with an effective charge and using the results of Ref. 17. This procedure yields the results shown in Table V.

It is to be stressed that the figures of Table V can only be considered as rough estimates; in the case of closed-shell atoms, the use of hydrogen-like wave functions derived from an effective charge is a questionable procedure. Yet, in view of the fact that we have used this procedure only for the ratio  $\sigma_n^e/\sigma_1^e$ , the results must be considered as providing lower limits, in the same sense as our figures for lower limits for hydrogen in Sec. 2. On this basis we expect that the maxima for the inert gases, occurring in the region 50–70 keV, are almost certainly somewhat in excess of the 20-keV maximum for hydrogen.

The actual values of the exchange cross sections for the inert gases are roughly an order of magnitude smaller in the 50–70-keV region than in the case of

hydrogen at 20 keV. Thus, the density must be chosen roughly an order of magnitude larger than condition (15) for hydrogen. That the maximum production for the inert gases occurs in the 50–70-keV region is of interest in that the characteristic decay lengths—Eq. (11)—for the highly excited hydrogen atoms will be more than a factor of 2 greater than at the 20-keV maximum for hydrogen.

#### SUMMARY

We have considered the production of highly excited states of atomic hydrogen by charge-exchange reactions, discussing first the reaction  $H^+ + H(1s) \rightarrow H(\text{excited}) + H^+$ ; we have then used the results for this reaction as a basis with which to compare the excited state production for neutralizing gases other than atomic hydrogen. We have expressed our results in terms of the equilibrium ratio  $i_n/i_c$ , that is, the ratio of the flux of excited states with principal quantum number  $n$  to the (constant) flux of protons incident on the neutralizer.

We find that with atomic hydrogen as neutralizer, the ratio  $i_n/i_c$  has a maximum of about  $0.75/n^3$  at 20 keV. For alkali atoms such as lithium and sodium, the maximum value of  $i_n/i_c$  occurs at about 10 keV being of similar magnitude to, but probably smaller than, the value for atomic hydrogen.

In the case of the inert gases helium, neon, and argon, the maximum value of  $i_n/i_c$  occurs in the vicinity of 50–70 keV, and again is of similar magnitude to, but probably slightly larger than, the maximum value for atomic hydrogen.

The inert gases thus offer a number of advantages over alkali gases as neutralizers: They probably yield as large, if not larger, values of  $i_n/i_c$  at maximum; the maximum occurs at higher energies so that the natural decay lengths of the excited atoms are correspondingly longer; they may be very much more easily handled experimentally.

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