Production of Metastable Hydrogen Atoms in Proton–Rare-Gas Collisions*

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Cross sections for electron capture into the metastable hydrogen $2s$ level for proton–rare-gas collisions have been measured. The $2s$ state population is determined by application of an electric field in the reaction region leading to a mixing of the $2s_{1/2}$ and the $2p_{1/2}$ atomic substates. The increase in the detected Lyman-alpha signal over that observed in the field-free case is related to the ratio of the cross sections for capture into the $2s$ and the $2p$ states. The cross sections, measured for proton energies between 1.5 and 23 keV, exhibit structure similar to that observed in $2p$-state capture. Possible interpretations of the results are discussed.

INTORDUCTION

RECENTLY, Pretzer, Van Zyl, and Geballe[1,2] have reported measurements of the cross sections for Lyman-alpha emission resulting from proton- and deuteron–rare-gas collisions. In these measurements, a variable energy (1.5 to 23 keV) ion beam was allowed to traverse a pressure-monitored target gas cell. The Lyman-alpha radiation resulting from charge capture by protons was detected with an oxygen-filtered, iodine-filled Geiger counter similar to that designed by Brackmann, Fite, and Hagen.[3]

These earlier investigations show that definite structure exists in the cross sections for $2p$ state capture. These cross sections are included in Figs. 4 through 7 of this paper. The interesting results obtained in these measurements led us to a study of the $2s$ capture cross sections in order to gain further information about the processes responsible for the double-peak structure present in the $2p$ cross-section curves.

The natural lifetime of the $2s_{1/2}$ hydrogen state is about 0.14 sec.[4] However, when a hydrogen atom in its metastable state is in an external electric field, its lifetime becomes a function of the applied field strength because the $2s_{1/2}$ state is mixed with the $2p_{1/2}$ state, which can decay to the ground state with emission of Lyman-alpha radiation. The time required for the $2s_{1/2} \rightarrow 2p_{1/2} \rightarrow 1s_{1/2}$ decay sequence varies from about 20 years in the zero field case to twice the $2p$ state lifetime (3×10^{-9} sec) for large fields. Several calculations giving the lifetime of the $2s_{1/2}$ state as a function of the applied field have been made.[5–7] The results of Lüder's calculations,[7] with and without the Lamb shift corrections, are shown in Fig. 1. An experimental determination of the lifetime versus applied field strength between 50 and 500 V/cm has been made by Sellin.[8] His results are included in Fig. 1.

TECHNIQUE

Several experimental techniques are available for measuring the intensity of a beam of metastable atoms. Lamb and Retherford[6] and Lichten and Schultz[9] have observed the secondary electron emission resulting from $2s$ atoms impinging on metal surfaces. Owing to the high, variable kinetic energy of the metastable atoms to be detected in the present experiment, this method was considered less desirable than an alternate method of "electric field quenching" of the $2s$ level. This second

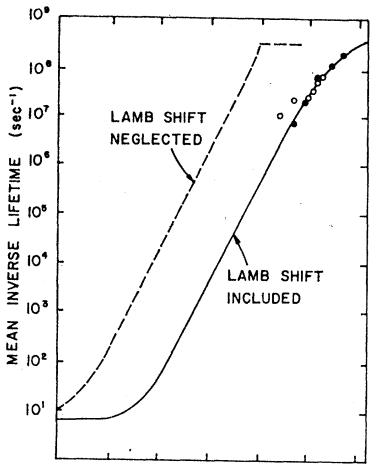

FIG. 1. Mean lifetime of H($2s$) state as a function of electric field strength calculated by Lüders (Ref. 7). Closed circle data are experimental values of Sellin (Ref. 8). Open circle data are from present experiment using Eq. (1) and saturation curve (Fig. 3.)

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[1] D. Pretzer, B. Van Zyl, and R. Geballe, Phys. Rev. Letters **10**, 340 (1963).

[2] D. Pretzer, B. Van Zyl, and R. Geballe, *Proceedings of the Third International Conference on the Physics of Electronic and Atomic Collisions* (North-Holland Publishing Company, Amsterdam, 1964), p. 618.

[3] R. F. Brackmann, W. L. Fite, and K. E. Hagen, Rev. Sci. Instr. **29**, 125 (1958).

[4] J. Shapiro and G. Breit, Phys. Rev. **113**, 179 (1959).

[5] H. A. Bethe and E. E. Salpeter, *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1957), Vol. XXXV, p. 370.

[6] W. E. Lamb and R. C. Retherford, Phys. Rev. **79**, 549 (1950).

[7] G. Lüders, Z. Naturforsch. **5a**, 608 (1950).

[8] I. A. Sellin (unpublished).

[9] W. Lichten and S. Schultz, Phys. Rev. **116**, 1132 (1959).

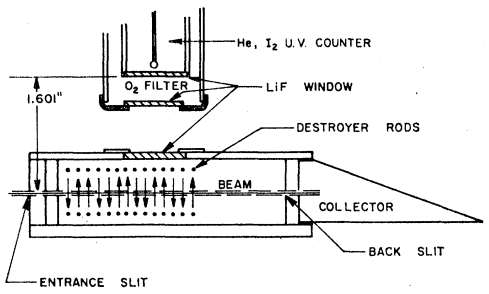

FIG. 2. Collision cell and detector geometry.

method has been employed by several investigators.[8,10–12] In these experiments, the technique employed was to form $2s$ atoms in a collision region, allow them to drift into a field quenching region and detect the resultant Lyman-alpha radiation. This procedure presents difficulties since the number of metastable atoms entering the quenching region represents an unknown fraction of those produced in the interaction chamber. Scattering at the time of capture and the large collision deexcitation cross section of $H(2s)$ place stringent requirements on the vacuum and mechanical design of the apparatus.

In order to overcome these difficulties, the perturbing electric field in the present experiment was applied within the collision region. The design of the collision chamber employed for this purpose is shown in Fig. 2. The proton beam enters the reaction chamber through the entrance apertures, traverses the collision cell and leaves the chamber through the exit slit in the chamber wall. Beam depletion in the collision chamber is negligible because of the low target gas pressures employed. As the beam passes through the reaction region, it encounters a spatially varying, transverse electric field of variable magnitude. This field design was chosen to give small net deflection of the proton beam in the collision chamber. Lyman-alpha photons, produced as a result of charge transfer, may escape through the lithium fluoride window at the top of the collision cell into the photon counter. When the electric field is off, only Lyman-alpha emission resulting from decay of the $2p$ state is detected. With the field on, radiation from both the $2s$ and $2p$ states is observed. The increase in signal over the field-free case is related to the cross section for capture into the $2s$ state. The absolute values of the $2s$ cross sections are determined by comparing it with the previously determined $2p$ state capture cross sections.[2]

As the lifetime of the $2s$ state is a function of the magnitude of the applied electric field, the increase in Lyman-alpha signal over the field-free case should also depend on the field strength. A necessary condition imposed on the experiment is that as the applied field is increased, the Lyman-alpha radiation from quenching the $2s$ level must reach a saturation value. This ensures equilibrium between the rates of formation and decay of the $2s$ state in the observation region since the lifetime of the $2s$ state has been made short compared to the transit time through the collision cell. An example of such a saturation curve obtained experimentally is shown in Fig. 3.

It is necessary to show that the detected signal does not arise from slow charged particles (produced by ionization and charge transfer) which are accelerated towards and strike the destroyer rods. As the destroyer rods and the chamber walls are coated with a layer of black gold (to prevent reflection of Lyman-alpha radiation into the counter[13]), they could be covered with surface layers of hydrogen which could lead to Lyman alpha radiation following bombardment by charged particles. To determine the importance of this effect, a beam of He^+ ions was allowed to pass through the collision cell. The resultant signal was small compared to that obtained with protons as projectiles (10% for He targets and less for the others). In addition, part of the observed radiation in the He^+ case comes from the He^+ ion itself as its $n=4$ to $n=2$ transition has the same wavelength as the Lyman-alpha line. The ratio of the signals resulting from He^+ and H^+ bombardment was independent of the applied field. Additional evidence of the small size of this effect may be obtained from the saturation curve of Fig. 3. One would not expect a saturation curve if a significant part of the signal arose from such secondary processes.

A measure of the proton beam divergence in the collision cell was determined by comparing the ion current traveling through the chamber exit slit and into the collector with that striking the back wall of the chamber. The back-wall current was normally less than 4% of the collector current and remained constant regardless of the magnitude of the applied field. This condition was placed on the beam focus in order that the solid angle viewed by the detector did not change with application of the quenching field. Thus the spa-

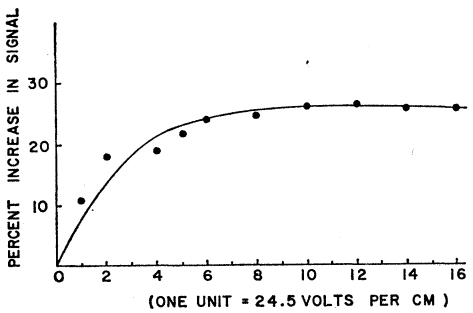

FIG. 3. Signal saturation curve for D^++Ar at 6 keV.

[10] R. F. Stebbings, W. L. Fite, D. G. Hummer, and R. T. Brackmann, Phys. Rev. **119**, 1939 (1960).

[11] L. Colli, F. Cristofori, G. E. Frigerio, and P. G. Sona, Phys. Letters **3**, 62 (1962).

[12] B. L. Donnally, T. Clapp, W. Sawyer, and M. Schultz, Phys. Rev. Letters **12**, 502 (1964).

[13] L. Harris and J. K. Beasley, J. Opt. Soc. Am. **42**, 135 (1952).

tially varying field in the collision chamber did not alter the projectile trajectories to a measurable degree.

The beam currents employed were normally about 0.05 μA and the target gas pressures were approximately 2×10^{-5} Torr. The measured Lyman-alpha signal was found to depend linearly on both beam current and target pressure. The target gas samples of Ne and Xe were "spectroscopically pure" and those of He and Ar were taken from standard gas cylinders. Each was passed through a dry-ice trap before admission into the collision cell.

Krypton was not included as a target gas in the present measurements since earlier studies[2] with a Kr target indicated that one of the Kr emission lines (1165 Å) lies near a transmission window of the oxygen wavelength filter (1167 Å) and that some of this radiation could penetrate the wavelength filter to a noticeable extent. The wavelength discrimination properties of oxygen have been measured by Watanabe[14] and are discussed for this application in Ref. 2.

The high transverse field gradients in the collision cell imposed a lower limit of 2 keV on the projectile energies attainable in the apparatus. The measurements were, however, extended to an effective proton energy of 1.5 keV by employing deuterons as incident ions. Our earlier work on $2p$ state capture indicated that all major features of the cross sections for D^+ rare gas interactions were similar to those for H^+ target encounters when plotted at equivalent projectile velocities.[2]

The emitted radiation in the present experiment is assumed to possess an isotropic spatial distribution. The radiation distribution is determined by the polarization P' which is defined[15] as $P'=(I_{\parallel}-I_{\perp})/(I_{\parallel}+I_{\perp})$. $I_{\parallel}$ and $I_{\perp}$ are the intensities parallel and perpendicular to the beam axis for $\theta=\pi/2$ and are proportional to the dipole interaction terms given, for example, by Condon and Shortley.[16] From these dipole interaction terms, one can show that the $2p_{1/2}-1s_{1/2}$ transition is isotropic. Since the $2s_{1/2}$ level mixes only with the $2p_{1/2}$ level, radiation resulting from depopulation of the metastable level will also be isotropic. In the earlier $2p$ capture cross section measurements (used here as the calibration standard), the radiation was assumed isotropic. The degree of polarization, if any, could not be determined theoretically as the distribution of population in the substates of the $2p$ level was not known. If the $2p$ radiation were found to be polarized and the degree of polarization were to be determined, the present data could be adjusted accordingly.

The estimated uncertainty associated with the present measurements is $\pm50\%$. This large uncertainty is primarily the result of the $\pm45\%$ uncertainty quoted for the $2p$ state capture results.

[14] K. Watanabe, *Advances in Geophysics* (Academic Press Inc., New York, 1958), Vol. 5, p. 153.
[15] J. A. Smit, Physica 2, 104 (1935).
[16] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, London 1963), p. 387.

RESULTS

The results of the cross-section measurements are presented in units of 10^{-16} cm^2. Proton energies are expressed in keV and deuteron energies in keV/2. The total charge-transfer cross sections for the proton–rare-gas systems as reported by Stedeford and Hasted[17] are also presented. The general appearance of the cross-section curves will be discussed here while the discussions of the over-all continuity of the results and their possible interpretation will be deferred until the next section.

H^++He. The results of the measurements for this reacting pair are shown in Fig. 4. The $2p$ capture cross section reported by Pretzer, Van Zyl, and Geballe,[2] the $2s$ capture cross section reported by Colli, Cristofori, Frigerio, and Sona,[11] and the total charge-transfer cross section reported by Stedeford and Hasted[17] are also included in Fig. 4.

Within the energy range investigated, the $2s$ capture cross section is smaller than that for the $2p$ state. At low energies, the ratio of the cross-sections for $2p$ and $2s$ state capture is approximately equal to the ratio of the statistical weights of the levels (3 to 1). At the higher energies, the magnitude of the $2s$ capture cross section becomes a larger percentage of the $2p$ cross section. The shape of the cross-section curves suggest that the $2s$ cross section may exceed the $2p$ cross section and that its maximum will occur at an incident projectile energy larger than that for the maximum of the $2p$ state capture cross section.

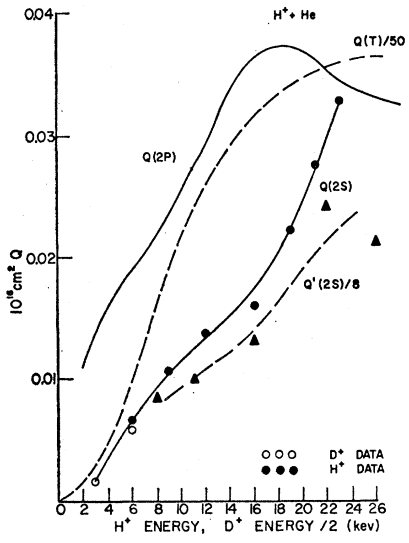

FIG. 4. Cross sections for proton-helium collisions. $Q(T)$ is the total charge-transfer cross section measured by Stedeford and Hasted (Ref. 17). $Q(2P)$ is the $2p$ capture cross section measured by Pretzer, Van Zyl, and Geballe (Ref. 2). $Q^1(2S)$ is the $2s$ capture cross section measured by Colli, Cristofori, Frigerio, and Sona (Ref. 11). $Q(2S)$ are present data.

[17] J. B. H. Stedeford and J. B. Hasted, Proc. Roy. Soc. (London) A227, 466 (1955).

Although the present data and those reported by Colli, Cristofori, Frigerio, and Sona[11] disagree to a considerable extent with regard to the magnitude of the cross section, shapes of the curves are quite similar. The discrepancy in magnitude may be accounted for by the method of normalization employed by Colli *et al.* Their results were normalized to the Born approximation[18] at a proton energy of 40 keV. The Born approximation generally overestimates the cross section at energies near its maximum.

$H^+ + Ne$. The data for this reacting pair are shown in Fig. 5. Again the $2p$ capture cross section[2] and the total charge-transfer cross section[17] are given.

Note that the $2s$ capture cross section exhibits definite structure. As in the $2p$ capture cross section, the $2s$ capture cross section has a pronounced maximum in the energy region where the total charge-transfer process is most efficient. In the 2s curve, however, the low energy maximum is only $\frac{1}{3}$ as large as that of the $2p$. Again this ratio is in agreement with the ratio of statistical weights of the states. The high energy maximum in the $2s$ cross section occurs at a higher projectile energy than in the $2p$ case and is larger in absolute value.

$H^+ + Ar$. The results of investigations on this projectile-target system are shown in Fig. 6. The data are similar to those exhibited by the Ne target except that the high energy $2s$ capture peak is smaller in magnitude than the corresponding $2p$ peak. Again the ratio of the magnitudes in the vicinity of the low energy maximum is nearly in the ratio of statistical weights.

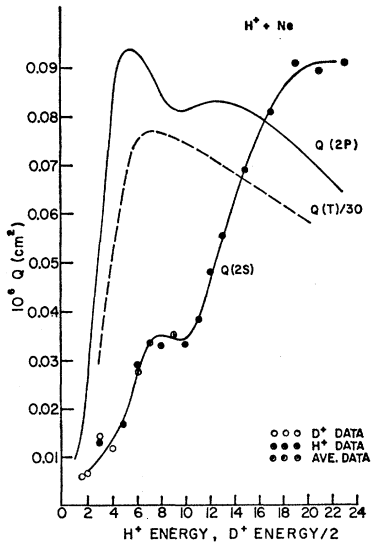

FIG. 5. Cross sections for proton-neon collisions. $Q(T)$ is the total charge-transfer cross section measured by Stedeford and Hasted (Ref. 17). $Q(2P)$ is the $2p$ capture cross section measured by Pretzer, Van Zyl, and Geballe (Ref. 2). $Q(2S)$ are present data. "Ave. Data" are the combined weighted averages of D^+ and H^+ runs.

[18] R. A. Mapleton, Phys. Rev. **122**, 528 (1960).

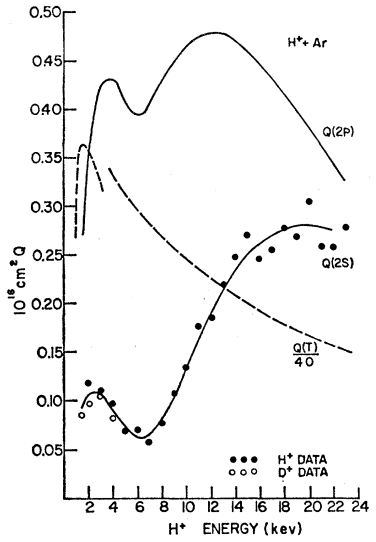

FIG. 6. Cross sections for proton-argon collisions. $Q(T)$ is the total charge-transfer cross section measured by Stedeford and Hasted (Ref. 17). $Q(2P)$ is the $2p$ capture cross section measured by Pretzer, Van Zyl, and Geballe (Ref. 2). $Q(2S)$ are present data.

$H^+ + Xe$. The cross sections for $2s$ and $2p$ state capture for $H^+ + Xe$ collisions are shown in Fig. 7. The $2p$ capture cross section has a low energy maximum at a projectile energy of about 0.4 keV. The $2s$ capture measurements do not extend below 1.5 keV and the existence of a corresponding low energy peak in the $2s$ capture process remains uncertain. The high energy peak in the $2s$ cross section occurs at about 15 keV as compared to the 10 keV maximum for the $2p$ capture results. The magnitude

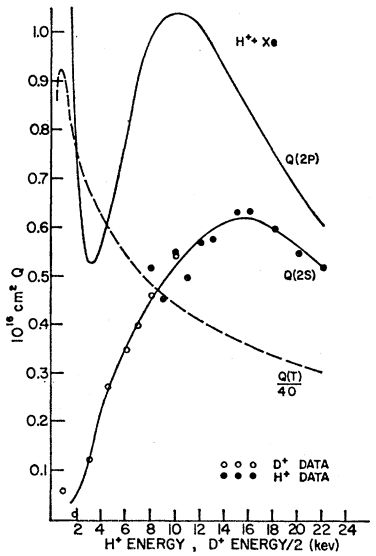

FIG. 7. Cross sections for proton-xenon collisions. $Q(T)$ is the total charge-transfer cross section measured by Stedeford and Hasted (Ref. 17). $Q(2P)$ is the $2p$ capture cross section measured by Pretzer, Van Zyl, and Geballe (Ref. 2). $Q(2S)$ are present data.

of the high energy $2s$ peak is smaller than the corresponding $2p$ capture maximum.

DISCUSSION

In addition to direct capture into the $2s$ and $2p$ states, these states may also be populated via capture into higher states which eventually arrive in the $n=2$ levels via Balmer-emission processes. The relative importance of these processes will now be discussed.

De Heer, van Eck, and Kistemaker[19] have measured the capture cross sections for Lyman-alpha and Lyman-beta radiation in proton collisions with He and Ne. Their results for the Lyman-alpha emission cross sections are about a factor of 3 larger in absolute value than those reported by this laboratory.[2] Of interest here are the relative populations of the $3p$ and $2p$ levels. De Heer *et al.* find that the ratio of Lyman-beta to Lyman-alpha emission is about 1/7. Since a large percentage of the $3p$ level population (88%) decays via Lyman beta,[16] this ratio gives a fair determination of the magnitudes of the capture cross sections into the $2p$ and $3p$ atomic levels and suggest that the capture cross section into the $3p$ level is significantly smaller than that for the $2p$ state. It is probable, therefore, that the entire $n=3$ level population is somewhat smaller than the $n=2$ level population, and that direct capture is the dominant mechanism for population of the $n=2$ levels.

In addition, the experimental apparatus employed in making the measurements reported here discriminates against Lyman-alpha emission resulting from cascade population of the $n=2$ levels by virtue of the long lifetimes of the $n=3$ states. Since the length of the collision chamber is short compared to the product of the beam velocity and the long decay times associated with the upper states, an equilibrium between population and decay of the upper states is not reached in the length of the path provided by the apparatus. An estimate of the ratio of the intensity of radiation from each of the upper states to the equilibrium or saturation value can be obtained from the relation

$$I/I_0=1-\exp(-xP/v), \qquad (1)$$

where x is the distance from the entrance of the collision chamber to the point at which the radiation is detected, P is the total transition probability from the upper state, and v the projectile velocity. However, since the relative population densities of the upper states are not known, the exact amount of cascade to the $n=2$ levels cannot be estimated accurately.

In a previous paper,[20] we reported a test on the relative importance of the cascade mechanism for population of the $2p$ level in collisions of H^+ on Ar. We measured the cross section for Lyman-alpha emission at two values of the variable x of Eq. (1). At the larger value, the cascade population of the $2p$ level should be enhanced due to the additional time available for cascade. The two cross sections were found to be equal within $\pm 5\%$ at all energies, when normalized at 6 keV. We concluded that cascade processes were not responsible for a major portion of the Lyman alpha signal in any particular energy range.

In the present measurements, an electric field is applied to the collision products, which mixes the upper states with the same n and j values. Thus, a larger percentage of the upper levels may now decay directly to the $1s$ state with the emission of shorter wavelength members of the Lyman series (i.e., L_β, L_γ, etc.). The effect of this mixing will be to decrease the total cascade contribution to the $n=2$ levels below that of the field-free case.

Equation (1) can also be employed to estimate the lifetime of the $2s$ state in the presence of an external electric field. The ratio I/I_0 for radiation arising from destruction of the $2s$ state, can be obtained as a function of the applied electric field from a saturation curve such as shown in Fig. 3. Knowing x and v of Eq. (1), one can compute the transition probability P as a function of the applied field. The inverse lifetime of the $2s$ state determined in this manner is shown in Fig. 1.

The values of the lifetime determined by this process are only approximate because the values of the electric field on the beam axis are not known accurately. In addition, Eq. (1) is not exactly valid because the electric field is not uniform over the position variable x. The agreement between our points and the theoretical curve is satisfactory considering the uncertainties of the experimentally determined points.

An interesting feature of the data is that the $2s$ capture cross sections seem to reach their high energy maxima at projectile energies which are larger than the corresponding high energy maxima in the $2p$ capture cross sections. Van Eck, de Heer, and Kistemaker[21] have observed a similar behavior in cross sections for helium atom excitation by protons where, for example, the ^1S states are excited most effectively at proton energies in excess of 35 keV while the ^1D states reach maxima in their excitation functions at about 15 keV. They also find that the maxima in the cross sections for excitation of the $3\,^3D$, $3\,^3P$, and $4\,^3S$ states of the helium atom by hydrogen impact decrease as the angular momentum of the final state increases.[22]

This behavior is consistent with a simple argument based on the adiabatic maximum rule suggested by Massey,[23] which says that the maximum value of a cross

[19] F. J. de Heer, J. van Eck, and J. Kistemaker, *VI^e Conference sur les phenomenes d'ionisation dans les gas* (Paris, 1963), p. 73.

[20] B. Van Zyl, D. Jaecks, D. Pretzer, and R. Geballe (unpublished).

[21] J. van Eck, F. J. de Heer, and J. Kistemaker, Physica **28**, 1184 (1962).

[22] J. van Eck, F. J. de Heer, and J. Kistemaker, *Proceedings of the Third International Conference on the Physics of Electronic and Atomic Collisions* (North-Holland Publishing Company, Amsterdam, 1964), p. 624.

[23] H. S. W. Massey, Rept. Progr. Phys. **12**, 248 (1949).

section occurs at a projectile energy E_m in eV given by

$$E_m = 3m\,a^2(\Delta E)^2. \quad (2)$$

The a in Eq. (2) is the length in angstroms of the region over which a reaction can occur and is inversely related to the impact parameter for the collision, m is the mass of the projectile in amu and ΔE is the internal energy change in eV of the collisional system. Thus, for constant ΔE, Eq. (2) indicates that as the impact parameter for a collision decreases, the energy at which the maximum cross section occurs will increase. Alternatively, the expectation value of the velocity for an electron in the target system increases for decreasing impact parameter.[24] Since charge-transfer processes are usually most efficient when the projectile velocity matches the orbital velocity of a target electron, the energy at which maximum occurs is again inversely related to the impact parameter.

For $2s$ and $2p$ state capture, the values of ΔE involved are expected to be comparable and the above ideas may be applied. Since the p state wave functions are nonspherical and some of these extend along the internuclear axis (the axis of quantization) of the impacting systems, capture into a p state should occur for larger impact parameters than s state capture. From the above arguments, the $2p$ capture process should reach a maximum at lower projectile energies than the $2s$ capture reaction. This feature is found in the experimental results. The relative magnitudes of the $2s$ and $2p$ capture cross sections are also consistent with these ideas. When the peaks occur at low energy, as for example in Xe and Ar, the $2p$ capture is more efficient than the $2s$. At higher energies, however, the $2s$ cross sections seem to be the larger of the two. Although a qualitative description of the results can be fitted to this model, the obvious deficiencies of the model, such as the neglect of coupling during the collision period, make it unsuitable for quantitative analysis.

While the previous discussion has dealt with the high energy peak exhibited by the cross-section curves, nothing has been said about the low energy maximum. The occurrence of the low energy peak and its location at about the same projectile energy as the total charge-transfer maximum lead us to suggest that it is associated with the total charge-transfer reaction. This association could arise either from a process associated with the ground state capture reaction that gives an extraneous signal or from a real coupling between ground and excited states during the period of an interaction that constitutes the mechanism for "capture into an excited state."

There are three principal secondary mechanisms which could give rise to Lyman-alpha radiation in the reaction cell following a charge-transfer collision. These are: (1) collision of the fast hydrogen atom reaction product with another target rare-gas atom. (2) collision of the residual rare-gas ion with the chamber wall, and (3) collision of the fast H atom with the chamber wall.

Mechanism (1) can be ruled out because signals arising from this process would depend on the square of the target pressure. Reaction (2) has been ruled out by auxiliary experiments in which rare-gas ions are deliberately attracted to the chamber wall within view of the counter without an increase in signal. Mechanism (3) cannot be eliminated by simple experimental test but a number of considerations indicate that its contribution to the detected signal should be small. First, the differential scattering cross sections in charge-transfer collisions fall off very rapidly with increasing scattering angles[25] and only a very small percentage of the H atoms should be scattered through an angle large enough to strike the chamber wall within the field of view of the counter. Second, the efficiency of countable ultraviolet production for H_2^+ ions striking hydrogen covered surfaces is only about 5×10^{-3} photons/ion.[26] Fast H atoms could not be appreciably more efficient. Finally, as mentioned earlier, the $2p$ measurements were repeated with a different chamber geometry and the cross sections were found to be identical to those reported earlier. If wall collisions were important, they should depend upon the particular collision cell design.

On the other hand, it is possible that the populations of the $2s$ and $2p$ levels may be influenced by an intermediate state existing during the collision period. The calculations of Lovell and McElroy[27] indicate that the cross section for $2s$ capture in proton-H atom collisions is influenced by coupling to the ground-state capture configuration. They find that this coupling tends to increase the size of the $2s$ capture cross section in the energy where ground-state capture is large. Bates and McCarroll[27] and McDowell[28] have also indicated that intermediate state coupling is of considerable importance in atomic collisions.

If ground-state charge capture does influence excited state capture, one might expect that coupling would be strongest or be most effective in the energy region where the ground-state capture process is most efficient. Note that all the measured $2s$ and $2p$ capture cross sections (excepting Xe for the $2s$ case) show maxima in their cross-section curves at about the same energy as the maxima of the total charge-transfer cross-section results.

If one represents the reaction by a two-step process such as

$$H^+ + Ar \rightarrow H(1s) + Ar^+ \rightarrow H(2p) + Ar^+$$
$$H^+ + Ar \rightarrow H(1s) + Ar^+ \rightarrow H(2s) + Ar^+$$

[24] D. R. Bates and R. McCarroll, Advan. Phys. **11**, 39 (1962).

[25] E. N. Fuls, P. R. Jones, F. P. Ziemba, and E. Everhart, Phys. Rev. **107**, 704 (1957).

[26] G. H. Dunn, R. Geballe, and D. Pretzer, Phys. Rev. **128**, 2200 (1962).

[27] S. E. Lovell and M. B. McElroy, undated, Kitt Peak National Observatory report, received May 1964 (unpublished).

[28] M. R. C. McDowell, National Bureau of Standards Technical Note 185, 1963 (unpublished).

we can see that the first part is the same for both the $2s$ and $2p$ states. The second half is then merely a process involving the Coulomb excitation of H atoms by Ar^+ projectiles. If the ratio between the excitation probabilities for the $2p$ and $2s$ states is the same for ions as it is for electrons (about 5 or 3 to 1 according to Fite and Brackmann,[29] Fite Brackmann, Hummer, and Stebbings[30] and Lichten and Schultz[9]), then the ratio of the low energy $2p$ cross section maxima to the $2s$ should be of this order The experimental results have this property.

Although the above ideas are largely conjectural, they give a qualitative and consistent explanation for the appearance of the structure in the measured cross sections.

[29] W. L. Fite and R. T. Brackmann, Phys. Rev. **112**, 1151 (1958).

[30] W. L. Fite, R. T. Brackmann, D. G. Hummer, and R. F. Stebbings, Phys. Rev. **124**, 2051 (1961).

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Mobilities and Reaction Rates of Ions in Helium*

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The mobility of He^+ ions in helium gas was measured, the result being 10.40 ± 0.10 (cm^2/V-sec). The He^+ ions were observed to react with the helium gas, the attachment frequency being $[1.08\pm0.05]\times10^{-31}$ $n^2(sec^{-1})$, where n is the gas density in atoms/cm^3. This value of the attachment frequency is roughly compatible with the theoretical estimate of Bates and with the mass-spectrometric measurement of Phelps and Brown for the rate of the three-body attachment reaction producing He_2^+. The mobility of the product ion, He_2^+, was found to be 16.70 ± 0.17. This mobility is compatible with recent ambipolar diffusion coefficient measurements on cold afterglows, where the identification of the ion is confirmed by the spectroscopic observations of Kerr. This mobility is inconsistent with the widely quoted value of Biondi and Chanin and with the theoretical analyses of Geltman and Dalgarno. Mobility determinations were also made on another ion, which is possibly what Biondi and Chanin called He_2^+. The actual identity is unknown, the ion possibly being He^{2+}, possibly a contaminant.

I. INTRODUCTION

MOBILITIES of ions in gases have been studied for a long time with many confusing and contradictory conclusions. The mobilities are of widespread and continuing interest for two reasons. As a transport coefficient, the mobility constant is useful in certain applications where drift or diffusion is the principal mechanism for the loss of ionization. Also, the mobility constant provides an experimental measure of the effects of elastic scattering at thermal energies. Secondly, mobility studies provide a means of distinguishing the types of ions that are present in a relatively high-pressure gas. The existence of ionization in a low-temperature gas is an unstable state with the ions being neutralized either by free electrons, ions of the opposite charge, or at the solid surfaces. During the time required for this neutralization to occur the ions can react with the gas, the products in general having a different mobility. Mass spectrometers are capable of identifying the ions positively; however, it is necessary to extract the ions from the gas and accelerate them. Studies using mass spectrometers, even while having interpretation problems of their own, are being used to supplement mobility studies in supplying information on the chemical history as ions approach equilibrium in a gas.

Helium is in many ways the simplest of the gases, and accordingly, it has received a great deal of study in drift tubes, in afterglows, and in mass spectrometers. As new information has come in over the years the apparent understanding has oscillated wildly from poor to good and back again.

Electron impact in helium gas can produce only two kinds of ions, He^+ and He^{2+}. He^{2+} converts to He^+ rather rapidly by charge transfer, and the He^+ attaches to form He_2^+. It is almost certain that He^+ does not have excited states which can survive more than a few gas kinetic collisions. Thus, in pure helium gas, the ionic chemistry is expected to be very simple. The trouble is associated with the fact that small traces of contaminants become strongly preferentially ionized by charge transfer. The subject of this paper is a study of the behavior of a small density of helium ions as they approach thermal equilibrium.

II. DEFINITIONS AND CONVENTIONS

In accordance with traditional usage, ionic mobility is here defined as the ratio of the drift velocity, v(cm/sec),

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