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Formation and Destruction of Excited Hydrogen Atoms at High Impact Velocities*

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A study has been made of the charge-transfer processes whereby fast neutral atoms of hydrogen are formed in the $3s$, $3p$, and $3d$ excited states as a result of the impact of protons on targets of helium and of nitrogen. Impact energies ranged from 75 to 400 keV. The experimental procedure involved quantitative measurement of the Balmer α radiation emitted by the spontaneous decay of atoms in these three states. The three contributions to the emission were separately identified according to the different lifetimes of the $3s$, $3p$, and $3d$ states by means of a time-of-flight technique. A study was also made of the processes whereby the excited atoms were collisionally destroyed before undergoing spontaneous radiative decay. Detailed measurements are presented of the cross sections for capture into the $3s$ state, which are by far the largest of these capture cross sections. Cross sections for capture into the $3p$ and $3d$ states are one to two orders of magnitude smaller, and preliminary measurements are discussed. All of these cross sections decrease rapidly with increasing impact energy. There is general agreement between the present measurements and the predictions of the Born approximation for capture into the $3s$ and $3d$ states from a target of helium, but the calculation for the $3p$ state appears to overestimate the cross section by a factor of at least 4. The cross sections for collisional destruction of atoms in the $3s$ state are several orders of magnitude larger than for electron capture into this state, and the magnitude of the measured values are in agreement with theoretical predictions.

I. INTRODUCTION

The primary objective of the present research has been the measurement of cross sections for the formation of fast excited hydrogen atoms in the $3s$, $3p$, and $3d$ states by the impact of protons on gaseous targets. Impact energies ranged from 75 to 400 keV. The process of interest was the direct formation of these atoms by transfer of an electron from a target atom,

$$H^+ + X \rightarrow H^*(3s,\ 3p,\ \text{or}\ 3d) + X^+. \qquad (1)$$

Targets X of helium and of molecular nitrogen were used.

A process of secondary interest was the collisional destruction of excited atoms prior to their spontaneous radiative decay,

$$H^*(3s,\ 3p,\ \text{or}\ 3d) + X \rightarrow [H^+ + e] + [X]. \qquad (2)$$

The brackets are used to indicate that the present experiments provided no information on the states of the postcollision products except for the fact that the H atom was no longer in the $n = 3$ level. However, theoretical predictions[1] indicate that nearly all such collisions at the impact velocities of this experiment result in ionization of the hydrogen atoms.

Various approximation methods have been developed to predict cross sections for the types of collisions described by Eqs. (1) and (2). An important objective of the present experiment was to test theoretical predictions of charge-transfer cross sections.

It is argued that a detailed comparison of theory and experiment for the charge-transfer formation of specific excited states may be carried out effectively for the $n=3$ level. Measurements for states with $n\geq 6$ which have been made by field ionization techniques[2] provide, at best, a sum of the cross sections for the different angular momentum states l and often do not allow resolution of states with different principal quantum numbers n.[3] The features of the cross section for formation of a particular state nl are frequently hidden in such a sum and these experiments have therefore not provided very sensitive tests of theory. For states having $n\geq 4$, the stray fields commonly encountered in experimental systems are sufficient to cause Stark mixing of the sublevels, thereby destroying their separate identity. It is therefore concluded that a definitive comparison of experimentally measured cross sections for capture into excited states with theoretical predictions of such cross sections may be carried out only on the $n=3$ and $n=2$ states. The formation of the $n=2$ level has been extensively studied elsewhere.[4]

II. EXPERIMENTAL TECHNIQUE

The formation of excited H atoms in the $3s$, $3p$, and $3d$ states can be detected by the quantitative measurement of the Balmer α photons emitted as the excited atoms decay to the $n=2$ level. The Balmer α (H_α) emission is, in fact, due to three transitions: $3s\rightarrow 2p$, $3p\rightarrow 2s$, and $3d\rightarrow 2p$. These all emit photons of essentially the same wavelength and are therefore detected simultaneously. Other means than spectroscopic separation must be employed if the three contributions to the H_α emission are to be separately identified.

At the high impact energies utilized in this experiment, the product of the projectile's velocity and the lifetime of the excited state is a length of several cm. Therefore, in general, a projectile will move an appreciable distance from the point where it was excited before emitting a photon and decaying to a lower state. As a result, the intensity of emission from the projectiles is a function of the position along the flight path at which the observation is made and also of the lifetime of the emitting state. Measurement of the spatial variation of this emission intensity allows the contributions of the $3s$, $3p$, and $3d$ states to be separately identified and the populations of the three emitting states to be evaluated. This principle has been used by Hughes *et al.*[5-7] to measure electron capture cross sections at energies somewhat lower than those of the present experiment.

Consider the population of hydrogen atoms in an excited state j formed by charge transfer as protons traverse a target gas. Initially the assumptions are made that (a) the state j is populated only by direct electron capture and that cascade may be neglected and (b) atoms are removed from the state j only by the processes of spontaneous radiative decay; these assumptions will be reexamined later.

A current of F projectile ions per second is incident with a velocity v (cm/sec) on a target which has a number density ρ (molecules/cm^3). Let x be distance measured along the beam axis from the point of entrance to the target region. Let n_j^* be the number of excited atoms in state j per unit length along the beam axis. During time interval dt, the incremental change in n_j^* from direct collisional formation and spontaneous radiative decay is given by

$$dn_j^* = F\rho Q_j dt - n_j^* \sum_{i<j} A_{j\rightarrow i}\, dt\,. \tag{3}$$

Here $A_{j\rightarrow i}(\text{sec}^{-1})$ is the transition probability for spontaneous decay of the state j to the state i,

$$\sum_{i<j} A_{j\rightarrow i} = \tau_j^{-1}\,,$$

where τ_j is the lifetime of the excited state. Since $v=dx/dt$, Eq. (3) may be rewritten as a function of x instead of time,

$$\frac{dn_j^*}{dx} = \frac{F\rho Q_j}{v} - \frac{n_j^*(x)}{v\tau_j}\,. \tag{4}$$

If a further assumption is made that the proton beam current F is not significantly depleted in passing through the target region, the solution of Eq. (4) is given by

$$n_j^*(x) = F\rho Q_j \tau_j (1 - e^{-x/v\tau_j})\,. \tag{5}$$

Let J_{jk} be the number of photons emitted per second in the transition $j\rightarrow k$ from a segment of beam path whose center is at x and whose length is d. If d is much less than $v\tau_j$, as it is for the present experiment, the approximation may be used that

$$J_{jk}(x) = A_{j\rightarrow k} n_j^*(x)\, d\,.$$

It is convenient to define a normalized emission function $G_{jk}(x)$ as the number of photons emitted per second in transition $j\rightarrow k$ at position x per unit length of beam per unit incident beam flux per unit target density. Since, in the present experiment, the Balmer α emission is due to three transitions, the observed emission function is a sum of three terms of the type given in the equation for J. Using Eq. (5), we obtain

$$G_\alpha(x) \equiv \frac{J_\alpha(x)}{F\rho d} = I_{3s}\left[1 - \exp\left(-\frac{x}{v\tau_{3s}}\right)\right]$$

$$+I_{3p}\left[1-\exp\left(-\frac{x}{v\tau_{3p}}\right)\right]$$
$$+I_{3d}\left[1-\exp\left(-\frac{x}{v\tau_{3d}}\right)\right]+K, \qquad (6)$$

where $I_j = Q_j A_{j\to k}\tau_j$ and a term K, independent of position, has been included to allow for contributions to the detected signal from collisionally induced target emission. It happens that the $3s$ and $3d$ states can decay spontaneously only by the Balmer α transition and therefore the branching ratio $A_{j\to k}\tau_j$ for these states is unity. For the $3p \to 2s$ transition, this ratio is 0.118, [8] indicating that only 11.8% of the atoms in the $3p$ state decay by the emission of a Balmer α photon, the rest by Lyman β. Equation (6) represents a sum of three terms which increase exponentially with x toward an asymptote. Because the three lifetimes of the states are quite different, it is possible to compare the measured function $G_\alpha(x)$ with Eq. (6) and to evaluate the coefficients I_j. In this manner, the relative values of cross sections for the formation of the $3s$, $3p$, and $3d$ states may be measured using the different lifetimes to identify the three sublevels.

III. APPARATUS

The apparatus for the experiment is shown schematically in Fig. 1. The source of incident protons was a 1-MeV Van de Graaff positive-ion accelerator equipped with a beam analyzing and stabilizing system. The incident proton energy was determined to within ± 2 keV by deflection through 90° in a regulated magnetic field. Beam currents of 0.3–3.0 μA were typically employed. The incident proton beam was collimated to $\frac{1}{16}$ in. diam by two knife-edged orifices spaced 6 in. apart. A third orifice of larger diameter was suitably biased to collect secondary electrons. A fourth orifice in the form of a short channel provided the limiting aperture between the collision chamber and the accelerator to inhibit the loss of target gas from the cell. This orifice had a diameter such that no particles which had traversed the first two apertures could be incident upon it, thereby reducing the possibility of secondary electrons and sputtered material entering the observation region.

The ion beam was monitored after traversing the collision and detection region on a deep parallel-plate Faraday cup assembly with an inclined end. Suitable biases were applied to the beam collection system to insure complete suppression of secondary electrons and ions. Ion-beam currents were measured by an electronic microammeter. It was possible for particles in the beam to be scattered in passing through the target by an angle sufficient to prevent their entrance into the Faraday cup. An annular ring formed the entrance aperture of the Faraday cup and the current collected by it remained at all times below 1% of the current collected by the Faraday cup. A large grounded plate isolated the electrostatic fields of the Faraday cup from the collision region.

Under certain conditions of operation, appreciable neutralization of the projectile beam occurred in the long flight tube so that the current to the Faraday cup no longer represented the flux of ions at the entrance to the collision chamber. To obtain the current F of Eqs. (3)–(6), corrections were

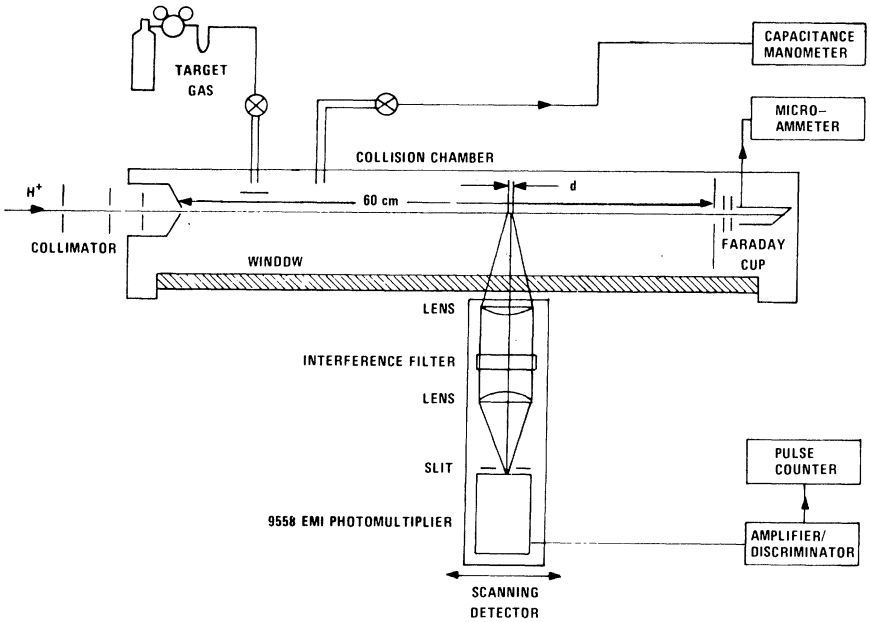

FIG. 1. Schematic diagram of the apparatus for measuring emission in the target region.

applied for beam neutralization using the well-known formulations by Hasted[9] with total charge transfer and stripping cross sections previously measured by Barnett and Reynolds[10] and by Stier and Barnett.[11]

The target gas was passed through a cold trap to remove any condensable materials and was leaked into the collision chamber. The purity of the helium used was stated by the manufacturer to be at least 99.999% and of the nitrogen 99.9%. The target gas pressure was monitored continuously by a capacitance manometer which had been calibrated against a McLeod gauge.

A window of crown glass in one side of the collision chamber allowed a view of the entire beam path. The Balmer α detector could be moved along a machined track to measure emission intensity at any position along a 60 cm length of the flight path. Light emitted from a short segment of the beam was focused at infinity by a lens, passed at normal incidence through an interference filter, and refocused by a second lens to form an image of the beam segment on the face of an EMI 9558 photomultiplier tube. A slit placed just in front of the tube's face limited its view to a 6-mm segment of the beam. A survey was made of the point-to-point variations in sensitivity over the face of the tube, and its orientation was chosen such that the variation in sensitivity over the exposed portion was less than 2%. The photomultiplier was operated in the pulse mode and its output was fed through a preamplifier, amplifier, and discriminator and counted by scaling equipment. The photomultiplier was thermoelectrically cooled to reduce its dark current.

In order to measure on an absolute basis the normalized emission function $G_\alpha(x)$, it is necessary to establish the efficiency with which photons are detected. This was accomplished by comparison of H_α emission intensity obtained from the charge exchange process with that obtained from the dissociative excitation of molecular hydrogen in the process

$$\begin{aligned} &H^+ + H_2 \rightarrow [H^+ + H] + H^*(n=3), \\ &H^*(n=3) \rightarrow H^*(n=2) + h\nu(\text{Balmer } \alpha). \end{aligned} \tag{7}$$

The absolute cross section for emission of the H_α line by dissociation of an H_2 target was measured previously in this laboratory[12] and was used as a transfer standard. Thus the present data are essentially normalized to a cross section of 29.9×10^{-20} cm^2 for the process described by Eq. (7) at an impact energy of 250 keV.

The optical system accepts light emitted into a 12° cone centered at 90° to the beam axis. Consequently the line emitted by the excited projectiles undergoes a Doppler broadening which ranges from 34 Å at 75 keV to 80 Å at 400 keV. Since the interference filter selects only a narrow bandwidth of radiation, the effective sensitivity of the detector varies with the impact energy of the incident protons. It should be emphasized that this dependence had no effect on measurements of the relative magnitudes of Q_{3s}, Q_{3p}, and Q_{3d} at a given energy, but it did affect the apparent dependence on energy of these cross sections. A technique was developed to correct for this variation in sensitivity. The relative sensitivity of the detector to photons incident on each element of the optical aperture was evaluated as a function of the wavelength of the incident photons. By summing over the entire aperture, the relative sensitivity of the detector could be determined as a function of the velocity of the source of emission. Figure 2 shows the relative variation of the detection sensitivity with projectile energy for the two filters used in these experiments.

IV. ANALYSIS OF THE NORMALIZED EMISSION FUNCTION

In Sec. II is an outline of the basic theory by which the signal from the experiment may be related to the required cross sections. This simple theory assumes that there is a negligible probability of a particle undergoing more than one collision as it traverses the target. In principle such "single collision" conditions can be obtained by operating at sufficiently low target densities. Unfortunately, the cross sections for certain of these secondary processes are some orders of magnitude greater than the cross section of interest. Consequently, single collision conditions can be guaranteed only at very low target densities where signal strengths are weak and the statistical accuracy with which $G_\alpha(x)$ may be

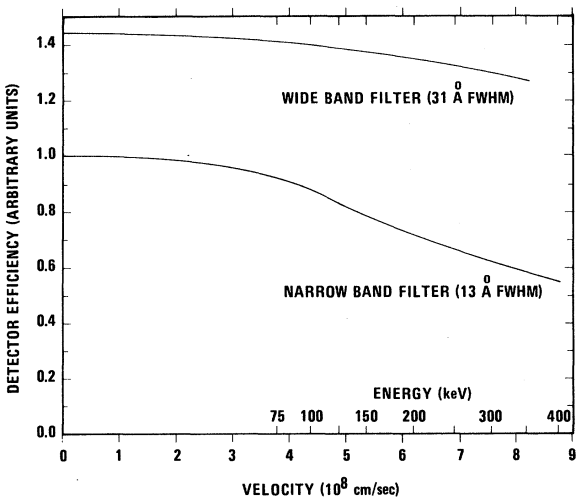

FIG. 2. Relative detection efficiency of the Balmer α detector as a function of the velocity of the emitting atom.

measured is inadequate to deconvolute the separate exponential terms. Consequently it proved necessary to operate the experiment under density conditions where some secondary collision mechanisms were important and to modify the analysis accordingly.

The major contributions to error in the simple analysis of Sec. II are as follows: (a) neglect of the process of collisional destruction of excited atoms (Eq. 2) before decay occurs; (b) neglect of the attenuation of the proton current with x; and (c) neglect of excitation to the $3s$, $3p$, and $3d$ states of H(1s) atoms formed in the beam by charge transfer. The most serious problem is the collisional destruction, the cross section for which we will denote as Q_I; its presence requires the inclusion of a loss term $-[n_j^*(x)\,\rho Q_I]$ to the right-hand side of Eq. (4). The effect of collisional destruction is to reduce the apparent lifetime of the excited state by collisional effects. The normal radiative lifetime τ_j in Eqs. (5) and (6) must then be replaced by an effective lifetime $\tau_j/(1+v\tau_j\rho Q_I)$. The decrease of ion flux and the excitation of neutral atoms have a complicated influence on the measurements; their effect on the signal does not vary with x in a simple manner.

If one allows for collisional destruction of excited atoms, attenuation of the proton current, and also excitation of neutral atoms, the equation governing the linear density of excited atoms in the $3s$ state n_{3s}^* is given by

$$\frac{dn_{3s}^*(x)}{dx} = -n_{3s}^*(x)\left(\frac{1}{v\tau_{3s}} + \rho Q_{I,3s}\right) + n_+(x)\rho Q_{3s} + n_0(x)\rho Q_{x,3s}, \tag{8}$$

where $n_+(x)$ is the proton density, $n_0(x)$ the density of neutrals, and $Q_{x,3s}$ is the cross section for excitation of fast neutral H(1s) atoms to the $3s$ state. Similar equations can be written for the $3p$ and $3d$ populations. The solution of (8) is

$$n_{3s}^*(x) = \frac{n_+(0)\rho}{\sigma_s+\sigma_c}\left[\left(\frac{\sigma_s Q_{3s}+\sigma_c Q_{x,3s}}{1/v\tau'_{3s}} + \frac{\sigma_c(Q_{3s}-Q_{x,3s})}{1/v\tau'_{3s}-\rho(\sigma_s+\sigma_c)}\right)\times(1-e^{-x/v\tau'_{3s}}) - \left(\frac{\sigma_c(Q_{3s}-Q_{x,3s})}{1/v\tau'_{3s}-\rho(\sigma_s+\sigma_c)}\right)(1-e^{-\rho(\sigma_s+\sigma_c)x})\right], \tag{9}$$

where

$$\tau'_{3s} = \tau_{3s}/(1+v\tau_{3s}\rho Q_{I,3s}).$$

Populations of the $3p$ and $3d$ states are given by similar equations. The complete equation for the normalized emission function, including a position-independent contribution from the target or background gas, is

$$G_\alpha(x) = (F\rho)^{-1}[A_{3s\to 2p}\,n_{3s}^*(x) + A_{3p\to 2s}n_{3p}^*(x) + A_{3d\to 2p}n_{3d}^*(x)] + K, \tag{10}$$

where $F = vn_+(0)$ is the proton current at $x=0$. Equation (10) replaces Eq. (6) as a description of the experiment. The three exponential terms of Eq. (6), which are dependent on the excited-state lifetimes, have remained in the expressions for n^*, but each decay length $v\tau$ has been replaced by $(1/v\tau + \rho Q_I)^{-1}$. The expressions corresponding to I_{3s}, I_{3p}, and I_{3d} have become more complicated. Also an additional exponential term in $(1-e^{-\rho(\sigma_s+\sigma_c)x})$ has appeared. It is important to note that Eq. (6) expresses the limiting value of $G_\alpha(x)$ as ρ, the target gas density, approaches zero.

Equations (8)–(10) are clearly very complete. Some simplifications are necessary in order that the measurements of $G_\alpha(x)$ may be analyzed to provide cross sections. There are no previous measurements of any of these cross sections to provide guidance as to which of the secondary processes might be neglected in the description of this experiment. Some guidance can be obtained by considering theoretical values of Q_{3l} and $Q_{x,3l}$ for $H^+ + H$ and $H + H$ collision combinations, but there must be doubt as to their applicability to the present situation.

An immediate simplification may be made in the use of the destruction cross section $Q_{I,3s}$. Bates and Walker[1] predict that the cross sections $Q_{I,3s}$, $Q_{I,3p}$, and $Q_{I,3d}$ are approximately equal. This result was assumed in the analysis of the present data and all three cross sections set equal to a single value Q_I. It should be noted that for the densities used in this experiment,

$$\rho Q_{I,3p} \ll 1/v\tau_{3p} \text{ and } \rho Q_{I,3d} \ll 1/v\tau_{3d},$$

thus the contribution of destruction to the depopulation of the $3p$ and $3d$ states is in any case negligible. Moderate errors in the assumed equality of these three cross sections will produce no appreciable errors in the analysis of the data.

To proceed further with the analysis, two basic assumptions were made that simplify Eqs. (9) and (10). First, the term involving $(1-e^{-\rho(\sigma_s+\sigma_c)x})$ is undoubtedly small and varies slowly over the observation region of the experiment; consequently it was neglected. Second, it was assumed that Q_x could be neglected in the remaining terms.

With these simplifications described above, the normalized emission function is given by

$$G_\alpha(x) = \sum_{l=s,p,d} I'_{3l}[1-e^{-x/v\tau'_{3l}}] + K, \tag{11}$$

where

$$I'_{3l} = \frac{A_{3l\to 2l'}\,Q_{3l}}{v(\sigma_s+\sigma_c)}\left(\frac{\sigma_s}{1/v\tau'_{3l}} + \frac{\sigma_c}{1/v\tau'_{3l}-\rho(\sigma_s+\sigma_c)}\right), \quad \times \begin{cases} l = s,p,d \\ l' = p,s,p \end{cases}.$$

In this equation only Q_{3s}, Q_{3p}, Q_{3d}, Q_I, and K are unknown.

The collisional destruction cross section Q_I was obtained for the 3s state by analysis of data for which x is sufficiently large that 3p and 3d contributions have essentially reached their equilibrium values. The data should fit an equation of the form

$$G_\alpha(x) = I'_{3s}(1 - e^{-x/v\tau'_{3s}}) + I'_{3p} + I'_{3d} + K . \qquad (12)$$

Q_I was determined by adjusting τ'_{3s} to obtain the best possible fit of Eq. (12) to the data according to the least-squares criterion. The measured $G_\alpha(x)$ was then fitted to an equation of the form of Eq. (11) and values of Q_{3s}, Q_{3p}, Q_{3d}, and K were determined. The entire procedure was repeated for data taken at different target densities.

Clearly the adequacy of this very complicated analysis requires some verification. The major criterion for its adequacy is that the cross sections obtained by fitting Eq. (11) to the measured emission function should be independent of target density. This criterion was demonstrated to be fulfilled at pressures from 0.2 up to 1.5 μ Hg in helium and from 0.2 to 1.0 μ Hg in nitrogen; lower target pressures could not be used due to degradation of the signal-to-noise ratio. It is concluded therefore that the analysis is satisfactory.

V. ASSESSMENT OF CASCADE

In addition to direct collisional excitation, the 3s, 3p, and 3d states may also be populated by cascade from higher levels. This fact has two important consequences. First, the measured cross section will then not represent only the formation of the state by collision but will include a component due to cascade. Second, and perhaps more important, the dependence of emission intensity on x will be different for atoms formed in $n = 3$ states through cascade than for atoms formed directly. This second problem might invalidate the analysis which separates the cross sections by means of a deconvolution technique based on the assumed values of 3s, 3p, and 3d state lifetimes.

The population of higher n states may be estimated by taking the present measurements of the 3s, 3p, and 3d state cross sections and scaling them to higher n states assuming that cross sections for a given angular momentum substate decrease as n^{-3}. This general rule is well established[13,14] by theory and serves well for an approximate assessment of the problem. The only cascade contributions of any significance are from the ns states into the 3p level.

Since at the energies of this experiment the n^{-3} rule is only an estimate (although measurements by Hughes *et al.*[15] of Q_{3s} and Q_{4s} near 100 keV tend to confirm it), no cascade correction has been applied to the data. The n^{-3} rule has been used only to estimate the uncertainty in the cross sections due to cascade. This uncertainty proved to be rather small in comparison with other known uncertainties.

VI. EFFECTS OF POLARIZATION

Emission from the 3p and 3d states may exhibit polarization. The polarization fraction is related to the population of the substates having different magnetic quantum numbers, and is zero if these states are all equally populated. It may be shown that the collisionally induced emission will be anisotropic if polarization is present. A measurement of emission at one angle does not allow the determination of a cross section unless correction is made for this anisotropy.

Much of the present research has been directed at the $3s \rightarrow 2p$ emission which is unpolarized and therefore emitted isotropically. No attempt has been made to measure polarization for the $3p \rightarrow 2s$ and $3d \rightarrow 2p$ emissions. Because of the small signal intensity from these states, the statistical accuracy would be so poor as to render the measurement meaningless. Consequently, it is not known whether the emissions are isotropic. However, upper and lower bounds can be placed on the degree of polarization possible in these emissions, and the resulting uncertainties in the 3p and 3d capture cross sections may be evaluated.

It should be noted that any polarization which may exist in the p- and d-state radiations can have no effect on the separation of the contributions of the three parent states to the detected radiation. The neglect of an anisotropic radiation pattern will, however, cause error in the values of the 3p and 3d capture cross sections interpreted from these contributions. If the polarization of these emissions varies with energy, its neglect will result in error in the energy dependence of the p and d cross sections.

It is possible to predict the polarization fraction of the $3p \rightarrow 2s$ and $3d \rightarrow 2p$ emissions in terms of the cross sections for populating the magnetic substates. Percival and Seaton[16] give an equation for the polarization of $2p \rightarrow 1s$ radiation which holds approximately for the $3p \rightarrow 2s$ radiation.[17] Hughes *et al.*[6] have derived an analogous expression for the $3d \rightarrow 2p$ emission. Maximum and minimum values of the polarization fraction will occur when the momentum transfer is, respectively, parallel to or perpendicular to the axis of quantization. On this basis the polarization P of the emission for the $3p \rightarrow 2s$ transition must lie in the range $-0.267 \leq P \leq 0.421$ and for the $3d \rightarrow 2p$ transition in the range $-0.32 \leq P \leq 0.48$. It may be readily inferred that absence of any detailed knowledge of the polarization fraction results in an uncertainty of $+9$ to -14% in the $3p \rightarrow 2s$ transition and $+11$ to -16% for the $3d \rightarrow 2p$.

No corrections have been made in the present work but these limitations are included in the estimated uncertainties of the data.

VII. STARK EFFECT MIXING

The experiment is designed to determine cross sections for the formation of the $3s$, $3p$, and $3d$ states using the lifetimes of these states for identification. However, if an electric field is applied to the excited atoms, the energy levels will be perturbed by the Stark effect, and "mixing" of certain states will cause changes in the effective lifetimes of the excited states. There is a danger that stray fields in the apparatus may cause this effect.

The states which are most vulnerable to mixing are those having the same value of the total angular momentum quantum number j. For the $n = 3$ level, the critical fields, i.e., the minimum fields which will cause full mixing, are 58 V/cm for the $3s_{1/2}$ and $3p_{1/2}$ states and 1.9 V/cm for the $3p_{3/2}$ and $3d_{3/2}$ states.[18]

Clearly, the weak-field Stark effect may distort the operation of the experiment. It would be impossible to correct the data for the effects of substantial stray fields since they would probably vary in space and time. In principle, it should be possible to design a field-free experiment, but this would entail a considerable increase in complexity. Instead, some simple precautions were taken to reduce the possibility of Stark mixing, and a test was made to determine whether mixing was affecting the experiment.

The collimation system was designed so that the beam could not strike any part of the gas cell aperture as it entered the target region. The only surfaces exposed to the beam were clean conducting surfaces so that accumulation of the static charge was unlikely. The window through which the H_α radiation was observed was an exception to this statement, but it was located at the largest practicable distance from the beam.

Finally, Stark plates were installed in the observation region so that electric fields could be intentionally applied to the beam. The application of these fields showed that the $3s$ state was not affected by any fields which might conceivably exist in the apparatus. It was not possible to prove conclusively that the $3p$ and $3d$ states, which made only small contributions to the total H_α emission, were completely free from mixing because the data were subject to random fluctuations from other sources. However, there was no detectable evidence that these states were mixed by fields which existed in the apparatus, and it will be assumed in the presentation of data that there was no mixing.

VIII. EXPERIMENTAL UNCERTAINTY

Uncertainties in the measurements arose from a number of independent causes. There were random errors in the data due to statistical variations and small short-term fluctuations in the measuring devices. Their effects have been assessed by noting the degree of reproducibility of the cross-section measurements from one set of data to another.

In addition, there was systematic uncertainty resulting from the possibility of errors in determination of the following quantities: target gas density, impact velocity, beam current, the correction for Doppler effects, the position of the effective boundary of the target region, the variations in sensitivity over the exposed portion of the photomultiplier face, and the intensity of the standard source of emission. Additional uncertainties were due to possible cascade contributions to the emission and, in the case of cross sections for capture into the $3p$ and $3d$ states, the possibility that the emission was polarized.

Most of the uncertainty in the measurements of the collisional destruction cross section Q_I is due to random fluctuations. Uncertainties in ρ, v, τ_{3s}, and cascade contributions result in relatively small uncertainties in Q_I. The values of Q_I presented in this paper are the weighted averages of several determinations made using Eq. (12). Greater weight was given to the determinations made at higher target densities because of their greater accuracy. The error bars shown in the figures include all but one of the determinations.

Ignoring for the moment the uncertainty in the absolute calibration, the measurements of cross sections for electron capture into the $3s$ state are estimated to have a total uncertainty of $\pm 15\%$ or less in almost all cases. The estimates are shown by the error bars in Figs. 3 and 4. The largest single contribution to the indicated uncertainties was due to the uncertainty in Q_I.

To these uncertainties must be added the $\pm 50\%$ uncertainty in the absolute calibration of the sensitivity of the H_α detector. This has been omitted from the figures for clarity since an error from this source cannot affect the energy dependence of the cross section but could only raise or lower all the points by equal distances on the figures.

Cross sections for capture into the $3d$ states are presented in a similar way in Figs. 5 and 6 omitting the estimated uncertainty in calibration. These cross sections are about two orders of magnitude smaller than the cross sections for formation of the $3s$ state. Therefore only a few percent of the measured light intensity is due to transitions from the $3d$ state. Random variations in the data are typically 1–2%, sometimes larger, As a result, the random variations in the measured values are sometimes more than 100% of the mean, and it has been necessary to assign an uncertainty factor of 2.5 (+150%, −60%) to these measurements.

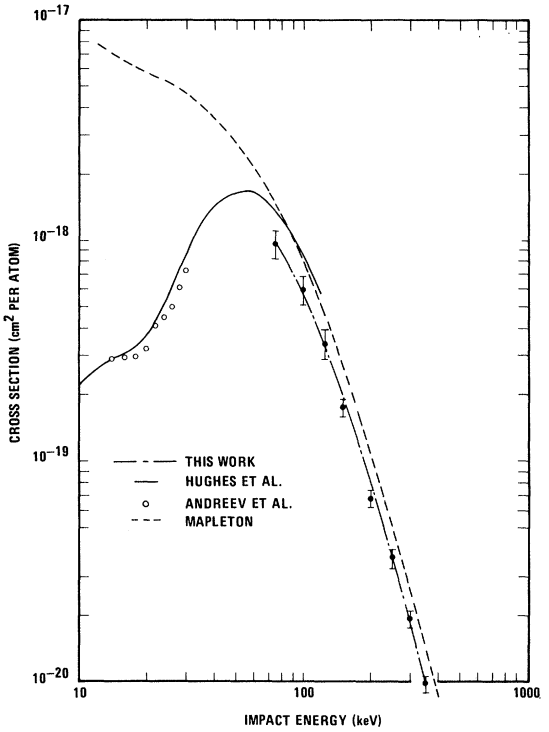

FIG. 3. Cross section for the formation of H(3*s*) atoms in helium. The process is represented by the equation $H^+ + He \rightarrow H(3s) + He^+$. Present measurements are shown along with those made by Hughes *et al.* (Ref. 7) and by Andreev *et al.* (Ref. 19). Also shown are predictions of the Born approximation calculated by Mapleton (Ref. 20) using the postcollision potential for the process $H^+ + He(1s^2) \rightarrow H(3s) + He^+(1s)$.

Random variations in the measurements of the cross section for capture into the 3*p* state are even larger than for the 3*d* state. The reason again is that only 1 or 2% of the Balmer α emission is due to radiation from atoms in the 3*p* state. The cross section for formation of this state is about an order of magnitude smaller than the cross section for formation of the 3*s* state, and less than 12% of the 3*p* atoms decay by emission of an H_α photon. The remainder decay by Lyman β. Because of large random variations in the measurements of the 3*p* cross sections, the results are not presented. However, the measured values do allow the establishment of an upper bound for the cross section.

IX. MEASURED VALUES OF THE CROSS SECTIONS

Measurements of the cross section for the formation of the excited states of atomic hydrogen are shown in Figs. 3–6 for targets of He and N_2. For comparison, the predictions by Mapleton and the previous measurements by Hughes *et al.*[7] and by Andreev *et al.*[19] are also shown.

Figure 3 shows that the general form of our measurements for capture into the 3*s* state from a target of helium is in agreement with Mapleton's predictions.[20] The systematic discrepancy between theory and experiment might be due to an erroneous calibration of detection sensitivity. It appears that the present measurements confirm the general validity of Mapleton's theory down to impact energies of 75 keV. The cross section for capture into the 3*d* state from helium (Fig. 5) is about two orders of magnitude smaller than the cross section for the formation of the 3*s* state. Despite the poor accuracy of the data on the 3*d* state, the agreement between theory[20] and experiment is surprisingly good.

The measurements of the cross sections for the formation of the 3*p* state in a target of helium are so poor that it would be misleading to present them here. Generally speaking, they do agree with the measurements by Hughes *et al.*[7] and lie below the theoretical predictions of Mapleton[20] by a factor of 4 or more at all energies from 75 to 400 keV. This very large discrepancy is surprising. There seems no obvious reason why theoretical predictions should be good for the 3*s* and 3*d* states while being very poor for the 3*p* level. There are also general theories for the prediction of relative populations of excited states from the work of Hiskes[21–23] and

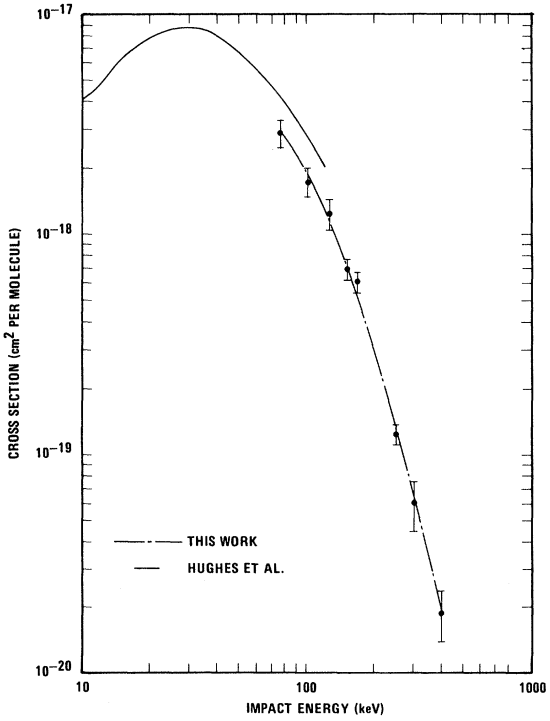

FIG. 4. Cross section for the formation of H(3*s*) atoms in nitrogen. The process is represented by the equation $H^+ + N_2 \rightarrow H(3s) + N_2^+$. Present measurements are shown along with those made by Hughes *et al.* (Ref. 7).

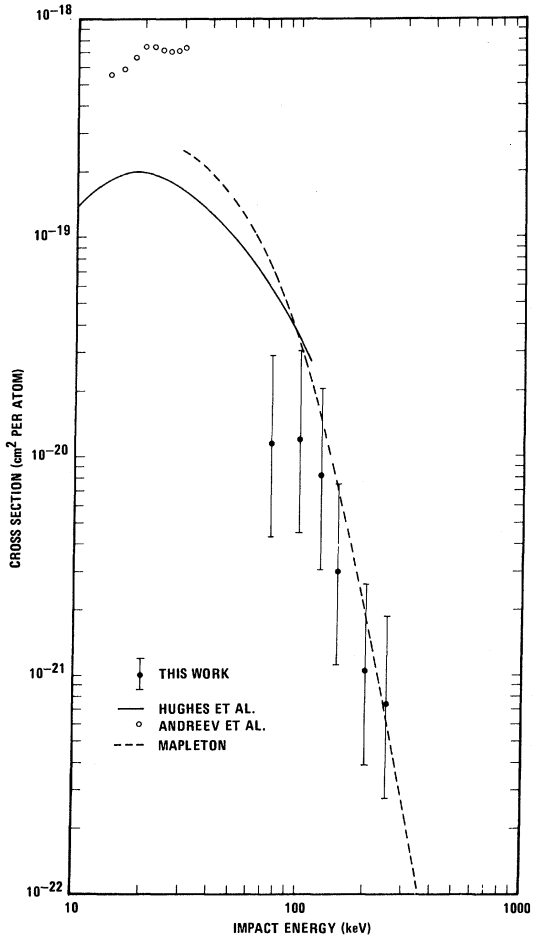

FIG. 5. Cross section for the formation of H(3*d*) atoms in helium. The process is represented by the equation $H^+ + He \rightarrow H(3d) + He^+$. Present measurements are shown along with those made by Hughes *et al.* (Ref. 7) and by Andreev *et al.* (Ref. 19). Also shown are predictions of the Born approximation calculated by Mapleton (Ref. 20) using the postcollision potential for the process $H^+ + He(1s^2) \rightarrow H(3d) + He^+(1s)$.

of Butler and May.[24] These theories are in general agreement with the work by Mapleton[20] and therefore in disagreement with the present experiment. The present work supports the conclusion by Hughes *et al.*[7] that the available theories seriously overestimate the 3*p* cross section.

The impact-parameter formulation has been used by Sin Fai Lam[25] to calculate cross sections for the electron capture reactions

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

over the energy range 1–1000 keV. The calculation was performed with allowance for coupling among these states during the collision process. Sin Fai Lam's predictions of the cross sections for capture into the 2*s* and 2*p* states have been scaled by the n^{-3} rule[13,14] to produce estimates of the 3*s* and 3*p* cross sections. Although the comparison of present measurements with these scaled results shows a substantial discrepancy in magnitude for both the *s* and *p* cross sections, the ratio of his predicted cross sections is much nearer the experimental results than the predictions of this ratio by Mapleton and by Hiskes. It would appear that further work to include coupling to yet other competing processes, as suggested by Sin Fai Lam[25] and by Bransden and Sin Fai Lam,[26] might further elucidate this problem.

Figures 4 and 6 show the cross sections for formation of the 3*s* and 3*d* states of H by impact of H^+ on a target of N_2. Their magnitudes are several times larger than for a helium target, but the energy dependences are similar. There are no detailed theoretical predictions with which these may be compared. Where a direct comparison with measurements by Hughes *et al.*[7] is possible, their cross

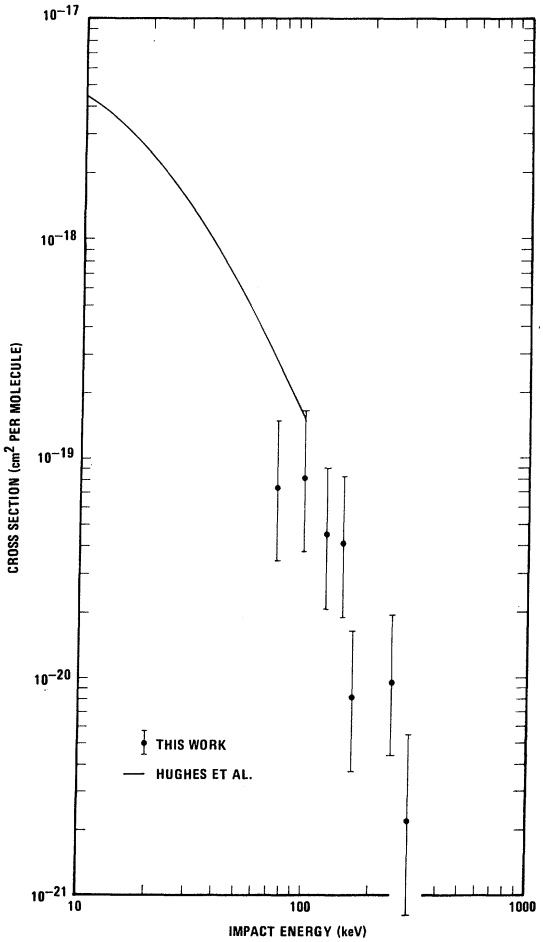

FIG. 6. Cross section for the formation of H(3*d*) atoms in nitrogen. The process is represented by the equation $H^+ + N_2 \rightarrow H(3d) + N_2^+$. Present measurements are shown along with those made by Hughes *et al.* (Ref. 7).

sections for electron capture appear to be about 40–50% larger than the present measurements due to a discrepancy in absolute calibration of detection efficiency. Even so, the two sets of measurements are within the combined experimental uncertainties and are considered to be in agreement.

Figures 7 and 8 show the cross sections for the destruction of the 3*s* excited state by impact on targets of helium and nitrogen, respectively. Comparisons are made with Bates and Walker's theoretical predictions of the cross sections for ionization of this state.[1] Ionization, they claim, is the principal mechanism for quenching H_α radiation under the conditions of the present experiment. In the case of a nitrogen target, the magnitude of the present measurements appears to be in agreement with these predictions within experimental uncertainty, although a somewhat different energy dependence is indicated. For a target of helium, the magnitudes of the predictions and the measurements differ by a little more than the estimated experimental uncertainties, but in no case more than a factor of 2. However, the energy dependences are substantially different.

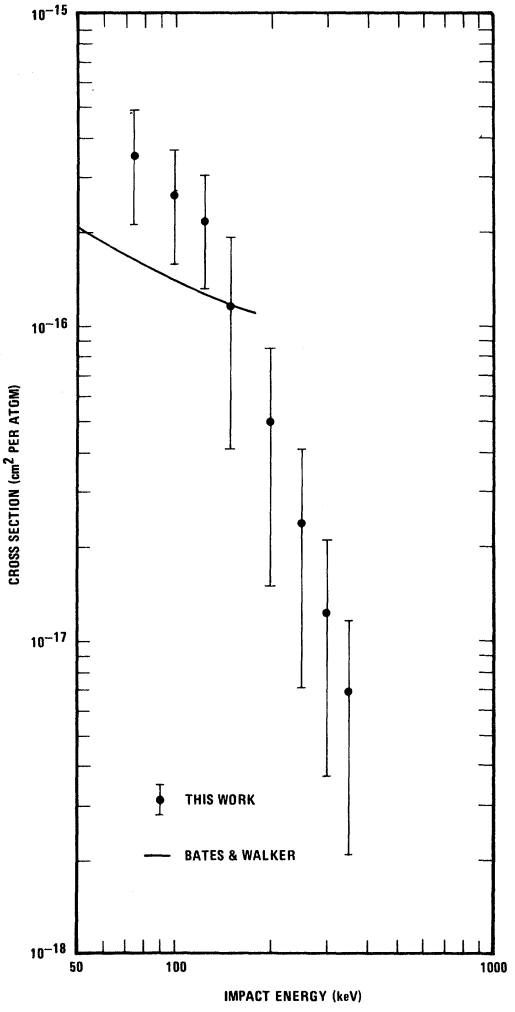

FIG. 7. Cross section for the collisional destruction of H(3*s*) atoms by impact on helium. Present measurements are shown along with predictions by Bates and Walker (Ref. 1) for the ionization of H(3*s*) atoms by impact on helium.

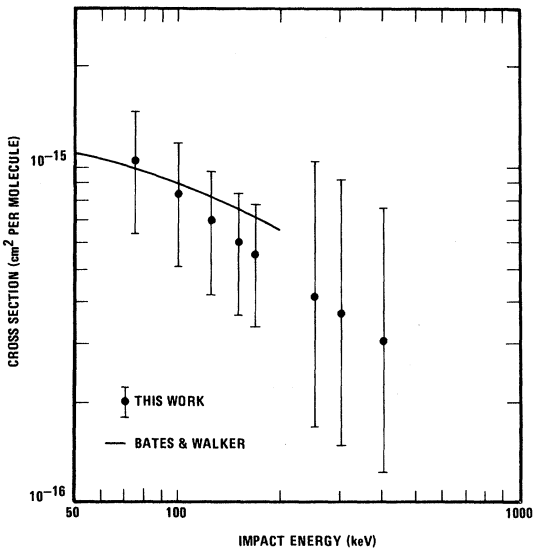

FIG. 8. Cross section for the collisional destruction of H(3*s*) atoms by impact on nitrogen. Present measurements are shown along with predictions by Bates and Walker (Ref. 1) for the ionization of H(3*s*) atoms by impact on nitrogen.

X. CONCLUSION

Charge transfer into the 3*s* and 3*d* states on helium target is in agreement with the theoretical predictions of Mapleton within the accuracy of the present experiments. The theoretical predictions for the 3*p* state, however, exceed the measured values by a factor of 4 or more. This discrepancy indicates a serious inadequacy of the theory. It seems likely that coupled state calculations might show an improved agreement between theory and experiment.

For a nitrogen target, the capture cross sections are about a factor of 3 higher than for helium, and slightly larger fractions of the $n = 3$ population go to the *p* and *d* states.

The present measurements of collisional destruction cross sections are not of high accuracy but do appear to confirm the general predictions of the theory by Bates and Walker. This cross section is several orders of magnitude larger than the cross section for capture into the 3*s* state.

*This work was supported in part by the Controlled Thermonuclear Division of the Atomic Energy Commission and a more detailed account is to be found in AEC Report No. ORO-2591-47.

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