Experiments on the Production and Extinction of the 2s State of the Hydrogen Atom*

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The rates of production and extinction of the metastable 2s state of the hydrogen atom during passage of an initially pure proton beam through molecular hydrogen are studied. Iodine-filled Geiger counters were used to detect the Lyman alpha radiation from the metastable beam component, which was quenched by electric fields for purposes of observation. An observed peak in the growth of the metastable beam component at 15 keV projectile energy leads to a cross section for the extinction of the metastable atom by collision with molecular hydrogen in excess of 10^{-15} cm^2 per target atom. The rate of extinction of the metastable atom by dc electric fields between 50 and 500 V/cm is studied and the weak-field time-independent perturbation-theory treatment of this process is verified. This extinction rate is obtained by observing the decay length of the Lyman alpha radiation emerging from the metastable beam component as a function of electric field. The effective cross section for electron capture into the 2s state by 15-keV protons in hydrogen is compared with the best available cross section for the process $e+H_2 \rightarrow$ countable uv, and is estimated to be 1×10^{-17} cm^2 per target atom. For the same energy and target, the effective cross section for excitation of 1s projectiles to the 2s state is estimated to be 1.5×10^{-18} cm^2 per target atom.

INTRODUCTION

A BEAM of kilovolt energy metastable hydrogen atoms in the 2s state can be obtained as one constituent of a neutral beam formed by electron capture when a beam of protons passes through a gas target.[1] One of the principal contributions to the total electron-capture cross section at these energies for protons in hydrogen comes from capture into the 2s state.[2,3] Once formed, this fragile metastable component of the neutral beam has a lifetime limited by collisions with the target gas or with residual gases in the vacuum system, and by any electric or magnetic fields present. The primary purpose of the experiments reported here was to study these production and extinction processes for protons in hydrogen.

The experiments to be described give information about the rates of production and extinction of the metastable atoms formed as an initially pure proton beam of 15-keV energy passed through various amounts of dry hydrogen gas, as well as the rate of destruction of the metastable atoms by uniform electric fields between 50 and 500 V/cm. These topics have received limited experimental attention in the past.[2,4] No theoretical calculations of the cross sections for capture into the metastable state or for destruction of the metastable atom by collision with molecular hydrogen as the target gas exist. Theoretical accounts of the effect of electric fields on the lifetime of the metastable hydrogen atom are available.[5,6] Comparison with the theoretical calculations serves to test the perturbation-theory treatment of the quenching of the metastable atom by electric fields, including departures from the case of very weak fields. For the proper interpretation of the data collected on the rate of production of metastable atoms in the experiments reported here, it was essential to know the lifetime of the metastable atom at suitable field strengths.

APPARATUS

A hot-filament, magnetically collimated ion source of the type described by Abele and Meckbach[7] provided protons which were accelerated to various energies between 5 and 20 keV. After magnetic selection, the protons passed through several collimating apertures and a differentially pumped collision cell as indicated in the schematic diagram in Fig. 1. The collision cell is 10.16 cm long and has beam entrance and exit apertures A_2 and A_3 which are 1 mm in diameter. Apertures A_1 and A_4 determine the amount of angular spread in the 1-mm-diam beam which finally enters the observation region. Two differential pumping chambers are defined by A_1 and A_4, which have the same design as A_2 and

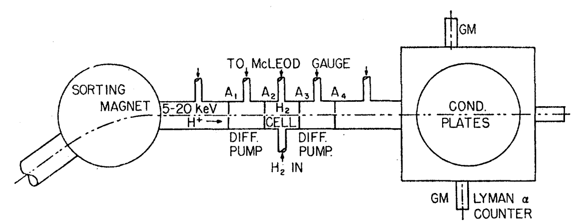

FIG. 1. Schematic diagram of the apparatus.

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1 L. Madansky and G. E. Owen, Phys. Rev. Letters **2**, 209 (1959).

2 L. Colli, F. Cristofori, G. Frigerio, and P. Sona, Phys. Letters 3, 62 (1962).

3 D. R. Bates, *Atomic and Molecular Processes* (Academic Press Inc., New York, 1962), Chap. 14.

4 W. L. Fite, R. T. Brackmann, D. G. Hummer, and R. F. Stebbings, Phys. Rev. **116**, 363 (1959).

5 H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic Press Inc., New York, 1957), Sec. 67.

6 W. E. Lamb, Jr., and R. C. Retherford, Phys. Rev. **79**, 571 (1950).

7 M. Abele and W. Meckbach, Rev. Sci. Instr. **30**, 335 (1959).

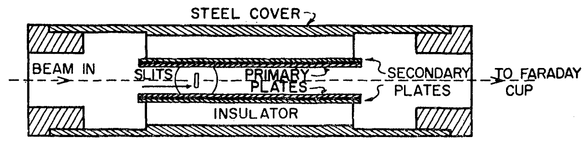

FIG. 2. Cross section of the observation chamber, showing the slits used to mask the Lyman alpha counters.

A_3 and are spaced 6.35 cm along the axis from them. Provision was made for pressure measurements inside and outside the gas cell by means of a McLeod gauge.

A cross section of the chamber used for observing the metastable component of the neutral beam and the quenching effect of an electric field on the metastable component appears in Fig. 2. The beam trajectory lies in the midplane of a pair of condenser plates which are symmetrically located with respect to the incoming beam and which provide an electric field which quenches the metastable component of the beam. The perturbing electric field mixes the $2s_{1/2}$ state with the $2p_{1/2}$ state which then radiates Lyman alpha photons in a further transition to the $1s$ state.[5,6] Observation of the number of Lyman alpha photons emerging per unit time from some section of the beam between the plates into detectors located at either side gives the relative population of metastable atoms in that section of the beam. The detection process is described below.

The polished brass condenser plates are 10.16 cm in diameter, 0.159 cm in thickness, and spaced 1.27 cm apart. Figure 2 shows an additional pair of plates spaced 0.076 cm from the pair which provides the internal field. Electrolytic-tank studies showed that when these secondary plates were maintained at equal but opposite potentials to the primary plates, the external fields tending to quench the metastable atoms before entering the internal field region were much reduced. At a point on the beam trajectory 3 mm upstream from the boundary of the internal-field region, for example, the ratio of measured fringing field with the secondary plates in place to measured fringing field when they were removed was about 15%. Similar precautions were taken against large stray magnetic fields by providing the entire observation chamber with steel covers. The motional electric field seen by metastable atoms moving at 10^8 cm/sec in a transverse field of 1 G is about 1 V/cm. Measurements of the magnetic field inside the observation chamber made with all magnets in use gave less than 1 G.

Detection of the metastables was accomplished by counting the Lyman alpha photons emerging from 3-mm sections of the beam into a solid angle of 1.53×10^{-3} sr perpendicular to the beam direction and in the midplane of the condenser plates. This solid angle was determined by a pair of rectangular brass slits placed in front of each detector and oriented vertically with respect to the beam direction, as in Fig. 2. All observations were made on sections of the beam at least 2.5 cm from any plate boundary to avoid any possible influence of the fringing field region on the measurements. Two detectors, mounted as in Fig. 1, were used to view the Lyman alpha photons emitted into the direction and solid angle indicated. The number of Lyman alpha photons emerging from a selected 3-mm section of the beam depended on the beam energy, the beam current passing through the apparatus, the pressure in the gas cell, the voltage between the primary condenser plates, and the linear distance of the section being observed from the beam's point of entrance into appreciable electric field. Both detectors were movable parallel to the beam direction during experimental runs. In this way the variation of metastable population in the beam with the distance traveled downstream from some reference point in the uniform field region could be obtained. It was convenient to keep one counter stationary viewing the reference section while moving the other counter downstream in intervals up to a total displacement of 5 cm. The ratio of the counting rates N_1/N_2, after making the background corrections to be discussed, was then independent of any assumption about constant beam current. As an independent check, a Faraday cup which collected the entire observed beam and a beam integration circuit were used to establish the proportionality of counter output to beam current for given experimental conditions.

The counters used were Geiger tubes filled with iodine vapor and helium, similar to those described by Brackmann *et al.*[8] Normally incident Lyman alpha photons were transmitted through lithium fluoride entrance windows into the counter interiors, where photoionization of the iodine, followed by electron multiplication and quenching, resulted in countable pulses from the charge collected at the center wires of the tubes. The counter efficiencies are on the order of 1% per photon for normally incident Lyman alpha photons. The efficiency of one of the counters used was measured under typical operating conditions by a method to be described and came out to be 1.2%. The spectral windows of such counters lie approximately between 1050 and 1300 Å. Photons of shorter wavelength are strongly absorbed by the window, while those with longer wavelengths cannot ionize. The dead times of the counters were large, ranging from 0.3 to 1.5 msec. Counting rates of several hundred photons per minute were used in these experiments with proton beam currents at the Faraday cup on the order of 1 mμA to avoid a large dead-time correction.

An end window geometry very similar to that of Brackmann *et al.* was used.[8,9] The stainless steel counter shells were typically 2.5 cm in diameter and a few centimeters in length. Into a retaining well at one end a lithium fluoride window was fastened with epoxy

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

[9] The first of two similar designs used was provided by Professor Bailey Donnally of Lake Forest College, Lake Forest, Illinois, and the second by Dr. C. Y. Fan and the author at the University of Chicago.

cement. Into the other end a Pyrex filling tube and a glass seal containing the 1-mm-diam polished center wire were sealed. The tip of the center wire typically terminated about 3 mm from the inner surface of the crystal window, as in Fig. 1. The length of center wire exposed to the counter interior was typically about 3.2 cm.

The sloping Geiger plateaus of these counters usually lay in the vicinity of 700 to 800 V. One counter was successfully operated over a plateau region of about 100 V, although most show narrower plateaus. The efficiency rose by roughly a factor of 2 over the plateau region. Moreover, the efficiencies of the counters used varied strongly with temperature, in a very similar way to that described by Brackmann *et al.*[8]

The counters were operated with the shell grounded and the center wire biased with batteries at some positive operating voltage in the plateau region. The outputs were then coupled to amplifiers, discriminators, and scalers. The large pulse amplitudes allowed very efficient discrimination against electronic noise. There was some variation in pulse height with counting rate during experimental runs, but never enough to require discriminator adjustments. No deterioration of counter characteristics occurred during these experiments. The shelf life appears to be at least several months long.

EXPERIMENTAL

A. Production and Equilibration of the Metastable Beam Component

The equilibration of the metastable component of the beam was studied by observing the relative intensity of the Lyman alpha line as various amounts of dried hydrogen gas were admitted to the gas cell. Pressures in the gas cell ranged from about 0.2 μ to 35 μ as measured by a McLeod gauge. Pressures on the downstream side of the cell were commonly about 10^{-3} μ, and on the upstream side 1 or 2×10^{-2} μ. Intensity measurements of the Lyman alpha line were made at all the various pressures for fixed placement of the two counters, a beam energy of 15 keV, an electric field between the primary plates of 275 V/cm, and fixed amounts of charge accumulated at the Faraday cup. The constancy of the ratio of the counting rates of the two detectors was a suitable reliability check. Because the counts accumulated at each pressure were normalized to unit charge deposited at the Faraday cup rather than to unit flux of particles, it was necessary to account separately for the total neutral component of the beam. Measurements of the percentage neutralization of the incoming proton beam for given pressure in the gas cell at 15-keV projectile energy were consistent with the neutral growth curve for 15-keV protons in hydrogen in a review article by Allison.[10] It was then possible to normalize the Lyman alpha intensity observed at each pressure to unit particle flux reaching the Faraday cup. The basic observations at each pressure were thus the number of Lyman alpha photons seen by each counter per unit particle flux at the Faraday cup. Small corrections to the pressure readings because of residual gases outside the gas cell were necessary. These corrections were a source of error at the smallest pressures in the gas cell, as were the readings of the McLeod gauge itself at pressures below 1 μ, where the gauge accuracy falls off. During experimental runs, repeated measurements at the lowest pressures were periodically made to test for any systematic changes.

Inherent in every experimental run were background counts from two sources. Cosmic ray background was measured at the beginning and end of each run. There was also ultraviolet radiation background from excitation of the residual gas in the observation chamber by the total incoming beam, which contained mostly protons and hydrogen atoms in the 1*s* state in addition to the small metastable component being studied. Because of the drift section in the apparatus between the gas cell and the detection chamber in Fig. 1, all the likely optical transitions associated with collision processes in the gas cell had long since died away. With pressures in the observation region at about 10^{-6} mm and the 200-Å windows of the counters, this background was never troublesome. It was separately determined by removing the electric field between the condenser plates, which effectively eliminated any contribution to the total signal from the now unquenched metastable component of the beam. Moreover, this radiative background was found to be substantially independent of the positions of the counters, indicating that the total beam passed well within the view of the two counters. One might surmise that the deviation of the proton component of the beam occurring when the electric field was switched on might change this background. Rapid switching of the field from one polarity to the other showed no systematic variation of the counting rate in either counter. This constancy of radiative and cosmic ray background was fairly unimportant anyway, since the background seldom exceeded 10% of the total number of counts, except during experiments with the largest quenching fields where the number of metastable atoms was severely attenuated before viewing by the counters, and during experiments with the smallest quenching fields, where the quenching rate was correspondingly small. For the most extreme of these cases, the background was sometimes as high as 30%. Dead-time corrections were a necessity at all but the lowest counting rates.

B. Quenching of the Metastable Component in dc Electric Fields

The rate of quenching of the metastable atoms as they traveled through dc electric fields between 50 and 500 V/cm was determined by observing the decay length of

[10] S. K. Allison, Rev. Mod. Phys. 30, 1137 (1958).

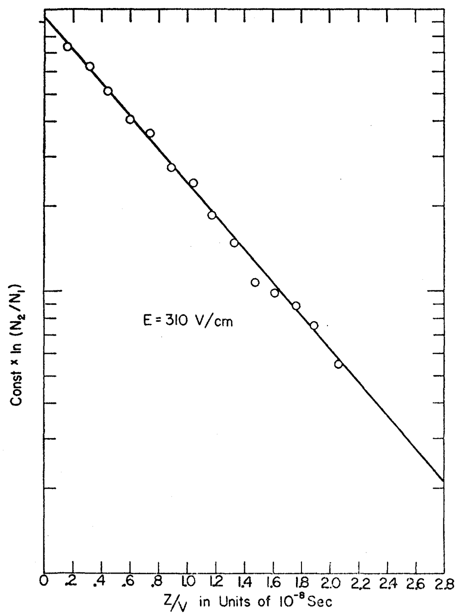

FIG. 3. Typical semilogarithmic plot of the counting rate ratio N_1/N_2 versus z/v, for E equal to 310 V/cm.

the Lyman alpha radiation entering the two counters. If z is the linear separation of the centers of the 3-mm sections being viewed by the two counters, then after corrections for different efficiencies of the two counters and total background as above, the ratio of the counting rates of the two counters N_1/N_2 is $\exp[-z/v\tau(E)]$, where $\tau(E)$ is the lifetime of the metastable atom in field E, and v is the velocity. For a given field strength and beam energy, comparison of the counting rates of the two detectors as they moved apart in intervals up to 5 cm total displacement gave a decay length for the quenching radiation. The procedure followed during each run at a given field strength involved: (1) aligning the counters so as to view the same section of the beam, at a minimum of 2.5 cm from any plate boundary; (2) obtaining the relative efficiency of the two counters from their counting rates at this position; (3) moving one counter downstream with respect to the other in intervals of 0.254 cm (0.1 in.) and measuring the two counting rates at each separation; (4) moving the same counter back upstream, again in intervals of 0.254 cm, but taking the counting rates at separations midway between the downstream separations; and (5) making a semilogarithmic plot of the corrected relative counting rates N_1/N_2 of the two counters at each point versus z/v. A typical plot from an experimental run is shown in Fig. 3, for the case $E=310$ V/cm. The inverse slope of a straight line statistically fitted to this plot gave, except for sign, the lifetime of the quenching radiation and thus of the perturbed metastable atom. The beam speed was determined from measurements of the accelerating voltage with a calibrated milliammeter and standard resistor. The field E was provided by batteries and measured with a calibrated electrostatic voltmeter. Errors of a few volts per cm in E were anticipated as a result of the motional electric field seen by the metastable atoms traveling through the inhomogeneous stray magnetic fields provided by magnets in the apparatus, meter accuracy, contact potential differences, and possibly surface charges accumulated on the condenser plates. This error was expected to cause greater error in the lifetime determinations than other systematic errors, the largest of which was no doubt the uncertainty in v. This uncertainty arose chiefly from instability in the proton accelerating voltage, causing an uncertainty of about 2% in $\tau(E)$ in the worst cases.

C. Measurement of the Effective Cross Section for Capture into the 2s State Including Cascade Contributions

The efficiency of one of the counters was measured by using it to view the radiation from the process $e+H_2 \rightarrow$ countable ultraviolet radiation, as a 150-eV electron beam passed through a hydrogen gas target.[11] The interaction region was viewed once again at 90 degrees to the beam direction, and care was taken to preserve the same geometry for the efficiency measurement as was used in the observations on metastable hydrogen atoms. This method was used by Dunn *et al.*[12] to measure the efficiency of such counters. The counter efficiency was then determined using the cross section published by these authors for the process $e+H_2 \rightarrow$ countable uv at the 150-eV beam energy. It should be emphasized that these cross sections were originally determined in the relative sense by Fite and Brackmann.[13] Measurements of the excitation of Lyman alpha radiation by electrons in atomic hydrogen were normalized to a Born approximation calculation at high energies. Then experimental comparison of the atomic and molecular excitation cross sections gave cross sections for $e+H_2 \rightarrow$ countable uv. Strictly speaking the efficiency of the counter in our experiments is expressed only in terms of the cross section for the process $e+H_2 \rightarrow$ countable ultraviolet radiation. The counter efficiency was thus about $0.012\times(1.2\times10^{-17}$ $\text{cm}^2/\sigma_{el})$, where σ_{el} is the correct excitation cross section. If the quoted value of σ_{el} of 1.2×10^{-17} cm^2 is correct, then the counter efficiency is about $1.2\%\pm0.4\%$. The uncertainty arises from the variation of the counter efficiency with room temperature, and from the possibility that the ion-gauge readings of the hydrogen pressure during the efficiency measurement were in error. The ion gauge used was a type RG-75 Veeco gauge; the sensitivity correction factor for measure-

[11] All of the experimental equipment for the efficiency measurement as well as much personal assistance was provided by Professor Bailey Donnally.

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

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

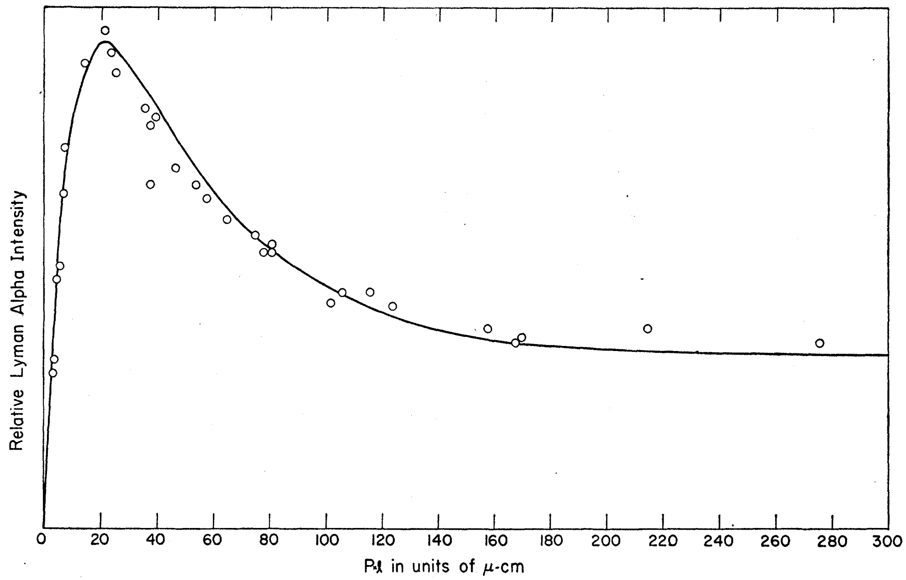

FIG. 4. Experimental and theoretical curves for the growth and equilibration of the metastable beam component as a function of the number of target atoms per cm², in units of pressure in microns times length in cm.

ments on hydrogen rather than on nitrogen was taken to give a pressure 2.39 times the gauge reading.

It should be noted that no filter was used to remove wavelengths other than Lyman alpha. Recent unpublished measurements by Donnally[11] indicate that about 65% of the countable radiation in the detector calibration is of molecular origin when no filter is used.

Once the efficiency of the counter had been established, it was possible to calculate the total number of metastable atoms in the total beam at a given counter position for the given experimental conditions already discussed, assuming the isotropic distribution of quenching radiation predicted by Lichten.[14] Using the experimental determination of the lifetime of the metastable atoms for the field strength being used of 275 V/cm, it was possible to determine the number of metastables entering the field region per unit time. Values for the counting rate, beam current, geometrical factor, counter efficiency, and pressure in the gas cell were then used to calculate $\sigma(1,2s)$, the effective cross section for the capture of an electron into the 2*s* state, including cascade contributions, by an incident proton in hydrogen gas, at 15 keV projectile energy. All measurements were made at pressures low enough to exclude secondary collisions, in the linear region of the production and extinction curve of the metastable component of the beam shown in Fig. 4.

RESULTS

A. Appearance of a Maximum in the Production and Extinction of the Metastable Beam Component

Figure 4 depicts the variation in intensity of the Lyman alpha line from the quenching of the metastable component versus the amount of hydrogen gas traversed in the gas cell, in units of pressure times length. There is a sharp peak in the production of metastable atoms between 17 and 23 μ-cm, corresponding to a hydrogen atom density between 1.0 and 1.5×10^{15} atoms/cm², this density being given by the molecular number density times the cell length times two. Superposed on the experimental points is a production and extinction curve, to be discussed below, obtained by considering the equilibration of a three-component ion beam composed of protons, hydrogen atoms in the 1*s* state, and hydrogen atoms in the metastable 2*s* state. There is first the expected fairly linear rise in the Lyman alpha quenching signal with pressure as the number of metastable producing collisions increases. Then, because of an apparently very large destruction cross section for the metastable component and neutralization of the proton component of the beam, the metastable component declines and then equilibrates. The observations of Colli *et al.*[2] do not show any such maximum.

B. Quenching by Electric Fields between 50 and 500 V/cm

Figure 5 shows experimentally measured values of the metastable lifetime versus field strength, obtained from the semilogarithmic plots of Lyman alpha intensity versus z/v at each field strength E. A straight line was fitted to each plot by the method of least squares.[15] The error bars appearing at some of the experimental points were deduced directly from the probable errors of the slopes determined in the linear least-squares fitting procedure. At points where no such error bars appear, this statistical error was too small to display, typically on the order of 2%.

[14] W. Lichten, Phys. Rev. Letters 6, 12 (1961).

[15] H. Margenau and G. M. Murphy, *The Mathematics of Physics and Chemistry* (D. Van Nostrand Co., Inc., New York, 1957), p. 518.

A weak-field time-independent perturbation-theory calculation of the effect of dc electric fields on the lifetime of the metastable hydrogen atom has been given by Bethe and Salpeter.[5] Evaluation of their formulas at the electric fields used in these experiments gives the dashed curve plotted in Fig. 5. The lower curve comes from the perturbation theory calculation for very weak fields,[5] as described in Fig. 5.

C. Effective Cross Section for Capture into the 2s State

Six determinations of $\sigma(1,2s)$ were made at pressures in the linear region of Fig. 4, all giving about 1×10^{-17} $\text{cm}^2\times(\sigma_{el}/1.2\times10^{-17}\ \text{cm}^2)$, where σ_{el} is expected to be about 1.2×10^{-17} cm^2. Because of the uncertainties in the counter efficiency already discussed, an error of 30% in the value of $\sigma(1,2s)$ relative to σ_{el} may be present. Unfortunately, an erroneous value of the capture cross section of 5×10^{-17} cm^2 per atom was quoted in Phys. Rev. Letters **13**, A2 (1964).

Once this capture cross section is specified, Fig. 4 yields a value for the effective cross section for excitation of 1s projectiles to the 2s state of 1.5×10^{-18} cm^2 per target atom.

DISCUSSION

A. Significance of a Maximum in the Growth of the Metastable Beam Component

The solutions to the differential equations governing the equilibration of an initially pure ion beam among three possible charge components have been discussed by Allison.[10] In Sec. III, Part A, of this review article, the significance of a maximum in the growth of a charge component is discussed. Six effective cross sections $\sigma(i,f)$ are needed to define the transition rates among the three components, each of which takes into account the multiple processes which may lead from the initial charge state i to the final charge state f. In our case two of the states, the metastable 2s state and the 1s state, are electrically neutral, but the same differential equations apply to a three-component system consisting of protons, 1s atoms, and 2s atoms, provided the appropriate effective cross sections are used. The crudest assumption in this interpretation of the experimental production and extinction curve in Fig. 4 is that only three components essentially govern the relative metastable population in the beam. A small H^- component that is known to appear[10] is, for example, neglected. Other excited states might compete with the population of the metastable state. Population of excited states other than the 2s state in a collision inside the cell usually results in a rapid decay to the 1s state, or occasionally to the 2s state; but in either case, after a time short compared to that of a subsequent collision and certainly to that at which the beam is first observed, the usual result is production of one of the three

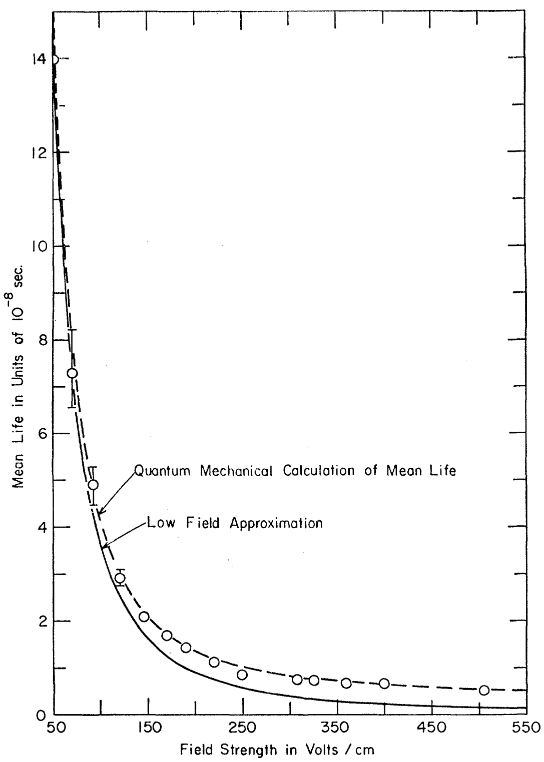

FIG. 5. Experimental and theoretical determinations of the lifetimes of metastable hydrogen atoms in dc electric fields. The dashed curve is drawn from Eq. (10). The solid curve corresponds to the first term in the expansion of Eq. (10), for $\delta\ll1$.

components being considered. We assume that population of excited states, having lifetimes such that further collisions may occur before radiative decay to the 1s state or the 2s state, occurs so rarely as not to have any effect on the interpretation of Fig. 4. A three-component system thus appears to be the first reasonable approximation in considering the production and extinction of the metastable component in this experimental situation and indeed will be found to give a reasonable interpretation of Fig. 4.

The six effective cross sections describing the equilibration process will be labeled $\sigma(1,1s)$, $\sigma(1,2s)$, $\sigma(1s,1)$, $\sigma(1s,2s)$, $\sigma(2s,1)$, and $\sigma(2s,1s)$. $\sigma(i,f)$ refers to the effective cross section per target atom for those collisions in which the projectile changes from state i to state f. $\sigma(2s,1)$, for example, refers to the net cross section per target atom for ionization of a metastable projectile in collision with a target molecule. Once values are specified for each of these cross sections, formula II-12 of the article by Allison[10] may be evaluated for the fraction F_{2s} of the total number of projectiles to be found in the metastable state as a function of the number of atoms of target gas per square centimeter which the initially pure proton beam has traversed:

$$F_{2s}=F_i=F_{i\infty}+[Pe^{\pi q}+Ne^{-\pi q}]\exp-\tfrac{1}{2}\pi\sum\sigma(i,f). \quad (1)$$

F_i is the fraction of the total beam to be found in the state i after traversal of π atoms/cm² of target gas. $\sum \sigma(i,f)$ is the sum of the six cross sections. $F_{i\infty}$, P, N, and q are all functions of the six cross sections; to describe their dependence it is convenient to use a notation similar to Allison's[10]:

$$\alpha \equiv \sigma(1s,2s)+\sigma(1s,1),$$
$$\beta \equiv \sigma_e \equiv \sigma(2s,1s)+\sigma(2s,1), \quad (2)$$
$$\gamma \equiv \sigma(1,1s)+\sigma(1,2s).$$

$$a \equiv -[\beta+\sigma(1,2s)], \qquad f \equiv [\sigma(2s,1s)-\sigma(1,1s)],$$
$$b \equiv [\sigma(1s,2s)-\sigma(1,2s)], \quad g \equiv -[\alpha+\sigma(1,1s)]. \quad (3)$$

$$q=\tfrac{1}{2}[(g-a)^2+4bf]^{1/2}. \quad (4)$$

$$\sum \sigma(i,f) = -(a+g). \quad (5)$$

$$D=(ag-bf). \quad (6)$$

$$s \equiv \tfrac{1}{2}(g-a). \quad (7)$$

Using the symbols thus defined,

$$F_{2s\infty}=[\alpha\sigma(1,2s)+\sigma(1s,2s)\sigma(1,1s)]/D,$$
$$F_{1s\infty}=[\beta\sigma(1,1s)+\sigma(2s,1s)\sigma(1,2s)]/D, \quad (8)$$

$$P=(1/2q)[F_{2s\infty}(s-q)-bF_{1s\infty}],$$
$$N=-(1/2q)[F_{2s\infty}(s+q)-bF_{1s\infty}]. \quad (9)$$

The term in brackets in Eq. (1) is an increasing function of π and all the damping of the production peak comes from $\exp[-\frac{1}{2}\pi \sum \sigma(i,f)]$.

Various rough but reasonable estimates and calculations can be invoked to approximately specify $\sigma(1,1s)$, $\sigma(1,2s)$, $\sigma(1s,1)$, and $\sigma(1s,2s)$.

Fortunately it is $\sigma(2s,1)$ and $\sigma(2s,1s)$, the two cross sections which describe the extinction of the 2s state, which largely determine the position of the maximum of the peak in Fig. 4, and to a somewhat lesser extent, the height of the peak referred to the equilibrium fraction. An illustration of this statement will be found in Fig. 6 after specification of the necessary cross sections. Why this is the case may be seen in the exponential damping terms in Eq. (1). No estimates of cross sections other than $\sigma(2s,1)$ and $\sigma(2s,1s)$ will be anywhere near large enough to account for the observed experimental damping in Fig. 4, and these two cross sections dominate the damping to such an extent that the location of the peak in Fig. 4 yields considerable information about the size of the cross section for the collisional destruction of the metastable atoms.

Allison[10] quotes experimental values for the total electron capture cross section for 15-keV protons in hydrogen gas at about 35×10^{-17} cm²/atom, which includes, of course, the very dominant contribution from[3] $\sigma(1,1s)$; this cross section should then have a value on the order of 30×10^{-17} cm²/atom.

Our measured value of $\sigma(1,2s)$ was about 1×10^{-17} cm²/target atom.

$\sigma(1s,1)$ should be nearly equal to the ionization cross section for the composite neutral beam, since the vast majority of atoms in the neutral beam is expected to be in the 1s state. Values for the ionization cross section for 15-keV atoms in hydrogen gas are quoted by Allison.[10] On this basis $\sigma(1s,1)$ should be about 5×10^{-17} cm²/atom.

$\sigma(1s,2s)$ has been calculated by Bates and Griffing[16] for protons in atomic hydrogen, and at 15 keV their calculations give about 0.5×10^{-17} cm²/atom. We shall use the estimate obtained from Fig. 4 of 0.15×10^{-17} cm² per atom, which will be discussed later in this section.

The estimates of four of the six cross sections describing the equilibration of the metastable component of the beam, in units of 10^{-17} cm²/target atom, thus become $\sigma(1,1s)=30$, $\sigma(1,2s)=1$, $\sigma(1s,1)=5$, and $\sigma(1s,2s)=0.15$.

Unfortunately, the production and extinction curve is very insensitive to the partition of σ_e between $\sigma(2s,1)$ and $\sigma(2s,1s)$. It is not possible to decide from the curve in Fig. 4 whether the destruction of the metastable atom comes about predominantly by ionization of the metastable atom, or by de-excitation to the ground state. It might be expected that the 2s level is much less stable against ionization than the 1s level. Some evidence based on measurements of the stopping power of hydrogen gas for neutral hydrogen atoms indicates that probably more than 90% of the stopping losses occur through ionization rather than through energy loss processes involving discrete atomic states of the projectile, at projectile energies on the order of 50 keV.[17] On the other hand, the small separation of the $2s_{1/2}$ and $2p_{1/2}$ states makes a de-excitation to the ground state through collisional mixing of these two states extremely easy. Fite *et al.*[4] have emphasized the importance of this process in collisional destruction of the metastable atoms at thermal energies. For our energy of 15 keV, however, it is easy to mix the 2s state with any number of discrete states, so that the nearby 2p state may lose its special significance.

The upshot is that a reasonable fit to the experimental equilibration curve in Fig. 4 can be obtained by taking the total extinction cross section σ_e for metastable atoms of 15 keV energy in hydrogen gas to be on the order of 200×10^{-17} cm²/target atom. The theoretical curve in Fig. 4 is constructed for $\sigma_e=200\times10^{-17}$ cm²/target atom. That the partition between ionization and de-excitation makes little difference can be seen in the exponential damping term in Eq. (1), since $\sigma(2s,1)$ and $\sigma(2s,1s)$ enter as a sum. In addition, q depends only weakly on the partition. The differences between various partitions are so small that they could not conveniently be displayed in either Fig. 4 or Fig. 6.

[16] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **66**, 961 (1953).

[17] J. Cuevas, M. Garcia-Munoz, P. Torres, and S. K. Allison, Phys. Rev. **135**, A335 (1964).

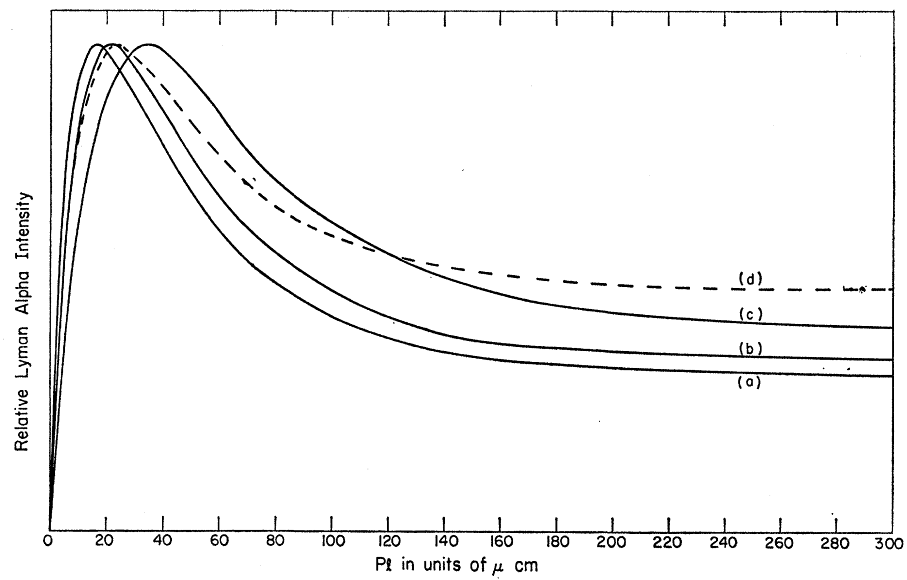

FIG. 6. Theoretical curves describing the growth and equilibration of the metastable beam component for several different values of the total extinction cross section per atom σ_e. See Table I for values of the cross sections used in constructing these curves.

To what extent σ_e is determined by the production and equilibration data of Fig. 4 may be seen in Fig. 6. Four growth and equilibration curves are plotted. Table I summarizes the various cross sections assumed in the construction of the four curves. Curve (b) is the same as the curve fitted to the experimental data in Fig. 4. Curve (c) is obviously a poorer choice to fit the experimental data, and we see that as σ_e decreases, the fit to the data gets rapidly worse.

It would be difficult to reconcile the experimental data with any value of σ_e less than 100×10^{-17} cm^2 per atom. The resulting curve would peak at too large a pressure in the target cell, and would have a ratio of peak height to F_{2s_∞} which was too small. A value for σ_e in excess of 100×10^{-17} cm^2/atom for 15-keV metastable projectiles is needed to give the production and equilibration curve a steep enough exponential tail to match the experimental data. Such a value can be compared with the value of Fite *et al.*[4] at energies on the order of 1 eV. Their value is about 350×10^{-17} cm^2/atom.

Such a large cross section for destruction of the metastable atom by ionization would appreciably alter the electron loss section for hydrogen atoms in various gases if any sizable admixture of the 2*s* state were present. Such an effect has been hypothesized by Barnett and Stier[18] in their work on electron-loss cross sections by helium neutrals in various gases. They found a dependence of electron-loss cross-section measurements on pressure in their beam neutralizer, and sought to explain it through preferential destruction of metastable helium atoms as pressure in the neutralizer was increased. The large value of σ_e deduced from Fig. 4 leads to a similar expectation in the case of electron loss by hydrogen atoms, if ionization of the metastable atoms by collision predominates over other extinction processes.

The ratio of peak height to equilibrium fraction is quite sensitive to $\sigma(1s,2s)$, reaching a value of about 5 if this cross section were zero. Curve (d) differs from curve (b) only through a change in this cross section estimate from 1.5 to 3×10^{-18} cm^2 per atom.

B. Metastable Lifetime Determinations Through Radiative Decay Length Measurements

Figure 5 shows the degree of verification of the quantum-mechanical-lifetime calculation previously referred to.[5] The calculation gives a formula for $\tau(E)$, the lifetime of the perturbed state which is metastable in the absence of the field, in terms of the lifetime of the

TABLE I. Cross sections assumed in the construction of the four growth and equilibration curves appearing in Fig. 6, in units of 10^{-17} cm^2/target atom.

Curve	$\sigma(1,1s)$	$\sigma(1,2s)$	$\sigma(1s,1)$	$\sigma(1s,2s)$	$\sigma(2s,1)$	$\sigma(2s,1s)$	σ_e
(a)	30	1	5	0.15	300	0	300
(b)	30	1	5	0.15	100	100	200
(c)	30	1	5	0.15	100	0	100
(d)	30	1	5	0.3	100	100	200

[18] C. F. Barnett and P. M. Stier, Phys. Rev. **109**, 385 (1958).

$2p$ state $\tau(2p)$ and δ:

$$\tau(E)=\tau(2p)\left\{1+\frac{\delta^2}{[1-(1+\delta^2)^{1/2}]^2}\right\},\quad \delta=\frac{2\sqrt{3}Eea_0}{L}. \tag{10}$$

L is the Lamb shift and a_0 the Bohr radius. Equation (10) is valid as long as mixing of $p_{3/2}$ states is negligible, say for $\delta<8$.

In addition to illustrating a convenient technique for making reasonably accurate lifetime measurements, the measurements verify the first-order perturbation treatment of a metastable hydrogen atom in dc electric fields. The measurements do not test the several finer approximations involved.[5,6] All effects due to hyperfine structure are ignored. It is assumed that there is no mixing of levels of different j, which implies field strengths much less than 3000 V/cm. Lamb's time-dependent[6] treatment takes explicit account of the radiative width of the $2p$ level, which the time-independent treatment of Bethe and Salpeter[5] does not. Four different decay probabilities appear in Lamb's treatment, only one of which enters in a material way in these experiments. Lamb's sudden initial conditions were not applicable, since the entrance of the metastable atoms into the field was fairly adiabatic. All of these differences were too small to observe by the experimental techniques used. Measurements were never made on the beam until times long compared to the lifetime of tne $2p$ state had passed. The fringing field had no transient effect, then, at the times of measurement. The time-independent perturbation treatment was thus quite adequate to explain the measurements. The confirmation of its predictions illustrates the utility of this kind of lifetime determination when accuracies on the order of 1% are considered sufficient.

C. Effective Cross Section for Capture into the 2s State

Comparison of the value of $\sigma(1,2s)$ obtained in these experiments can be made with the Born approximation calculations by Bates and Dalgarno[19] and with the most recent results of Lovell and McElroy[20] on the cross section for capture into the $2s$ state for protons in atomic hydrogen. At 15 keV the Born approximation result is about 4×10^{-17} cm^2/atom. The result of Lovell and McElroy, who used an expansion in atomic eigenfunctions and took account of coupling between the $1s$ and $2s$ state, is about 1.5×10^{-17} cm^2/atom.

Comparison of experimental results can be made with the work of Colli *et al.*[2] Their result at 15 keV for the case of molecular hydrogen indicated a value between 1 and 2×10^{-17} cm^2/atom. This experimental value is a relative one, however, normalized to the same Born approximation calculation of Bates and Dalgarno at 40 keV. The values for $\sigma(1,2s)$ quoted by this group[2] agree in behavior with the Born approximation calculations for energies above 23 keV, but the more recent absolute values emerging from their absolute calibration scheme differ from these calculations by about a factor of 50.[21] Our result is in agreement with their relative value at 15 keV. It is certainly not possible to reach agreement with the absolute value quoted, the difference being on the order of a factor of 50.

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[20] M. B. McElroy (private communication).

[21] F. Cristofori, G. Frigerio, N. Molho, and P. Sona, *Proceedings of the Sixth International Conference of Ionization Phenomena in Gases* (Paris, 1963) Vol. 1, p. 69–71.

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