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Radiative Mean-Life Measurements of Some Atomic-Hydrogen Excited States Using Beam-Foil Excitation*

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The beam-foil excitation method has been used to measure the radiative mean lives of the $2p$ and $3p$ states of atomic hydrogen. The results obtained were $(1.60 \pm 0.01) \times 10^{-9}$ sec and $(5.5 \pm 0.2) \times 10^{-9}$ sec, respectively. A study of the results of this experiment has led to the following conclusions: (a) the Lyman-α decay curves are best fitted by the sum of two exponential terms whose characteristic decay times correspond to spontaneous decay of the $2p$ state and repopulation of this state by higher states, (b) the shapes of the Lyman-α decay curves vary with foil thickness, and (c) the ion energy loss in the foils used in this experiment changes with the age of the foil. A review of previous measurements on atomic-hydrogen radiative mean lives is also made and compared with theory.

I. INTRODUCTION

A method of determining the radiative mean lives of excited states of atoms and ions of astrophysical interest has recently been suggested.[1] This technique, which has become known as the beam-foil excitation method, is actually a modern refinement of a method first used by Wien.[2,3] In the present

TABLE I. Measurements of mean lives of some excited states of hydrogen (10^{-9} sec).

Reference	$2p$	$n=2$	$3s$	$3p$	$3d$	$n=3$	$4s$	$4p$	$4d$	$4f$	$n=4$	$n=5$
Ref. 6						12.5 ± 1.0					34.0 ± 1.5	78 ±15
Ref. 8			135 ± 14		16.1 ± 0.6		186 ± 27	14.6 ± 2.5	37.7 ± 5.5			
Refs. 10, 11			160	5.4	15.6		230					
Ref. 9	1.600 ± 0.004			5.58 ± 0.13								
This paper	1.60 ± 0.01			5.5 ± 0.2								
Theory, Ref. 4	1.6	2.1	160	5.4	15.6	10.2	230	12.4	36.5	73	33.5	88
Theory[a]	1.596	2.128	158.4	5.273	15.47	10.02	226.6	12.36	36.16	72.56	33.13	85.54

[a]See Ref. 5; obtained by summing referenced values of the transition probabilities over the final states and inverting.

work, a study has been made of the radiative decay of some excited states of atomic hydrogen using this technique. Exact agreement between experiment and quantum-mechanical calculations of radiative mean lives of hydrogen would give confidence in knowing to what extent unknown mean lives can be reliably measured. This program also provides an opportunity for a review of the status of recent experiments on atomic-hydrogen mean lives as well as to point out some basic limitations of the beam-foil method.

There have been surprisingly few measurements of the radiative mean lives of the excited states of atomic hydrogen. Up to 1965 there does not appear to be any accurate direct measurements of hydrogen excited-state radiative mean lives. All recent significant results known to us are tabulated in Table I. The Table also shows two sets of theoretical values[4,5] for all hydrogen-state mean lives through principal quantum number $n = 5$ for comparison with one another and with all experimental values shown.

Ankudinov and collaborators[6] observed the build-up of light intensity in a gas chamber as H_2^+ ions passed through it. It was assumed in their analysis of results that repopulation of the state of interest by cascade transitions was negligible and that there were no secondary collisions that might cause nonradiative decay. The average mean lives of the $n = 3,4$, and 5 levels of atomic hydrogen were measured by this method and the results are shown in Table I. The use of photometric detection techniques in this experiment enabled these workers to obtain the first accurate values for radiative mean lives in hydrogen. The mean lives of the various fine-structure states contributing to Balmer radiation were not measurable due to mixing of such states by the presence of a strong magnetic field.

The beam-foil method, first introduced by Kay[7] and developed by Bashkin,[1] was used by Goodman and Donahue[8] to study the spectrum of atomic hydrogen. The excited beam was obtained by passing H_3^+ ions through thin carbon foils. The decay in intensity of the emitted H_α and H_β radiation as a function of distance along the beam was measured using interference filters placed in front of photomultipliers. Analysis of the decay curves gave the radiative mean lives of the $3s,3d,4s,4p$, and $4d$ states shown in Table I. Cascading from higher states may have been a factor in these results; however, it was not considered. It was also found that the orbital-angular-momentum states within an n level were not populated according to their statistical weights in the excitation process. Lower orbital-angular-momentum states appeared to be preferentially populated.

Bickel and Goodman[9] studied radiation from the Lyman series of atomic hydrogen using essentially the same experimental system as that of Goodman and Donahue. A vacuum-ultraviolet monochromator was used to record the intensity of the Lyman α and Lyman-β radiation from the excited beam. The errors shown on the results given in Table I were determined by curve-fitting analysis alone and are probably optimistic. In the case of the $2p$ state, these authors note the effect of cascades by observing that the Lyman α decay curve is best fitted by one exponential plus a constant (the cascade term). They also find that the $3p$ state mean life is about 3% larger than the accepted theoretical value and attribute the difference to the effect of cascades.

Hughes *et al.*[10,11] used a technique similar to the beam-foil method except that a gas target served as the excitation medium instead of a foil. The radiative mean lives of the $3s,3p,3d$, and $4s$ states of atomic hydrogen were measured. The effect of cascading has been neglected in the analysis of the results. The results quoted in Table I are in precise agreement with theory[4]; however, no experimental errors are given in the papers, thus it is difficult to assess an actual comparison with theory.

The present paper describes results obtained for

the radiative mean lives of the $2p$ and $3p$ states of atomic hydrogen using the beam-foil technique.

II. EXPERIMENTAL METHOD

The experimental system used to obtain and analyze the foil excited beam is shown in Fig. 1. A 4–5-μA beam of 340.5-keV H_2^+ ions from the University of New Hampshire 400-kV Van de Graaff accelerator was passed through carbon foils[12] having thichnesses of 5, 20, and 40 $\mu g/cm^2$. The cross-sectional area of the excited beam was approximately 0.7 cm^2. A 2.5-mm section of this beam was viewed by a McPherson $\frac{1}{2}$-m vacuum-ultraviolet monochromator (Seya-Namioka type) set for the emission line of interest. An EMI 6256S photomultiplier, cooled to dry-ice temperature, and a sodium salycilate window were used together as the uv photon detector. A monitor photomultiplier (RCA 1P21), attached to the foil holder, viewed the beam through an H_β interference filter at a fixed distance downstream from the foil. All intensity measurements were normalized to this monitor signal. The signals from both tubes were recorded using standard pulse-counting techniques.

Intensity decay curves for a given spectral line were obtained by moving the foil upstream from the entrance slit of the monochromator in 5-mm steps. The data at each foil position were taken in the following sequence:

(1) The tube background counting rates were determined by closing the gate valve in the beam port of the accelerator with the accelerator running. These counting rates were of the order of 25 counts/sec in both tubes.

(2) The signals from the monochromator and monitor photomultipliers were counted four times consecutively, for equal periods of time.

(3) The tube background counting rates were again taken. The average background rate of the tubes was determined from Steps 1 and 3 and then subtracted from the data in Step 2. The resulting corrected monochromator counting rate was normalized to the corrected monitor photomultiplier counting rate. The background-gas contribution, although small (approximately 1 count/sec) at the chamber pressures attained (5×10^{-6} Torr), was then subtracted from the average of the four normalized counting rates. The background-gas contribution was determined (under comparable experimental conditions) by taking data in the above described manner with the foil removed from the holder. This was repeated several times and an average value was used as the background-gas contribution. Data were taken out to about thirteen $2p$ state mean lives from the point of initial excitation. The ratio of signal to background gas counting rate for the Lyman-α radiation was ≈ 700 near the foil and ≈ 8 at a distance corresponding to thirteen $2p$ state mean lives. This detailed data-taking procedure was necessitated by our observations that the RCA 1P21 monitor photomultiplier dark current increased slightly under steady irradiation from the beam.

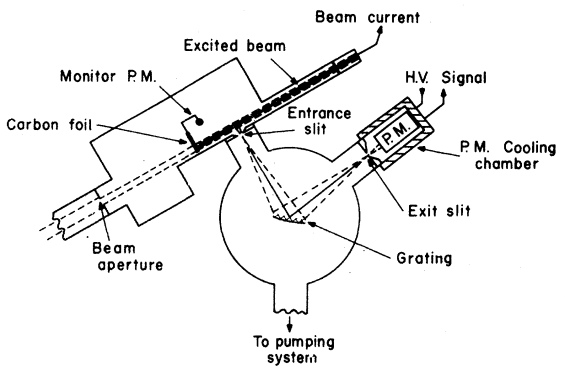

FIG. 1. The experimental arrangement is shown (not to scale). Normalization of the monochromator counting rate (EMI 6256S) is based on the counting rate of the monitor photomultiplier (RCA 1P21).

The ion energy was determined by using the 340.5-keV resonance in the F^{19} $(p, \alpha\gamma)O^{16}$ reaction to calibrate the terminal voltage of the accelerator. A beam energy of 340.5 keV was used throughout the experiment. Beam-profile measurements and the uncertainty in energy calibration resulted in a 5 keV uncertainty in the H_2^+ ion energy before it passed through the foil. The total error in the energy of the emerging atoms is due to a combination of the above mentioned error, the error in the calculated energy loss in the foil, and the error in foil thickness. The energy loss in the foil was calculated from Northcliffe's energy-loss curves.[13] It has been estimated that this value has an uncertainty of 10%[14] at the energy used in this experiment. The foil-thickness error, as quoted by the manufacturer,[12] was 60% for the 5-$\mu g/cm^2$ foils, 25% for 20-$\mu g/cm^2$ foils, and 20% for 40-$\mu g/cm^2$ foils. This uncertainty in foil thickness gives a large error in energy loss which itself is small compared to the beam energy. The net result, including the above three sources, is approximately a 1% uncertainty in the atomic-beam velocity when using either a 5 or 20-$\mu g/cm^2$ foil, and approximately 2% when using a 40-$\mu g/cm^2$ foil. The energy-loss calculations are based on the assumption that the H_2^+ ion dissociates immediately upon entering the foil and that the energy loss in the foil is that for a proton of half the energy passing through carbon.

In principle the finite velocity spread in the beam must be considered in converting the decay distance to time in the data analysis. This correction was found to be negligible compared with the previously discussed errors for all data analyzed.

III. RESULTS AND ANALYSIS

1. Lyman α Results

A typical Lyman-α decay curve is shown in Fig. 2. The slight bump in the middle of the curve appeared in all of our results at the same distance downstream from the foil. We conclude this was due to 180° reflections of the Lyman-α radiation from the beam by the H_β interference filter attached

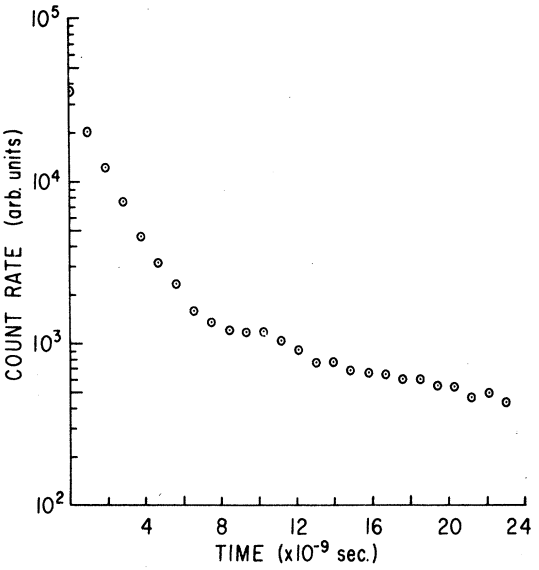

FIG. 2. A typical intensity decay curve of the Lyman α line of atomic hydrogen, obtained with 340.5-keV H_2^+ ions and a 20-$\mu g/cm^2$ carbon foil. The source of the anomaly in the curve at about 10×10^{-9} sec is discussed in the text. Counting rate errors are insignificant compared to the size of the symbols used.

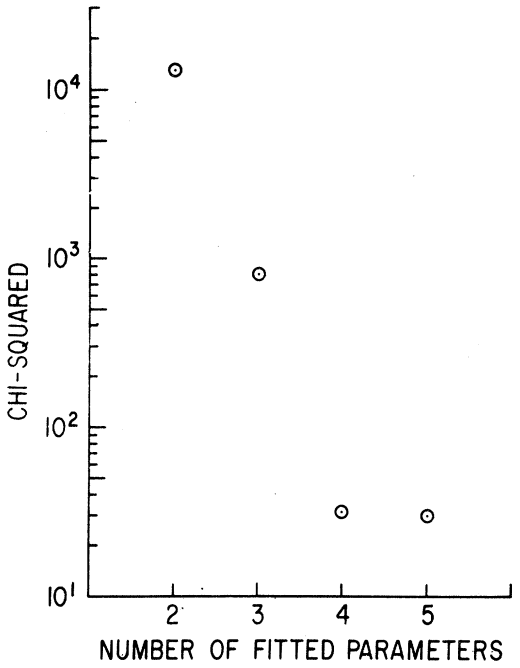

FIG. 4. Values of goodness-of-fit parameter χ^2 plotted versus the number of parameters used to fit the Lyman α data.

to the monitor photomultiplier since the signal of the monochromator photomultiplier increased as the monitor photomultiplier passed in front of the entrance slit of the monochromator. For this reason, this section of the curves was not considered during the final curve-fitting analysis. The decay curves were fitted to the sum of exponentials using a computer program called "EXFIT 2"

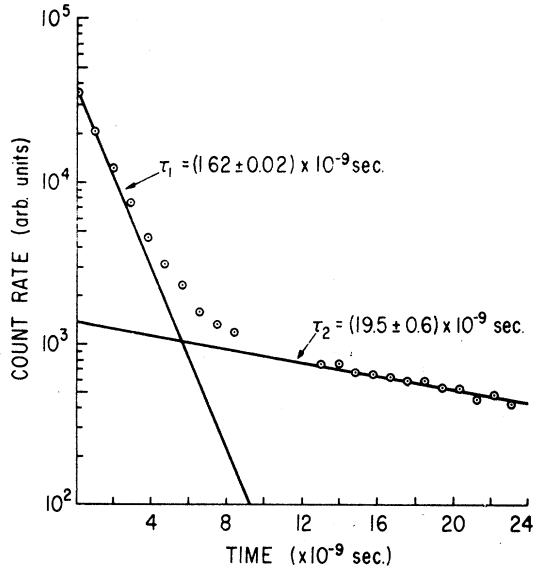

FIG. 3. Curve fitting analysis of the data in Fig. 2 is shown. The two straight lines represent the best fit to experimental data.

which is a variation of the program "AD-COS" originally developed by Grard[15] and modified by Estes.[16]

As an example, Fig. 3 shows the results from curve fitting to the data in Fig. 2, where the errors in the decay times are a combination of the curve-fitting error and the previously mentioned uncertainty in atom velocity. The decay curves were fitted to one exponential, one exponential plus a constant, two exponentials, and two exponentials plus a constant. A graph of Chi-squared plotted against the corresponding number of fitted parameters is shown in Fig. 4. Using an F-distribution table[17] in conjunction with the χ^2 test it was determined, at the 95% confidence level, that there was no significant difference between a four and five parameter fit indicating that a two exponential fit is the best fit to the Lyman-α data. However, it was also found that if the data are not taken as far downstream from the foil as was done in this experiment, the best fit to the data begins to approach the one exponential plus a constant, which is in agreement with the results of Bickel and Goodman.[9]

In Fig. 3, τ_1 and τ_2 are the characteristic decay times of the first and second exponentials which provide the best fit to the decay curve. The first exponential represents the decay of the $2p$ state to the $1s$ state and the second exponential is the result of cascading into the $2p$ state from higher states. Each cascading state in principle would contribute an additional exponential term whose decay time would be the radiative mean life of that state. The contribution of any one particular repopulating state to the Lyman α intensity depends on the product of the population of that state

TABLE II. Results of measurements of Lyman-α radiation.

Foil thickness $\mu g/cm^2$	Experimental mean life (10^{-9} sec)
5	$\tau_1 = 1.60 \pm 0.02$ $\tau_2 = 14.0 \pm 0.5$
20	$\tau_1 = 1.58 \pm 0.02$ $\tau_2 = 18.5 \pm 0.8$
20	$\tau_1 = 1.62 \pm 0.02$ $\tau_2 = 19.5 \pm 0.6$
40	$\tau_1 = 1.58 \pm 0.03$ $\tau_2 = 17.0 \pm 0.6$
40	$\tau_1 = 1.65 \pm 0.04$ $\tau_2 = 19.4 \pm 1.0$
40	$\tau_1 = 1.61 \pm 0.03$ $\tau_2 = 19.7 \pm 0.8$

and the transition probability to the $2p$ state. Since there is little understanding concerning the mechanism of production of excited states by the foil and therefore the population of these states, it is not possible to uniquely identify τ_2 with a particular state or states.

Table II shows the result of curve fitting to several sets of Lyman-α decay curves taken with previously unused 5, 20, and 40-$\mu g/cm^2$ carbon foils as the excitation medium. The values of τ_1 in Table II are to be associated with the radiative mean life of the $2p$ state of atomic hydrogen. It is seen that the values of τ_1 are all in relatively close agreement and constitute a set of independent measurements. The weighted average value of these measurements, $\bar{\tau}_{2p} = (1.60 \pm 0.01) \times 10^{-9}$ sec, is in agreement with either theoretical value listed in Table I.

The values for τ_2 for all foils of thickness 20 and 40 $\mu g/cm^2$ are seen to agree within experimental error. On the other hand, the value of τ_2 for the 5-$\mu g/cm^2$ foil is significantly below the weighted average of the values for the thicker foils of $(18.8 \pm 1.1) \times 10^{-9}$ sec. As discussed later in this paper (see Fig. 5), the shape of a Lyman-α intensity decay curve varies with the thickness of the foil used. This effect may reasonably be interpreted as due to a change in the population of the higher cascading states compared with the $2p$ state population for different foil thicknesses. A population redistribution among the excited states as foil thickness is changed could change the value of τ_2, since τ_2 describes cascading to the $2p$ state. If, for example, the $3s$ and $3d$ states were the only cascading states of significance, then the change in τ_2 for a thin foil could be due merely to a change in the ratios of the $3s$ to $2p$ and $3d$ to $2p$ populations as compared to a thicker foil. This variation in the cascade contributions, which affects the value of τ_2, would not be expected to appreciably affect the value of τ_1, since as is seen in Fig. 3, the repopulation contributions to the Lyman-α counting rate at distances corresponding to two $2p$ state mean lives is only $\frac{1}{10}$ that due to the counting rate produced by the initial $2p$ state population decay.

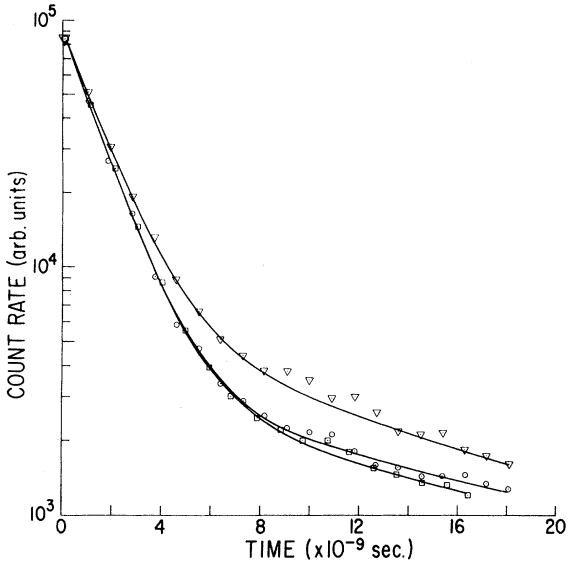

FIG. 5. Lyman-α intensity decay curves taken with a new 5-$\mu g/cm^2$ carbon foil (upper curve), a used 5-$\mu g/cm^2$ foil (middle curve), and a new 40-$\mu g/cm^2$ carbon foil (lower curve). Data taken with a 20-$\mu g/cm^2$ carbon foil are difficult to distinguish from the lower curve. The curves are normalized at the first datum point. Counting rate errors are insignificant compared to the size of the symbols used. The solid curves are the result of the curve fitting analysis. The four points between 9 and 12 nsec in the upper curves and the four points between 9 and 13 nsec in the lower curve were not used during the curve-fitting analysis for reasons explained in the text.

Calculations have been made to estimate the effects of any possible fields on the otherwise free atoms in the excited beam. It can be shown from Stark effect mixing calculations[18] that an electric field of the order of 50 V/cm or an equivalent magnetic field of 10 G would be needed to noticeably affect our results. At this field strength, the mean life of the $2p$ state of hydrogen would be increased to 1.62×10^{-9} sec. Since there is no evident source of such fields being present in our experimental chamber, Stark mixing has been considered negligible.

2. Lyman β Results

Figure 6 shows a sample curve from the measurements on Lyman-β radiation. The Lyman-β results were curve fitted in the manner described previously and the best fit to the data was found to be a single exponential. Table III shows the curve-fitting results for three different sets of data. The mean value of these results is $(5.5 \pm 0.2) \times 10^{-9}$ sec. No cascading effects can be seen from the curves; however, the large statistical error due to the weak-line intensity would overshadow a small cascade effect.

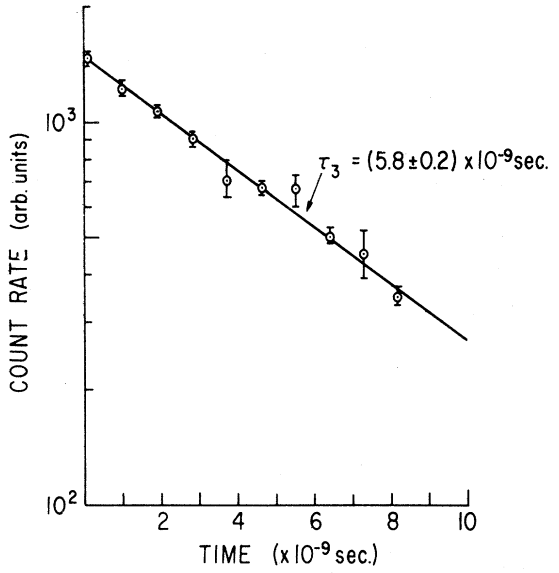

FIG. 6. A typical intensity decay curve of the Lyman-β line of atomic hydrogen, obtained with 340.5-keV H_2^+ ions and a 5-μg/cm^2 carbon foil. The straight line represents the best fit to the experimental data.

3. Foil Effects

During the course of these experiments it became apparent that the foil properties were changing with time. It was found upon analyzing the results that the computed $2p$ state mean life would agree with the theoretical value for only the first and second sets of Lyman-α intensity data obtained with the same foil. Whenever decay curves were generated using older foils, it was found that the computed value for the decay time was considerably shorter. It is obvious not to expect the fundamental atomic properties to change; therefore, it is concluded that the foil properties are changing in such a way as to change the energy loss of the ions in the foil so that the excited atoms have a lower emergent velocity than when the foils were fresh. Thus we conclude that the change in the decay times (which are all based on the same atom velocity) as the foil ages is due to the use of the incorrect velocity to transform from the distance scale to a time scale when plotting the decay curves. A clear example of this phenomenon is shown in Table IV for a 5-μg/cm^2 foil. The experimentally determined decay time is seen to decrease in value as the foil ages. This is the best example of this phenomenon using a single foil; however, several cases of runs with old foils have been analyzed which consistently gave a low value for τ_1. Presumably then, as foils age in our vacuum system and undergo H_2^+ bombardment, a significant change in energy-loss results. This interpretation suggests that the energy-loss properties of the foils have changed sufficiently to produce, in some cases, as much as a 5% change in atom velocity. The diffusion pumps used in the present accelerator vacuum system all used silicone oils and the system was trapped with liquid

TABLE IV. The effect of foil age on measured decay times for a 5 μg/cm^2 carbon foil.

Foil age[a]	Experimental decay time τ_1 (10^{-9} sec)
1(1)	1.60 ± 0.02
3(3)	1.54 ± 0.02
11(7)	1.50 ± 0.02
13(8)	1.51 ± 0.02

[a]The foil age indicates the number of days the foil had been in the system. The number of experiments completed with the foil is indicated in parenthesis.

N_2 only when Lyman-α intensity decay data were taken. Thus a large change in energy loss due to surface contamination might not be surprising. Similar difficulties in experiments designed for studying the interaction of various ions with metallic foils are discussed by Allison and Warshaw[19] and Allison.[20] Further evidence for effective thickening of foils in experiments of this nature has been observed by Bickel[21] in passing ion beams of S and FD_2 through the foils. Changing foil properties in the present experiment were also evident from the fact that new foils were translucent, whereas old foils became nearly opaque.

The analysis of the data from many runs with foils of different thicknesses has shown that the shape of the Lyman-α decay curve is a function of foil thickness. This is illustrated in Fig. 5, where results from runs using fresh foils of thicknesses 5 μg/cm^2 and 40 μg/cm^2 are compared and show a dramatic difference in shape. The difference is undoubtedly due to a difference in initial-state populations. This figure also shows the effect of foil aging on a 5-μg/cm^2 foil. The difference in shape of the upper (new 5-μg/cm^2 foil) and middle (old 5-μg/cm^2 foil) curves is due to the combined effects of energy-loss change and change in initial-state populations.

It is also to be expected that the foil effects observed in the present experiments would be dependent on the type of ion accelerated as well as its energy.

IV. CONCLUSIONS

The radiative mean lives of the $2p$ and $3p$ states of atomic hydrogen have been determined. The measured value for τ_{2p} is in good agreement with the theoretical values given in Table I. Although the two sets of theoretical values quoted in the table are different, it is presumed that the more recent calculations[5] are appropriate for comparison with experiment. In this regard it is seen that the experimental value for τ_{3p} is in slight disagreement with the theoretical value.[5] Repopulation effects may be responsible for the difference.

It has been determined that the Lyman-α decay curves produced by using previously unused foils are best fitted by two exponential terms whose characteristic decay times correspond to spontaneous decay of the $2p$ state and repopulation of that

state by higher states. It was observed that the problem of cascading may complicate the analysis of data obtained using the beam-foil method and could result in erroneous conclusions.

Changing foil characteristics have also been observed in the course of these experiments. Aging of foils in the accelerator vacuum system and under ion bombardment produced apparent changes in ion-energy loss. Consistent results were obtained only by using fresh foils for mean-life measurements. It is also evident that the initial populations of atomic states depend on the thickness of the foil used.

The actual mechanism of the excitation process within the foil is not yet fully understood. Due to the fact that the mean-free paths of the incident ions are small compared to the foil thickness, many ion-foil atom collisons will occur. It may be that the actual final excitation process occurs at the emergent surface of the foil and if this is the case, surface contamination would affect the initial-state populations. Some understanding of the excitation mechanism may be reached through polarization studies of the emitted radiation. Preliminary experimental studies in this endeavor have not thus far revealed any polarization.

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Regularities in Atomic Oscillator Strengths*

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Many regularities and systematic trends among atomic oscillator strengths have been studied using the extensive material which has recently become available for the lighter elements. The quantum-mechanical background for the existence of these regularities is discussed, and in particular the relationship between oscillator strength and nuclear charge as predicted from conventional perturbation theory is reviewed in detail. A number of characteristic numerical examples are then presented. The regularities are of great practical importance, since they may be exploited to obtain additional oscillator strengths by simple interpolation techniques as well as to evaluate the reliability of existing data by the degree of fit into established systematic trends.