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Mean Lives of the 2*p* and 3*p* Levels in Atomic Hydrogen*

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Radiation from hydrogen atoms in excited states produced by the beam-foil method was studied as a function of time using a vacuum ultraviolet monochromator. Measured values of the mean lives of the $2p$ and $3p$ levels in hydrogen are presented and compared with theory. The effects of cascades from levels of higher n were observed and have been included in the mean-life calculations.

I. INTRODUCTION

ATTEMPTS to measure atomic mean lives began with Wein[1] but the amount of accurate experimental data on this subject is still surprisingly meager. The past few years have seen an increase in activity in this field, thanks in part to the development of the beam-foil excitation technique[2] which is directly applicable to this research. Recently, mean lives of excited states formed by beam-foil excitation have been investigated in nitrogen,[3] oxygen,[4] hydrogen,[5] neon,[6] helium,[7] and the iso-electric sequence of lithium-like ions.[8]

We have used the beam-foil excitation technique to measure the mean lives of the $2p$ and $3p$ levels of hydrogen. Cascades from higher levels into the $2p$ and $3p$ levels were observed and their effect on the rate of decay of the $2p$ and $3p$ levels has been determined. Hydrogen has been chosen for these initial experiments because its wave functions are exactly known and allow all observed effects to be amenable to exact calculation.

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[1] W. Wein, Ann. Physik **60**, 39 (1919); **66**, 16 (1921); **73**, 32 (1924); **83**, 1 (1927).

[2] S. Bashkin, Nucl. Instr. Methods **28**, 88 (1964).

[3] L. Kay, Proc. Phys. Soc. (London) **85**, 163 (1965).

[4] W. S. Bickel and S. Bashkin, Phys. Letters **20**, 488 (1966).

[5] A. S. Goodman and D. J. Donahue, Phys. Rev. **141**, 1 (1966).

[6] S. Bashkin, L. Heroux, and J. Shaw, Phys. Letters **13**, 229 (1964).

[7] S. Bashkin, D. Fink, P. R. Malmberg, A. B. Meinel, and S. G. Tilford, J. Opt. Soc. Am. (to be published).

[8] K. Berkner, W. S. Cooper, S. N. Kaplan, and R. V. Pyle, Phys. Letters **16**, 35 (1965).

II. THEORY

The intensity of a spectral line (in photons/sec) resulting from a radiative electric dipole transition between the atomic levels at energies E and E' and described by the quantum numbers nlj and $n'l'j'$ ($E_{nlj}>E'_{n'l'j'}$) is given by

$$I_{n'l'j'}{}^{nlj}(t)=A(nlj\rightarrow n'l'j')N_{nlj}(t)\,, \qquad (1)$$

where the Einstein coefficient, $A(nlj\rightarrow n'l'j')$, is the probability per unit time that the transition will occur and $N_{nlj}(t)$ is the population of the upper level. If the contributions to the population of the upper level due to cascades from higher levels are negligible,

$$N_{nlj}(t)=N_{nlj}{}^0e^{-\alpha_{nlj}t}\,, \qquad (2)$$

where $N_{nlj}{}^0$ is the population of the level $|nlj\rangle$ at $t=0$ and $\alpha_{nlj}=\sum_{n'l'j'}A(nlj\rightarrow n'l'j')$ is the reciprocal of the mean life of the level $|nlj\rangle$. Therefore, we have

$$I_{n'l'j'}{}^{nlj}(t)=A(nlj\rightarrow n'l'j')N_{nlj}{}^0e^{-\alpha_{nlj}t}. \qquad (3)$$

It has been shown that $A(nlj\rightarrow n'l'j')$, and therefore α_{nlj}, is independent of j, m_j and j', m_j'.[9,10] Dropping the subscript j, we have

$$I_{n'l'}{}^{nl}(t)=A(nl\rightarrow n'l')N_{nl}{}^0e^{-\alpha_{nl}t}\equiv I_{nl}{}^0e^{-\alpha_{nl}t}. \qquad (4)$$

When cascades cannot be neglected, the expression for $I_{n'l'}{}^{nl}$ becomes more complicated. If the levels n_il_i, $i=2\cdots M$, cascade into the level n_1l_1, Eq. (2) must

[9] E. V. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1963), p. 71.

[10] L. R. Maxwell, Phys. Rev. **38**, 1664 (1931). Proof due to J. H. Van Vleck.

be replaced by

$$N_1(t)=N_1^0e^{-\alpha_1 t}+\sum_i^M \beta_{1i}N_i^0(e^{-\alpha_i t}-e^{-\alpha_1 t})\,, \qquad (5)$$

where the subscripts 1 and i represent n_1l_1 and n_il_i, respectively, and

$$\beta_{1i}=A\,(n_il_i\to n_1l_1)/(\alpha_1-g_i)\,. \qquad (6)$$

We note that if the $1/\alpha_i$ are large compared to $1/\alpha_1$, the effect of cascades is very small[11] and

$$N_1(t)\approx N_1^0e^{-\alpha_1 t}. \qquad (7)$$

In any method of detection employing the counting of photons emitted during atomic transitions from $|nl\rangle$ to $|n'l'\rangle$, the intensity of the spectral line is related to the actual counting rate $c(t)$ in the absence of background, by

$$c(t)=kI_{n'l'}{}^{nl}=kA\,(nl\to n'l')N_{nl}(t)\,, \qquad (8)$$

where k is a constant which depends on geometrical factors and detection efficiency.

In the beam-foil excitation technique, ions traveling with a well-defined velocity v in a well-defined beam are put into various excited levels on passing through foil. The emergent particles, which are atoms or ions, generally radiate as they travel downstream from the foil, and the change in the intensity of this radiation as a function of distance x from the foil may be used to determine the mean lives of the excited levels.

Equation (5) may be written

$$N_1(t)=\sum_{i=1}^M K_ie^{-\alpha_i t}\,,$$

with (9)

$$K_1=(N_1^0-\sum_{i=2}^M \beta_{1i}N_i^0)\,,$$

and

$$K_i=\beta_{1i}N_i^0\,, \quad i\neq 1\,;$$

or, using $t=x/v$,

$$N_1(x)=\sum_{i=1}^M K_ie^{-\alpha_i x/v}\,, \qquad (10)$$

where K_i is independent of x. Therefore,

$$c(x)=kA\,(nl\to n'l')\sum_{i=1}^M K_ie^{-\alpha_i x/v}. \qquad (11)$$

The general form of $c(x)$ is shown in Fig. 1.

Since a finite length of the radiating beam is observed by the detector, the total counting rate is given by the integral of $c(x)dx$ over this length. If the center of the interval with length 2δ is at x_c we have

$$C(x_c)=\int_{x_c-\delta}^{x_c+\delta} c(x)dx=kA\,(nl\to n'l')\sum_{l=1}^M K_i\int_{x_c-\delta}^{x_c+\delta} e^{-\alpha_i x/v}dx$$

$$=kA\,(nl\to n'l')\sum_i^M 2K_i\frac{v}{\alpha_i}\sinh\left(\frac{\alpha_i\delta}{v}\right)e^{-\alpha_i x_c/v}.$$

Note that if $\alpha_i\delta/v$ is small, $2(v/\alpha_i)\sinh(\alpha_i\delta/v)\cong 2\delta$ and the functional dependence of the change in counting rate with distance downstream from the foil is essentially independent of the length of beam observed.

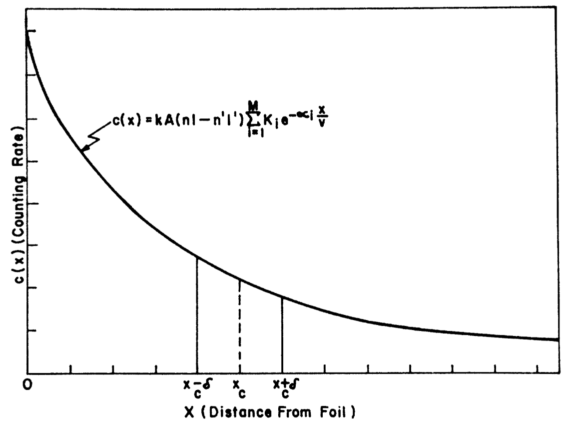

FIG. 1. The general form of the dependence of counting rate on distance. This curve represents radiation from an excited state enhanced by cascades from higher states.

III. EXPERIMENTAL TECHNIQUE

Singly ionized hydrogen molecular ions of mass three (HHH^+) were accelerated by a Van de Graaff accelerator and allowed to pass through a thin ($\sim$10 μg/cm^2), self-supporting, carbon foil which was perpendicular to the beam. Excitation of the beam particles by the foil caused the beam to become self-luminous. A 0.5-cm length of this beam was viewed with a McPherson model-235 half-meter Seya-Namioka vacuum uv scanning monochromator (see Fig. 2). For measurements dealing with the $3p$ states of hydrogen the beam energy was 347 keV and the spectrometer was set at Lyman β, $1s$-$3p$ λ1025.7 Å. For measurements dealing with the $2p$ states of hydrogen the beam energy was 964 keV and the spectrometer was set at Lyman α, $1s$-$2p$ λ1215.6 Å. In both cases the entrance and exit slit widths were 1.0 mm. The light intensity at the exit slit was detected by an EMI photomultiplier (9514S) coated with sodium salycilate and radiation-cooled with liquid nitrogen. The photomultiplier was used as a photon counter. Its output was amplified by an Ortec 101-201 charge sensitive preamplifier-amplifier system and the pulses counted on a Hamner (N-276) scaler. The beam current was 2 μA and was monitored with a Faraday cup connected to a current integrator. All photomultiplier

[11] This also depends on the reasonable assumption that the higher excited states are not as profusely populated as the lower states. Relative intensity measurements of Ly_α and Ly_β indicate that the populations fall rapidly with increasing n.

counts were normalized to equal amounts of charge collected in the Faraday cup in order to ensure that the decrease in intensity with distance was due to decay of the atoms in the beam from excited states and not to fluctuations in the beam current.

The carbon foil was translated upstream from the entrance slit of the spectrometer by means of a precision screw of 32 threads per inch. Moving the foil upstream increased the distance between the observed section of the beam and the point of initial excitation. This allowed us to measure the decay of intensity of the light from the beam as a function of distance from the exciter foil. The counting rates observed were determined to be the sum of three contributions:

(1) the dark current of the photomultiplier,
(2) the background light caused by collisions between beam particles and the residual gas atoms in the target chamber,
(3) the signal from the beam caused by decays of atoms initially excited by the foil.

In the measurement of the Lyman α and Lyman β light, the two sources of background were determined separately. The background due to the photomultiplier dark current was determined by closing the exit slit of the spectrometer and counting for 100 sec. This background was determined to an accuracy of $\pm 2\%$ and amounted to less than 10% of the total signal for Lyman α and less than 30% for Lyman β. The background from random excitations was measured by rotating the foil out of the beam, counting for 100 sec and subtracting the photomultiplier dark current. This source of background amounted to less than 4% of the total signal.

We emphasize that all measurements of intensity from both Lyman α and Lyman β were made with the same foil. From other experiments, we have learned that the nature of the light emitted by this beam is a function of the thickness of the foil and perhaps of the nature of its preparation. It cannot be assumed that

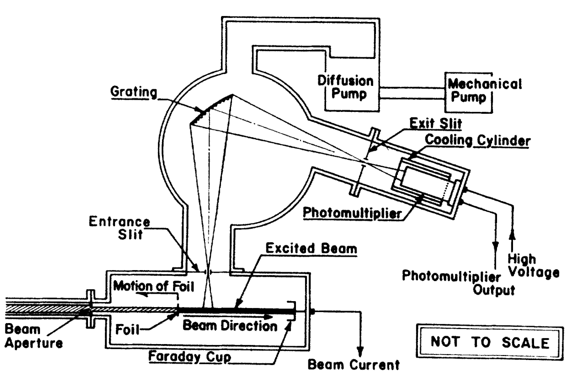

FIG. 2. Experimental arrangement showing spectrometer, target chamber, and detector chamber. Suppresser plate and grounded shield on Faraday cup are not shown.

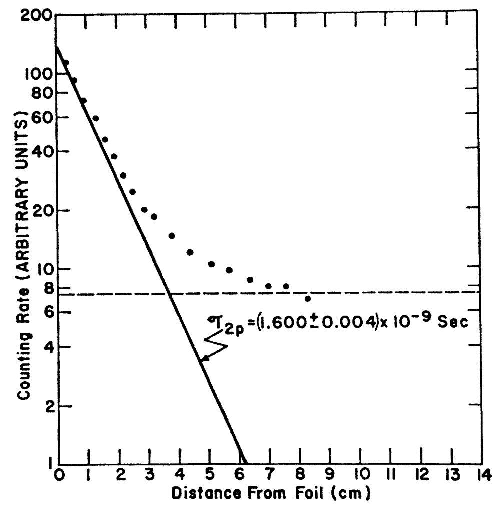

FIG. 3. Results of measurements of Lyman α radiation. Solid points are experimental data. Solid straight line represents the mean life of the $2p$ state, while the dashed line is the computer-fitted constant term.

excitation equilibrium exists in foils with the thickness ($\sim 10\ \mu g/cm^2$) we used.

IV. RESULTS

A. Lyman α Measurements

The decay of Lyman α was observed over 8.27 cm. The beam velocity in this case was 7.92×10^8 cm/sec, which gave an observation time of 1.04×10^{-8} sec or about 6.5 theoretical mean lives.

Figure 3 shows a semilog plot of the decrease in counting rate from Lyman α light as a function of distance downstream from the foil. This composite curve was decomposed using a version of the "Malik" curve-fitting program.[12] These data were first fitted to functions of several exponentials but the fits were poor. In the case of the two exponentials, the value for the mean life of the second term was on the order of 2×10^{-4} sec. The best fit to the data was obtained when the data were fitted to one exponential plus a constant. A constant is acceptable since the states which can cascade into the $2p$ level are long-lived and our data in the region beyond 6.5 cm are too poor to provide discrimination between mean lives of ∞ and 2×10^{-7} sec. The computer fit yields

$$1/\alpha_{2p}=\tau_{2p}=(1.600\pm 0.004)\times 10^{-9}\text{ sec}.$$

B. Lyman β Measurements

The decay of intensity of Lyman β was measured over a distance of 3.92 cm. The beam velocity was 4.74×10^8 cm/sec. Therefore, the decay of the $3p$ states was

[12] F. Grard, University of California Radiation Laboratory Report No. UCRL-10153, T.I.D.-4500, 17th ed. (unpublished).

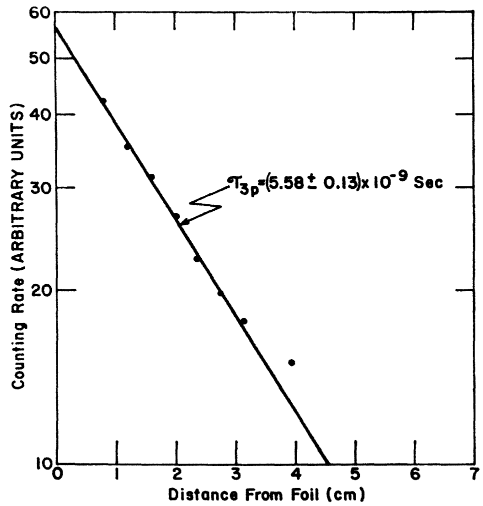

FIG. 4. Results of measurements of Lyman β radiation. Solid points are experimental data. The straight line results from a least-squares fit to the experimental points.

viewed for 5.9×10^{-9} sec or about 1.1 theoretical mean lives.

Figure 4 shows the results of observations of the decrease in intensity of Lyman β light. Cascade effects are again apparent but are too weak to warrant analysis by the use of "Malik." The mean life of the $3p$ state was determined by a least-squares fit to the first six points shown in Fig. 3. The solid line represents this decay. The least-squares fit yields

$$1/\alpha_{3p}=\tau_{3p}=(5.58\pm0.13)\times10^{-9}\text{ sec}.$$

The experimental results are shown in Table I, along with the theoretical values of τ_{2p} and τ_{3p} from Condon and Shortley.[13]

We note that the velocities used in this experiment were determined to better than 0.5% and that there was essentially no error in the location of the position of the foil. In the data used to determine the mean life of the $3p$ level the statistics on the experimental points are better than 2%. The 3% deviation of our measured value for τ_{3p} from the theoretical value given by Condon and Shortley cannot, therefore, be accounted for by any experimental error. This deviation is interpreted as being caused by the effect of cascades on the decrease of intensity of Ly_β radiation.

We have considered the effect of small extraneous electric fields on the mean lives of the $2p$ and $3p$ levels.

[13] Reference 9, p. 136, Table 5^6.

TABLE I. Summary of results of Lyman α and Lyman β measurements.

Spectral line	Transition	Experimental mean life (10^{-9} sec)	Theoretical mean life (10^{-9} sec)
1215.6 Å Ly_α	$1s$-$2p$	1.600 ± 0.004	1.60
1025.7 Å Ly_β	$1s$-$3p$	$5.58\ \pm0.13$	5.40

These fields might arise from accumulations of charge on nearby surfaces in the target chamber or from electrons or ions in the beam itself. Our calculations, which follow the treatment of the Stark effect in hydrogen by Bethe and Salpeter[14] show that although mixing of these states occurs for very small fields, stray fields would have to be greater than 30 V/cm before our measurements would be affected. At 30 V/cm, the state that reduces to pure $2p$ for zero electric field has a mean life of 1.606×10^{-9} sec and the mean life of the state that reduces to pure $2s$ for zero field is 3.75×10^{-7} sec. Specific studies[15] of the effect of electric fields on the intensity of light radiated downstream from the foil confirm the fact that any fields present in these experiments are less than 5 V/cm.

V. SUMMARY

Experimental mean lives for the $2p$ and $3p$ levels in hydrogen have been determined. The measured value for τ_{2p} agrees with theory and the 3% deviation of the measured value of τ_{3p} from the theoretical value is attributed to cascade effects.

The general technique outlined here may be extended to yield data on the relative initial populations of decaying states and those states which cascade into them. Such work is in progress.

The accuracy with which we were able to determine these mean lives demonstrates the elegance of the thin-foil excitation technique when applied to measurements of mean lives.

ACKNOWLEDGMENTS

It is our pleasure to thank Dr. S. Bashkin and Dr. L. Marquet for many helpful discussions concerning various phases of this experiment.

[14] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic Press Inc., New York, 1957), p. 284.

[15] S. Bashkin, W. S. Bickel, D. Fink, and R. K. Wangsness, Phys. Rev. Letters **15**, 284 (1965).