THE

PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 141, No. 1

JANUARY 1966

Mean Lives of Some States in Atomic Hydrogen*

A. S. GOODMAN AND D. J. DONAHUE Department of Physics, University of Arizona, Tucson, Arizona (Received 16 August 1965)

Hydrogen atoms in excited states were produced by passing a beam of fast-moving hydrogen molecules through a thin carbon foil. Radiation from these atoms was studied as a function of time using filtered photomultiplier tubes. Measured values of the mean lives of the 3s, 3d, 4s, 4p, and 4d states in hydrogen are presented and compared with theory. Results of measurements of relative initial intensities of radiation from fine-structure states of the n=3 and n=4 levels in hydrogen are also presented.

I. INTRODUCTION

N important property of an excited state of an atom is its mean life. A comparison of experimental and calculated values of such a mean life provides a sensitive test of the correctness of the atomic wave functions used in the calculations. In addition, the mean lives of excited atomic states are crucial in some calculations of the abundances of elements in stars.

Although several methods have been developed for measuring mean lives, most of them are limited in application, and the information available concerning decav constants of atomic states is meager. In this paper we describe the application of a new technique to the measurement of the mean lives of some states in atomic hydrogen. The method is capable of giving precise values for mean lives, and is potentially applicable to a large number of atomic states. In fact, experiments quite similar to the one described here have recently been performed on states in various light elements.¹⁻⁴ When applied to states in hydrogen, the method yields not only the mean lives of some of the individual fine-structure states, but also gives information concerning the relative populations of these states. This information could be of use in studying the processes by which the excited states were produced.

The measurement of the mean lives of states in hydrogen is an old problem, and was first undertaken by Wien⁵ in 1919. His efforts proved to be unsuccessful, and vielded results in disagreement with quantum-mechanical predictions as well as with later experiments. Slack in 1926 measured the mean life of the 2p level in hydrogen. Griffiths,⁶ and Ankudinov et al.⁷ have measured the average mean lives of the n=3, 4, and 5 levels in hydrogen. A recent abstract⁸ indicates that Hughes and coworkers have done an experiment similar to the one described herein, with similar results.

II. EXPERIMENTAL METHOD

Hydrogen atoms in excited states were produced by the method described by Bashkin.9 A schematic diagram of the experimental arrangement is shown in Fig. 1. Singly ionized mass-three hydrogen molecules were accelerated to an energy of 157.3 keV in the University of Arizona Van de Graaff accelerator. The energy of the molecules was determined by magnetic analysis. The beam was directed at a self-supporting carbon foil. The thickness of this foil was determined by weighing it, and was $6\pm 2 \ \mu g/cm^2$. On the downstream side of the

⁶ F. G. Ślack, Phys. Rev. 28, 1 (1926).

Copyright © 1966 by The American Physical Society.

^{*} This work was supported in part by the National Aeronautics and Space Administration and is from the Master's degree thesis of A. S. Goodman.

¹S. Bashkin and G. N. McIntire, Bull. Am. Phys. Soc. 10, 50 (1965)

⁽¹⁾ ⁽¹⁾ ⁽²⁾ ⁽²

⁵ W. Wein, Ann. Physik **60**, 39 (1919); **66**, 16 (1921); **73**, 32 (1924); **83**, 1 (1927).

⁶ F. G. SIACK, Phys. Kev. 25, 1 (1920).
⁷ V. A. Ankudinov, S. V. Bobashiev, and E. P. Andreev, Zh. Eksperim. i Teor. Fiz. 48, 40 (1965) [English transl.: Soviet Phys.-JETP 21, 26 (1965)].
⁸ H. R. Dawson, B. M. Doughty, and R. H. Hughes, Bull. Am. Phys. Soc. 10, 170 (1965).
⁹ S. Bashkin, Nucl. Instr. Methods 28, 88 (1964).



foil the beam contained protons and hydrogen atoms, some in excited states, with speeds of $(3.08\pm0.05)\times10^8$ cm/sec. Since the velocity of the radiating atoms was accurately known, it was only necessary to measure the radiation emitted by these atoms as a function of distance from the foil in order to study the time behavior of the excited atoms.

Radiation from the atoms was detected by two RCA 1P21 multiplier phototubes which were mounted outside the vacuum system and viewed the beam through Lucite windows. One of these phototubes was fixed in position, and was used to monitor the number of excited atoms present in the beam. The second phototube was mounted so that it could be moved in a direction parallel to the direction of the beam over a distance from about 1 cm from the foil to about 30 cm from the foil. Collimators in front of this movable phototube insured that in a given position it viewed only about $\frac{1}{2}$ cm of the radiating beam. Both tubes were shielded by interference filters so that only radiation in a narrowwavelength region could reach the phototubes. In order to reduce noise, both phototubes were operated at the temperature of solid CO₂. Pulses produced in the phototubes were amplified with charge-sensitive preamplifier and amplifier systems, and were recorded in scalers.

A measurement of the time behavior of atomic states emitting radiation passed by the filters consisted simply of measuring the ratio of count rates in the stationary and movable phototubes as a function of the position of the movable tube. It was necessary to correct the



FIG. 2. Results of measurements on H_{α} radiation. Solid points are experimental data. Straight lines result from a least-squares fit to the experimental points.

data for radiation from atoms excited by the residual gas in the vacuum system. This background was measured periodically at various positions along the path of the beam by recording counts in the movable tube with the carbon foil removed from the beam.

III. RESULTS AND ANALYSIS

1. H_{α} Measurements

Results of the measurements using filters which pass radiation emitted in $n=3 \rightarrow n=2$ transitions in hydrogen are shown in Fig. 2. In this figure the ratio of counting rates in the stationary and movable tubes is plotted on a logarithmic scale as a function of the distance from the foil to that part of the beam observed by the movable tube. As can be seen, the experimental curve is not simple, and must result from the exponential decay of more than one state in the n=3 level of hydrogen. We would in fact expect to see the decay of the 3s, 3p, and 3d levels. Theoretically,¹⁰ most of the decays from the 3p state proceed to the 1s state, and the radiation from these transitions will not pass through the filters. Further, the 3p level is expected¹⁰ to have a mean life sufficiently short so that it is depleted by about a factor of 2 between the foil and the closest position of the movable tube to the foil. The combination of these two effects make it unlikely that we could observe decays from the 3p level. Accordingly, we have analyzed the experimental data assuming that they are composed of two exponential functions.

A least-squares fit was performed with a computer using a program described by Grard.¹¹ Four unknowns, the mean lives of the 3s and 3d states and the initial intensity of radiation from these states, as well as the standard deviations of these quantities, were obtained from the analysis. The results of this analysis are shown in Table I. The measured mean lives are in agreement with calculated values tabulated in Condon and Shortley.¹⁰ The calculated relative initial intensities shown in the table were obtained by assuming that the 3s and 3dstates were initially populated according to their statistical weights. As can be seen, this assumption is bad,

TABLE I. Results for H_{α} .

State	Mean life (1 Experimental	10 ⁻⁸ sec) Theoreticalª	Initial in (arbitrary Experimental	tensity y units) Theoretical ^b
3s	13.5 ± 1.4	15.9	4.6 ± 0.3	4.6
3p		0.54		
3d	1.61 ± 0.06	1.56	23.2 ± 1.7	

^a See Ref. 10.
 ^b Calculated assuming states are initially populated according to their statistical weights.

¹⁰ E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1963), p. 136, Table 5⁵.

¹¹ F. Grard, University of California Radiation Laboratory Report UCRL-10153, T.I.D.-4500, 17th ed. (unpublished).



FIG. 3. Results of measurements of H_{β} radiation. Solid points are experimental data. Straight lines result from a least-squares fit to the experimental points.

and in fact the data indicate that in our experiments the ratio of initial populations of the 3s and 3d states must be more nearly 2.0 than 0.2.

2. H_{β} Measurements

Results of measurements using filters which pass radiation emitted in $n=4 \rightarrow n=2$ transitions in hydrogen are shown in Fig. 3. These data are qualitatively similar to the H_{α} results. However, because the phototubes are more sensitive to H_{β} than to H_{α} radiation, the precision of the H_{θ} data is better than that for the H_{α} measurements. Further, the combination of theoretical initial intensity and mean life of the 4p state make it possible that radiation from that state as well as from the 4s and 4d states could be observed in these experiments. Accordingly, these data were analyzed with the assumption that they represent radiation from three states decaying exponentially. The results of a computer fit to the data are also shown in Fig. 3 and listed in Table II. The errors listed with the results are standard deviations and were obtained from the com-

TABLE II. Results for H_{β} .

State	Mean life (Experimental	10 ^{–8} sec) Theoreticalª	Initial in (arbitrary Experimental	tensity units) Theoretical ^b
4s	$\begin{array}{c} 18.6 \ \pm 2.7 \\ 1.46 \pm 0.25 \\ 3.77 \pm 0.55 \end{array}$	23.2	2.22 ± 0.35	2.22
4p		1.24	3.94 ± 1.1	25.3
4d		3.65	6.45 ± 1.1	90.5

^a See Ref. 10.
 ^b Calculated assuming states are initially populated according to their statistical weights.



puter analysis. Again the measured mean lives are in agreement with the calculated values listed in Condon and Shortley, and the relative initial intensities are in disagreement with the assumption that the individual levels were populated according to their statistical weights. As was the case with the H_{α} measurements, it appears that states with low orbital angular momentum are preferentially populated in these experiments.

One might wonder if the solutions listed in Table II are unique. In fact, without prior information we would be inclined to try to fit the data with only two exponential functions rather than with three. We have analyzed the data with two and with four exponentials, as well as with three and have obtained a goodness-of-fit parameter χ^2 for each of the fits. Relative values of χ^2 as a function of the number of exponentials used in a fit are plotted in Fig. 4. As illustrated, the fit with three exponentials is considerably better than that with two, and not much worse than that with four. Thus three exponentials seem to give the most satisfactory fit to the experimental results. However, it is clear that the analysis of data containing more than two exponentials must be performed with some care.

ACKNOWLEDGMENT

The authors express their gratitude to Professor E. Malamud for his assistance in performing the computer analyses described herein. They also thank Professor S. Bashkin for his help with various phases of the experiment.