

A Method for Determining Beam-Foil Excited Hydrogenic Populations from Lyman and Balmer Decay Curves which Exactly Accounts for Cascading

D. R. Bryant and L. J. Curtis

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio

Received February 11, 1974

Abstract. A method is presented which permits the determination of relative initial p -state populations and combined s - and d -state populations of beam-foil excited hydrogenic atoms from Lyman and Balmer decay curves. The method utilizes exact and linear cascade relationships which exist between corresponding portions of these decay curves when they are measured under common excitation conditions, and does not involve exponential curve fitting. The technique determines (but does not require a measurement of) the relative normalizations of the various decay curves by a simple linear regression, thus inferring initial populations from the renormalized relative intensities as extrapolated to the foil position. The basic relationships of the method are developed, and results are presented from a computer study of the quality of data which would be required to extract populations by this technique.

Introduction

The relative initial populations of beam-foil excited hydrogenic levels have been studied by a number of authors [1–11] using techniques such as multi-exponential curve fitting and electric field stark quenching. Recent increases in experimental resolution and in the time window after excitation which can be reliably viewed [13] have made possible highly reproducible results for the $n=3$ level populations [8, 11]. An interesting technique, developed by Tielert and Bukow [10], has been utilized to jointly analyze simultaneously measured Ly_{α} and H_{α} decay curves by a correlated non-linear curve fit, thus including effects of several cascades in the population determination. The success of this approach suggests that it is experimentally possible to include large numbers of cascades in such an analysis [12]. We have therefore developed a formalism by which a large fraction of or even the entire Lyman and Balmer series can be incorporated into a unified analysis which deter-

mines the relative p -state and admixed s - and d -state populations through a correlated linear regression, which utilizes the measured decay curves (rather than their multiexponential representations) as the basic fitting functions.

Computational Methodology

The $2p$ level in hydrogenic atoms has a single radiative decay to $1s$, and is repopulated by direct cascades from the various ns and nd levels, so that its population is governed by

$$dN_{2p}/dt = \sum_{n=3}^{\infty} [N_{ns}(t) A_{ns2p} + N_{nd}(t) A_{nd2p}] - N_{2p}(t)/\tau_{2p} \quad (1)$$

where N_{nl} denotes the instantaneous population of a level with principal quantum number n and orbital angular momentum quantum number l (summed

over the meanlife-degenerate fine and hyperfine sub-states), $A_{nl'n'}$ is the transition probability between such states and τ_{2p} is the meanlife of the $2p$ state. The decay curves of the Lyman and Balmer series depend explicitly upon these same quantities. Thus the measured intensity of the Lyman series can be written

$$I_{n1}(t) = \xi_{n1} N_{np}(t) A_{np1s} \quad (2)$$

while the Balmer series is described by the degenerate set of transitions

$$I_{n2}(t) = \xi_{n2} [N_{ns}(t) A_{ns2p} + N_{np}(t) A_{np2s} + N_{nd}(t) A_{nd2p}] \quad (3)$$

where $I_{nn'}$ denotes the intensity detected by the spectrometer system at the appropriate wavelength at a given time t after excitation, and $\xi_{nn'}$ is the detection efficiency at that wavelength. Notice that the n th element of the Balmer series contains the cascade terms from ns and nd into $2p$, and would correspond to the bracketed term in Eq. (1) if the np term were subtracted off. This can be done, since the corresponding np term is contained in the n th element of the Lyman series. Thus the substitution of Eqs. (2) and (3) into Eq. (1) yields

$$I_{21}(t) + \tau_{2p} dI_{21}/dt = \sum_{n=3}^{\infty} [a_n I_{n2}(t) - b_n I_{n1}(t)] \quad (4)$$

where

$$a_n = \xi_{21}/\xi_{n2}, \quad (5)$$

$$b_n = \xi_{21} A_{np2s}/\xi_{n1} A_{np1s}. \quad (6)$$

Eq. (4) interrelates the entire set of Lyman and Balmer decay curves (and the slope of Ly_α) for each point in time. Since a beam-foil experiment measures time as a flight path, t can conveniently be expressed in centimeters if τ_{2p} denotes the corresponding distance traversed by the beam in one $2p$ meanlife. The relationship is linear in the constant coefficients τ_{2p} , a_n and b_n , which can be evaluated by a linear regression. Although the number of fitting parameters is formally infinite, it can in practice reduce to a few. For example, if a sufficiently accurate velocity determination can be made, the $2p$ decay length can be fixed from the theoretical $2p$ meanlife (given by $(9/4\alpha Z)^4 a_0/c = 1.595/Z^4$). The criteria for this determination were studied by computer simulation and are discussed in the next section. With regard to the infinite sum, it is possible that the beam-foil excitation function decreases with increasing principal quantum number n strongly enough that direct cascades from the higher levels become negligible, allowing the sum to be terminated without significant error. Such a termina-

tion would not entirely neglect higher levels, since indirect cascading from them is inherently contained in the measured decay curves. Criteria for this possibility were also explored for computer simulated data, and are also discussed in the next section. However, even if direct cascades from very high lying levels are important, the number of effectively non-identical fitting parameters is quite limited, since the detection efficiencies become equal as the wavelengths converge at the Rydberg series limits. Further, the ratio A_{np1s}/A_{np2s} also converges for large n , as can be seen when it is written, using the formulae of Bethe and Salpeter [14], as

$$\frac{A_{np1s}}{A_{np2s}} = \frac{1}{2^3} \left(\frac{n^2 - 4}{n^2 - 1} \right)^3 \left[\frac{(n-1)(n+2)}{(n+1)(n-2)} \right]^{2n} \quad (7)$$

This ratio is 7.45, 7.05, 6.95 and 6.90 for $n=3, 4, 5$ and 6 , and converges to $e^4/2^3 = 6.825$ for large n . The wavelengths of the Lyman lines between Ly_γ at 973 \AA and the series limit at 912 \AA fall in a region for which the response of detectors such as the channel electron multiplier is reasonably flat, and it should therefore be possible to make the replacement

$$\sum_{n=v}^{\infty} b_n I_{n1}(t) \cong b_\infty \sum_{n=v}^{\infty} I_{n1}(t) \quad (8)$$

where v could possibly be as low as 4. The intensity summation could be obtained either by widening the bandpass of the spectrometer to include the desired lines, or by a point-by-point merging of the separately measured decay curves normalized to spectral scan intensities. A similar instrumental summation could be made for lines near the Balmer limit, provided the detection system possessed a reasonably flat response in the region of 3645 \AA .

Since Eq. (4) exposes a continuous set of linear relationships parameterized by time, there are many alternative techniques by which the measured decay curves $I_{nn'}$ can yield a discrete set of numerical relationships. For example, a set of suitably incremented time abscissae could be chosen, and numerical techniques used to obtain multipoint estimates of the local values and (for Ly_α) numerical derivatives for each. A number of numerical techniques accomplish this quite well without degrading the resolution of the data, e.g. Savitsky and Golay [15] have tabulated sets of weighting coefficients which smooth and differentiate by a local polynomial regression of a point and several of its near neighbors. An alternative but similarly directed approach is to convert the differential equation to an integral equation. Thus if we integrate both sides of Eq. (4) between the arbitrary limits T_i and T_f and exchange the orders

of integration and summation, it assumes the form (with the aforementioned terminations on the sum)

$$x(T_I, T_F) = \sum_{n=3} [a_n y_n(T_I, T_F) - b_n z_n(T_I, T_F)] \quad (7)$$

where

$$x(T_I, T_F) = \int_{T_I}^{T_F} dt I_{21}(t) + \tau_{2p} [I_{21}(T_F) - I_{21}(T_I)], \quad (8)$$

$$y_n(T_I, T_F) = \int_{T_I}^{T_F} dt I_{n2}(t), \quad (9)$$

$$z_n(T_I, T_F) = \int_{T_I}^{T_F} dt I_{n1}(t). \quad (10)$$

The integrations automatically average over fluctuations, and the subtraction in Eq. (8) can be smoothed by the techniques described above. In this case each suitably different choice of T_I and T_F will yield an independent relationship. Thus, with either the differential of integral approach, a standard linear regression (weighted, if criteria can be established) yields values for the various a_n and b_n , and if desired, τ_{2p} . These, in turn, infer population information when coupled with the decay curves, as extrapolated back to the $t=0$ foil position. Thus, from Eqs. (2) and (6) we can relate

$$b_n \frac{I_{n1}(0)}{I_{21}(0)} = \frac{N_{np}(0) A_{np2s}}{N_{2p}(0) A_{2p1s}} = \frac{N_{np}(0)}{N_{2p}(0)} \frac{2^2 3^6 n(n^2-1)}{(n^2-4)^3} \left(\frac{n-2}{n+2}\right)^{2n}. \quad (11)$$

Similarly, from Eqs. (2), (3), (5) and (6)

$$\frac{a_n}{b_n} \frac{I_{n2}(0)}{I_{n1}(0)} - 1 = \frac{N_{ns}(0) A_{ns2p} + N_{nd}(0) A_{nd2p}}{N_{np}(0) A_{np2s}} = \frac{N_{ns}(0)}{N_{np}(0)} \frac{n^2}{2^2(n^2-1)} + \frac{N_{nd}(0)}{N_{np}(0)} \frac{2^3 n^2}{5(n^2-4)} \quad (12)$$

where the transition probability ratios have been evaluated from their theoretical expressions [14]. The p -state populations are unilaterally determined by Eq. (11), while Eq. (12) determines only a weighted sum of the s - and d -state populations, weighted strongly, however, toward the d -states (10.24 to one for $n=3$, approaching 6.4 to one for large n).

Computer Simulation

In order to investigate the experimental precision necessary for accurate analysis by this method, simulated decay curves with controlled statistical errors were computer generated by the diagrammatic

mnemonic technique [16] using theoretical transition probabilities [14] and population models of the form

$$N_{nl}(0) = (2l+1) C/n^{s+2} \quad (13)$$

and

$$N_{nl}(0) = C/n^{s+1} \quad (14)$$

truncated, of computational necessity, above some value of n . The effects of this truncation can be estimated, since for $s > 1$ the untruncated models converge to a finite total population for an infinite number of states. Thus the total population of all states which cascade either directly or indirectly into $2p$ is, for both Eqs. (13) and (14), given by

$$N = \sum_{n=1}^{\infty} \sum_{l=0}^{n-1} N_{nl}(0) - N_{3p}(0) = C [\zeta(s) - 1 - 2^{-s} - 3^{-s-1}] \quad (15)$$

where $\zeta(s)$ is the Riemann Zeta-function, which is tabulated [17]. Thus one can compute, for example, that for $s=2$, 49% of the population lies above $n=6$, while for $s=3$, 21% of the population lies above $n=6$, etc. Thus a reasonable approximation of a fully populated system can be obtained from these truncated models.

The simulated decay curves were analyzed by the suggested method, and the accuracy with which the model populations were retrieved was studied as a function of the following experimental variables: (a) the time window, i.e., the total usable decay length (in ns) after the foil, (b) the statistical accuracy of the data, (c) the precision with which the beam velocity is known, (d) the number of decay curves included in the analysis, and (e) the number of fitting parameters allowed to vary. The results can be briefly summarized as follows. The minimum time window was about 20 ns, and shorter decay lengths required prohibitive statistical accuracies. Increasing the time window beyond 50 ns did not usually enhance the determination, since equal scaling times were used and statistical accuracy on the tails became low (particularly when simulated background subtractions exceeded decay intensities). In addition, when higher decay curves were omitted, extending the time window past 50 ns actually worsened the determination, as the omitted longlived states began to dominate. With an adequate time window a statistical accuracy of 1% at $t=0$ was usually sufficient to extract 10% populations in the absence of other errors, and in the presence of data omissions and velocity errors sufficient to prevent population extraction, increased statistical accuracy did not materially improve the populations.

When a fixed velocity was used, the technique proved to be very sensitive to errors in its value, and velocity errors as small as 0.2% sometimes introduced substantial errors in populations. Post-foil beam energy analyzers of 0.1% accuracy have been applied to beam-foil studies [18], but if a velocity measurement of such accuracy is not possible, the velocity should be treated as a free parameter. The neglect of higher decay curves produced sizable errors for the unnormalizable population models with $s=1$, but with $s=2$ and $s=3$ it was often possible to recover accurate populations with as few decay curves as Ly_α , Ly_β and H_α , or Ly_α , Ly_β , Ly_γ , H_α and H_β included. Predictably, as more fitting parameters were added, it became successively more difficult to accurately extract them. This indicated that the reliability of this technique would be considerably enhanced if it were possible to determine experimentally a few of the relative detection efficiencies of the higher lying lines of a given Rydberg series.

Conclusions

The results of this study indicate that, although special care must be given to some factors in a measurement intended for analysis by this technique, the required conditions should be experimentally attainable. This technique is therefore suggested as an appropriate means of correlating the Lyman and Balmer series to determine the initial population of beam-foil excited hydrogenic atoms. It would be particularly interesting to compare the results of an analysis of Ly_α , Ly_β and H_α by this technique with a similar analysis by the technique of Tielert and Bukow (treating the velocity as a free parameter) since each method has advantages and disadvantages. The former does not separately determine s - and d -state populations, but incorporates three decay curves with only three free

parameters. The latter separates the populations, but incorporates two decay curves with eight free parameters. Two of the parameters, τ_{2p} and $a_3 = \xi_{21}/\xi_{32}$ are common to both analyses, and agreement would provide an important consistency check.

References

1. Goodman, A.S., Donahue, D.J.: Phys. Rev. **141**, 1 (1966)
2. Jordan, J.A., Jr., Bakken, G.S., Yager, R.E.: J. Opt. Soc. Am. **57**, 530 (1967)
3. Bickel, W.S.: J. Opt. Soc. Am. **58**, 213 (1968)
4. Dotchin, L.W., Pegg, D.J., Chupp, E.L.: Phys. Lett. **31A**, 85 (1970)
5. Andr , H.J.: Phys. Rev. A **2**, 2200 (1970)
6. Sch rmann, D., Schlagheck, W., Heckmann, P.H., Bukow, H.H., Buttler, H. v.: Z. Physik **246**, 239 (1971)
7. Bukow, H.H., Buttler, H. v., Haas, D., Heckmann, P.H., Holl, M., Schlagheck, W., Sch rmann, R., Tielert, R., Woodruff, R.: Nucl. Instr. Methods **110**, 89 (1973)
8. Alguard, M.J., Drake, C.W.: Phys. Rev. A **8**, 27 (1973)
9. Heckmann, P.H., Bukow, H.H., Tielert, R.: Z. Physik **264**, 109 (1973)
10. Tielert, R., Bukow, H.H.: Z. Physik **264**, 119 (1973)
11. Tielert, R., Bukow, H.H., Heckmann, P.H., Woodruff, R., Buttler, H. v.: Z. Physik **264**, 129 (1973)
12. Curtis, L.J., Berry, H.G., Bromander, J.: Physica Scripta **2**, 126 (1970), cf. Section 2.2
13. Etherton, R.C., Beyer, L.M., Maddox, W.E., Bridwell, L.B.: Phys. Rev. A **2**, 2177 (1970)
14. Bethe, H.A., Salpeter, E.E.: Quantum Mechanics of One- and Two-Electron Atoms, Berlin-G ttingen-Heidelberg: Springer 1957
15. Savitsky, A., Golay, M.J.E.: Analytical Chemistry **36**, 1627 (1964)
16. Curtis, L.J.: Am. J. Phys. **36**, 1123 (1968)
17. Jahnke, E., Emde, F.: Table of Functions, New York: Dover 1945
18. Andr , H.J.: Private communication

M.Sc.D.R. Bryant
 Prof. Dr. L.J. Curtis
 Department of Physics and Astronomy
 University of Toledo
 Toledo, Ohio 43606
 U.S.A.