RECENT PROGRESS IN LIFETIME MEASUREMENTS

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Résumé - Un aperçu des récents développements de l'utilisation des faisceaux d'ions rapides pour la détermination des durées de vie atomiques est présenté. Plusieurs améliorations sont discutées et un certain nombre de succès et de difficultés d'application sont décrits.

Abstract - A review of recent developments in the use of fast ion beams for atomic lifetime measurements is presented. Several improvements in capabilities are discussed, and a number of successes as well as some difficulties in its application are described.

INTRODUCTION

Many uses for fast ion beams in atomic physics research have been developed since this series of conferences began, and our sessions now include many new and exciting topics. Nevertheless, lifetime measurements remain one of the most important applications of fast ion beam spectroscopy, not only because they are the primary source available for this type of atomic data, but also because the method is still in a developmental stage, with new adaptations occurring continually. I would like to review some of the recent improvements in these measurements, describe some of the problems which remain, and indicate the progress which has been made toward their solution.

RECENT IMPROVEMENTS

One of the recent advances involves precision. At the Gatlinburg Conference the Berlin group reported a cascade-free beam-laser lifetime measurement [1] which was accurate to within 1/4 %. The Stockholm group has since made a beam-foil measurement [2] of a favourable transition in He I which was of similar accuracy, demonstrating that the presence of cascading does not necessarily preclude a high precision measurement. However, the record for precision has now been broken by a beam-laser measurement of the $3p\ ^2P_{1/2}$ lifetime in Na I by Gaupp et al. [3] which claims an accuracy of better than 2 parts per thousand, with the major remaining uncertainty arising from the time calibration. Although this degree of precision is attainable only in very special situations, it illustrates a general trend toward higher precision, made possible through advances in instrumentation and measuring procedures.

The development of beam detection windows with very narrow spatial resolution is another important improvement. Direct measurements of lifetimes in the one picosecond range were reported already in Gatlinburg, and the techniques have since been extended and refined. For the EUV region the Laval group developed a near-beam auxiliary entrance slit modification for a grazing incidence monochromator which permits a window width as small as 80 μm [4]. For the X-ray region the Kansas State group utilised a near-beam source slit followed by a collimator slit to obtain a measured window width as small as 20 μm [5]. Alternatively, the Bochum [6] and Liège [7] groups have convoluted the experimental detection window into the fitting function to recover short lifetimes with a wider spatial resolution. By these various methods picosecond lifetimes are now being reported with uncertainties as low as 5%. At present this seems to be the lower limit for direct measurements, but shorter lifetimes can be measured by indirect methods such as non-proportional X-ray yields [8] and natural line width studies [9].

The wavelength resolution attainable in a lifetime measurement has also been improved substantially. Optical techniques which exploit the angular doppler dispersion to narrow and enhance lines from a moving source have been devised and developed by Stoner and Leavitt [10], Bergkvist [11] and Jelley et al. [12]. This has permitted separate lifetime measurements for near lying lines and has reduced line blending effects. For example, a recent measurement by Livingston and Berry [13] obtained a line width of 0.3 Å by refocusing a

concave grating VUV spectrometer to a moving source. This permitted resolution of the fine structure of doubly excited ^4P states in three electron ionisation stages of C, N and O. A detection window of 0.5 mm yielded 80 ps time resolution. The lifetimes were found to differ by an order of magnitude among the fs components of a given ion (from 0.1 to 1 ps) depending upon the availability of autoionisation channels.

New excitation methods have also been developed since Gatlinburg. As one example, Winter and Gaillard [14] obtained sharp impulsive excitation in a beam-laser decay curve, using velocity tuning to doppler-switch the resonance between superimposed laser and ion beams. Another example is the use by Ramanujam [15] of a pulsed ion beam incident upon a solid target to measure lifetimes of sputtered ions by delayed coincidence methods. This permits the study of very heavy atoms, such as uranium and lead. It is interesting that decay curves so obtained are cascade-free, presumably due to quenching of all but the smallest orbitals by the internal fields within the solid target. However only neutral atoms have been observed by this technique, illustrating an important fact - the only presently available technique to which high stages of ionisation are accessible is beam-foil excitation. Therefore the problem of cascades still confronts us.

SOME DIFFICULTIES FOR LIFETIME DETERMINATIONS

The Na I and Cu I isoelectronic sequences have recently provided a testing ground for the reliability of beam-foil measurements [16, 17]. It is either a blessing or a curse that these essentially single electron spectra are, at the same time, among the easiest to reliably calculate, and among the most difficult to measure by curve fitting techniques. These problems have been discussed in detail in the literature [16, 17], so I shall only mention here that there are a number of beam-foil measurements for ions in these sequences which have yielded values which are longer than the true lifetimes by substantially more than the quoted uncertainties. Part of the difficulty is seen from Fig.1, which is a plot of the relative lifetimes of the most important cascades into the 4p resonance level for the Cu I isoelectronic sequence. Notice that for Cu I these cascades are all longer-lived than the 4p, while for As V and above the opposite is true.

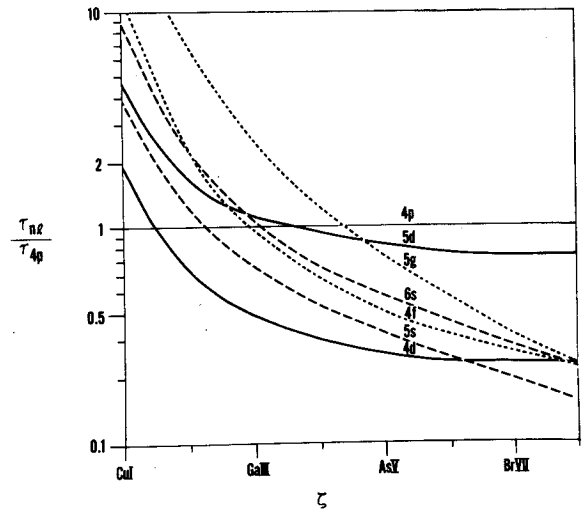

Fig. 1 - Relative cascade lifetimes into 4p for the Cu I sequence.

In the intermediate region there are many "lifetime crossings" which make lifetime extraction by exponential curve fitting difficult. For very high ionisation stages one would expect a heavy growing-in behaviour due to the short-lived cascades, but one might hope that the higher lying longer-lived levels would be sparsely populated, and would not strongly contribute [16]. This turns out to be not at all the case, as is shown in Fig. 2.

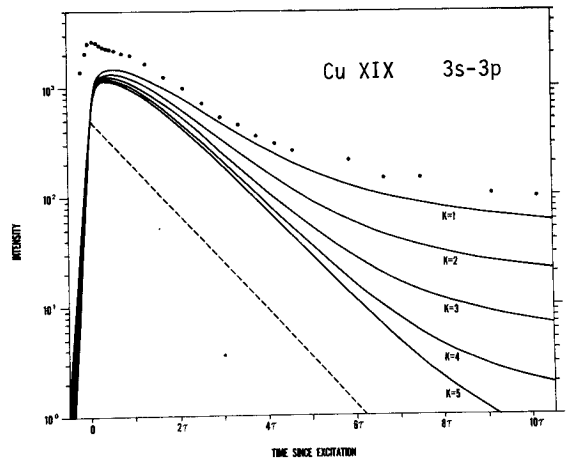

Fig. 2 - Measured and simulated decay curves for the 3p level in Cu XIX.

This is the decay curve of the 3p $^2P_{3/2}$ level in Cu XIX of the Na I sequence. The points denote measurements by Pegg et al. [18], and the lines are simulations made with a population model $(2\ell+1)/(n^*)^k$ truncated above n=25, for k=1-5. The simulation includes direct cascades from S and D levels, the yrast chain [19] and two-step indirect cascades from the lower P and F levels, convoluted

with an appropriate window function. The model is debatable, and I show the simulations here only to draw attention to one fact - there is a very long tail on the measured decay curve which is of puzzling origin. One possibility is that it arises from some sort of beam associated continuum radiation in the 300 Å wavelength region, which falls off with distance from the foil. Another possibility is that it arises from cascading from higher lying levels, which would mean that there is a much larger population of very high lying levels than we had previously suspected. This tail has a 600 ps e-folding time, and accounts for over 10% of the peak intensity. To find a 600 ps lifetime one must go above n=15 on the yrast chain and above n=30 for the S, P and D levels. Similar tails are seen on the homologous Cu I sequence transition in Kr VIII, with data of the Toledo-Chicago group [20] shown in Fig 3.

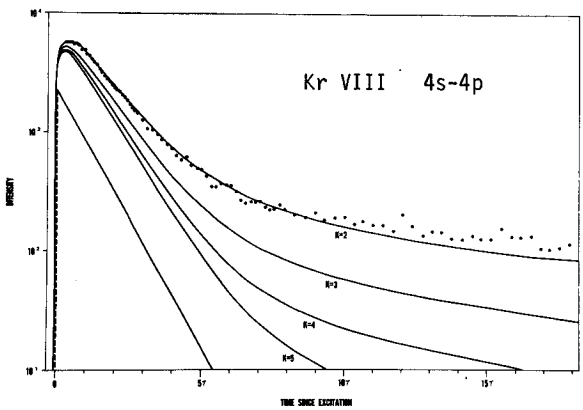

Fig. 3 - Measured and simulated decay curves for the 4p level in Kr VIII.

These simulations were extended to include S, D and yrast cascades as high as n=100 in an attempt to match the tails with a lower value of k, but such simulations were insensitive to these very high levels on the time scale shown here.

In discussing the various types of repopulation it is useful to adopt a terminology which differentiates between generically separable situations. Geologically speaking, a cascade is defined as "one of a series of small falls formed by water in its descent over rocks" [21]. A large volume of water falling sheer from a great height is a cataract, defined as "a waterfall of great size falling headlong over a precipice, thus distinguished from a cascade" [21]. We have found it convenient to refer to Rydberg transitions from high S and D levels to low P levels as cataracts, and sequential transitions such as the yrast chain as cascades.

Various methods for extraction of lifetimes from these decay curves are available, but this depends upon whether the tails arise from backgrounds, cataracts, or cascades. It has been suggested [22] that transitions from continuum states to lower lying bound states could be substantial (i.e., cataracts from the continuum). We have attempted to test this by applying small electric fields along the beam to sweep away free or loosely bound electrons, but we saw no effect on the tails of Kr VIII decay curves [20]. Ellis [23] has proposed a simple classical model which would populate levels of high n but limited ℓ, thus favouring cataracts. Some experimental effects have been noted. A dependence of decay curve tails upon beam current was reported at the Lysekil Conference [24]. In studies of chlorine, Forester et al. [25] have observed long tails in N, O and F like systems, but not on Li, Be, B or C like systems. If the tails are log-log plotted they are nearly straight lines, in accord with the power laws deduced by Hopkins and von Brentano [26] and by Ellis [23] by replacing discrete sums over high n state transitions by integrals. If these tails result from spurious backgrounds which mask the true decay curve, means can be found to discriminate against them. If they are a true manifestation of the dynamical level population, then they can be used to advantage to determine the lifetime.

JOINT ANALYSIS OF YRAST DOMINATED DECAY CURVES

If these tails are a result of heavy cascading along the yrast chain, as opposed to backgrounds or cataracts, then the analysis is extremely well suited to the technique which has become known as the ANDC method (the name arose from the "arbitrarily normalised decay curves" which are jointly analysed, but it has been suggested that "ACDC" for "analysis of correlated decay curves" would better describe the procedure). The method utilises the fact that the radiated line intensities $I_{n\ell}(t)$ are proportional to the corresponding upper level populations, and are therefore interrelated through the population rate equation. The use of such information to reduce beam-foil data

had already been suggested [27] before the first beam-foil conference and was discussed in Lysekil [28]. Its application has since been greatly simplified and extended [29]. For the Cu I isoelectronic sequence the instantaneous population equation for the 4p level can be written as

$$dI_{4p}/dt = -\alpha_{4p}I_{4p}(t) + \xi_{4d}I_{4d}(t) + \sum_{n=5}^{\infty} [\xi_{ns}I_{ns}(t) + \xi_{nd}I_{nd}(t)] \quad (1)$$

provided that all $I_{n\ell}(t)$ have a common t=0 and have been cleared of all backgrounds and blends. Here $\alpha_{4p} \equiv 1/\tau_{4p}$ is the reciprocal meanlife and the $\xi_{n\ell}$ are constant factors which renormalise the cascade and cataract decay curves relative to the 4p. Except for these constants, which can be determined from the fact that Eq(1) must hold for all values of t, $I_{4d}(t)$ contains all information about replenishment from the yrast cascades and the sum contains all information about replenishment from the $\Delta n \neq 0$ cataracts. If yrast cascades dominate, the cataract contributions will be small and Eq(1) is essentially a relationship between two decay curves, the 4p and 4d. The degree to which this is true becomes manifest if Eq(1) is rewritten

$$y = \alpha_{4p} - \xi_{4d}(x + x_o) \quad (2)$$

where the measured decay curves are used to compute the auxiliary quantities

$$y = -d(\ell n\, I_{4p})/dt \quad (3)$$

$$x = I_{4d}(t)/I_{4p}(t) \quad (4)$$

$$x_o = \sum_{n=5}^{\infty} [\xi_{ns}I_{ns}(t) + \xi_{nd}I_{nd}(t)]/\xi_{4d}I_{4p}(t) \quad (5)$$

in subsets of data points for various time regions, using numerical differentiation and averaging methods (which may include integrating both sides of Eq(1)).

A "Replenishment Plot" of y versus $(x+x_o)$ is shown in Fig. 4, from a Zn II study by the Lund-Stockholm group [30]. x_o is computed from the 5s decay curve, which was normalised to the 4d using relative population measurements of Hultberg et al. [31] (The relative normalisation need not be performed with the same detection equipment as the decay curve measurements, since it is only the ratio of the $\xi_{n\ell}I_{n\ell}(t)$ products which enters). The error bars denote statistical uncertainties which were propagated into the quantities y and $(x+x_o)$. The intercept of the fitted line with the y-axis is the reciprocal of the 4p meanlife, and the intercept with the $(x+x_o)$ axis is the reciprocal of the Replenishment Ratio R(0) [29]. The values (which are only slightly sensitive to the inclusion of the 5s) are τ_{4p}=2.1 ns and R(0)=0.34. A coulomb approximation calculation yields 2.0 ns [32] and a multiconfiguration Hartree-Fock calculation yields 2.5 ns [33]. Earlier experimental curve fitting results generally gave ∿3 ns.

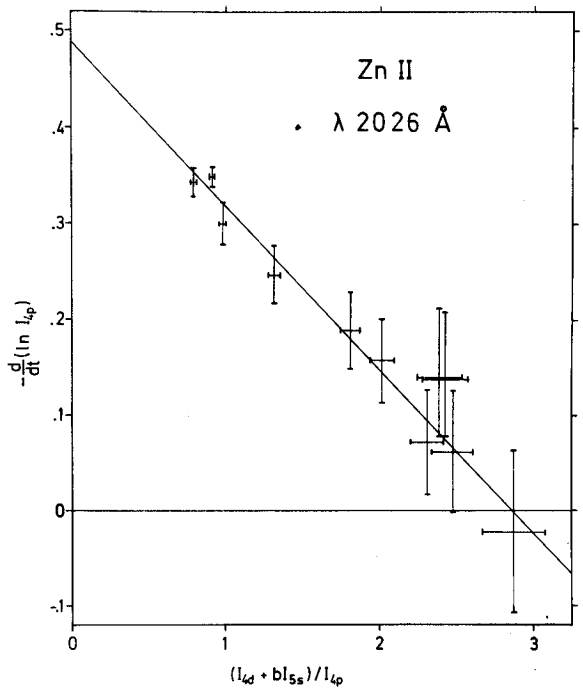

Fig. 4 - Replenishment Plot for the 4p level in Zn II.

A crude estimate of the relative importance of the various cascades into 4p can be gained from Fig. 5, which shows the principal cascade gA's relative to α_{4p} along the Cu I sequence.

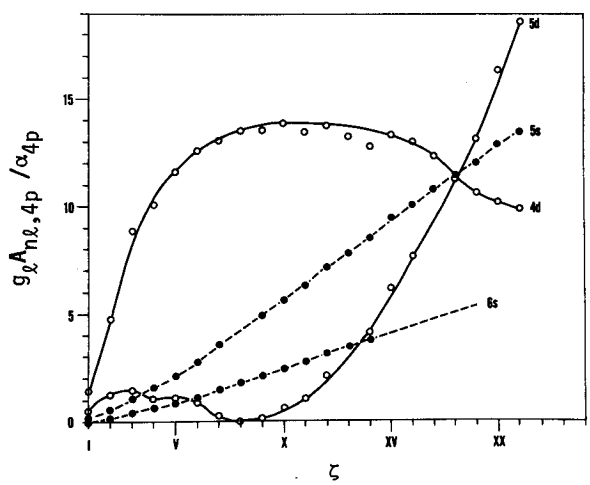

Fig. 5 - Weighted relative transition probabilities for the principal cascades into 4p for the Cu I sequence.

If populations were commensurate on this scale, the 4d would clearly dominate for the low charge stages. Notice how the 5d passes through a minimum and eventually dominates for very high charge stages. The 5d cascade is particularly interesting for reasons made clear in Fig. 6.

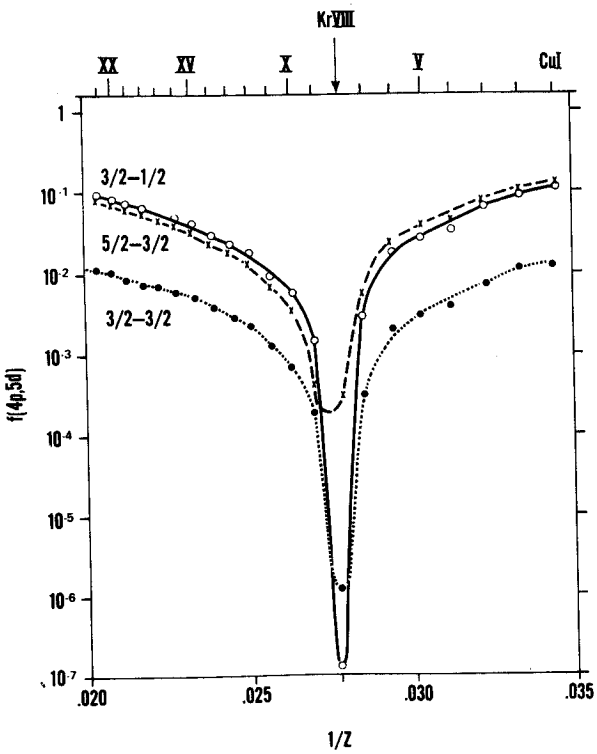

Fig. 6 – f versus 1/Z for the 4p-5d transition in the Cu I isoelectronic sequence

Due to cancellation effects in the transition integral the oscillator strength for the 4p-5d transition drops abruptly by several orders of magnitude for Kr VIII, and thus is reflected in the 4p decay curve only through its feeding of the yrast chain. Sharp cancellations such as this are not unusual in this type of spectrum, and a knowledge of their occurrence can be useful in analysing cascade effects. We have developed a simple graphical method [34] (which does not require computation of transition integrals) for determining if and where such cancellation effects will occur along the isoelectronic sequence of a given Rydberg transition . This cancellation in Kr VIII makes it an excellent candidate for ANDC analysis, since all n≤5 cascading is included in the 4d and 5s decay curves. Preliminary results from an ANDC analysis by the Toledo-Chicago group [20] are shown in Fig. 7. Although these plots include only the 4d yrast cascades, the measurements have now been improved both in statistical accuracy and by inclu-

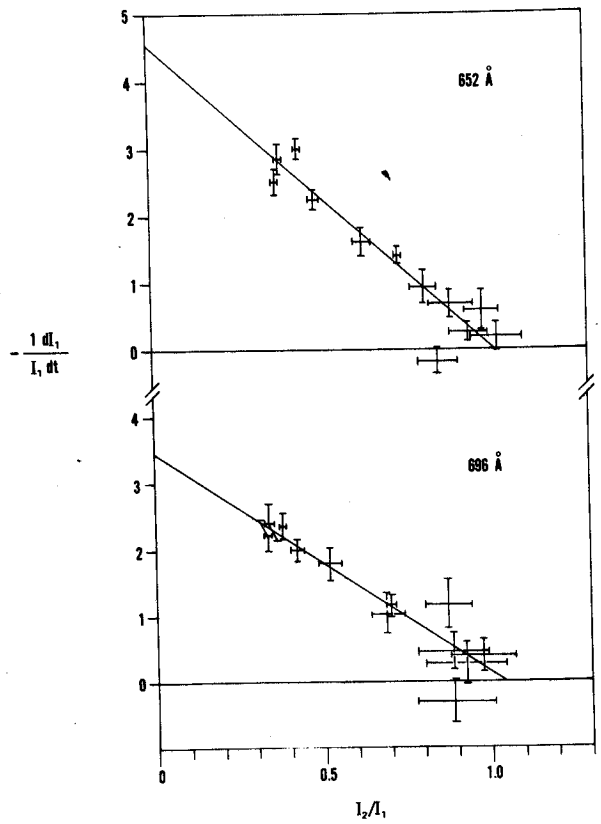

Fig. 7 – Replenishment Plot for the 4p levels in Kr VIII

sion of both the 4d and 5s cascades. Here α_{4p}, ξ_{4d} and ξ_{5s} were all left as free parameters. Since the analysis demanded a very small contribution from 5s (1/10 the 4d count rate at t=0), the results do not differ significantly from those depicted in Fig. 7, and yield τ_{4p}=220 and 290 ps for the J=3/2 and 1/2 levels, with R(0)≅1. This can be compared with MCHF calculations of 240 and 315 ps [33], while earlier curve fitted measurements were generally 40% longer. Although curve fitting analysis of these decay curves is very difficult, ANDC analysis is very easy, and succeeds even if the statistical accuracy is deliberately reduced and the 5s cascade is neglected. Unlike curve fitted results, the extracted lifetime has a validity test in the linearity of the Replenishment Plot. Thus in this case it is clear that the long tails are an essential part of the dynamical population propagated along the yrast chain, which indicates a very large population at very high values of n.

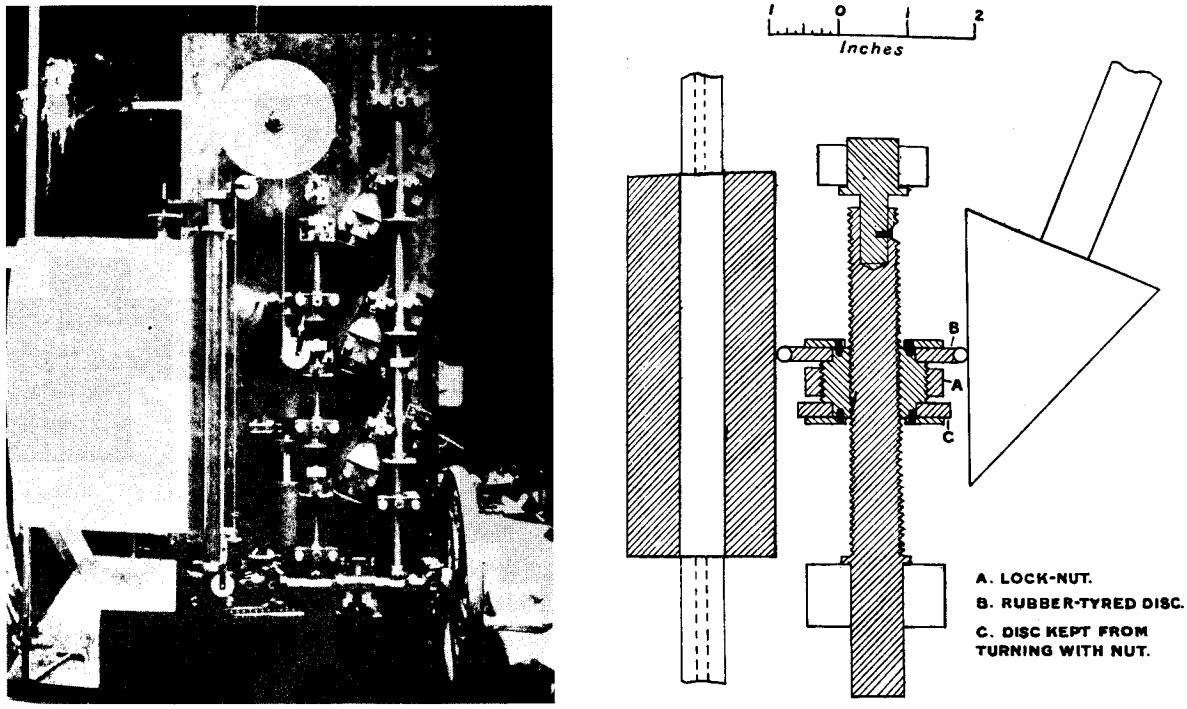

Fig. 8 - (left) Frederick Soddy's analogue computing machine [35] which simulated up to third order yrast decay curves and (right) a diagram of one of its three simulation units. The instantaneous population is represented by the height of a rubber-tyred nut on a screw. The nut is driven by a rotating cone at a speed proportional to its height above the point of the cone, while the screw is turned by gears from the nut of the next cascade unit. The nut thus moves relative to the screw and its height can be magnified and communicated to a crayon-holder by cords and pulleys.

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If, however, the substances are not independent, but are such that one of them is gradually formed from the other, we have a different system of equations to represent the reaction.

Let u, v be the residues of the substances after an interval x, $y(=u+v)$ being the total residue actually measured at that time. Let the initial values of u and v be $u=a$, $v=0$; let αu be the rate of diminution of u due to its reaction with one of the other elements of the system, and βu its rate of diminution due to its reaction with another of the elements of the system, by means of which v is formed, and let γv be the rate of diminution of v, then

$$\frac{du}{dx}=-(\alpha+\beta)u, \quad \ldots \quad (18)$$

$$\frac{dv}{dx}=\beta u-\gamma v, \quad \ldots \quad (19)$$

whence

$$u=ae^{-(\alpha+\beta)x}, \quad \ldots \quad (20)$$

$$v=\frac{a\beta}{\alpha+\beta-\gamma}\{e^{-\gamma x}-e^{-(\alpha+\beta)x}\}, \quad \ldots \quad (21)$$

$$y=\frac{a}{\alpha+\beta-\gamma}\{\beta e^{-\gamma x}+(\alpha-\gamma)e^{-(\alpha+\beta)x}\}. \quad \ldots \quad (22)$$

There are several particular cases of these equations which require to be considered separately.

Fig. 9 - Portion of the 1866 paper by Harcourt and Esson [36] which presents the "Esson Equations" for a branched repopulating cascade. The paper goes on to discuss special cases where the decay curve is a single exponential, a humped growing-in and a tailed growing-out, and presents examples of each case from chemical reaction rate measurements.

HISTORICAL FOOTNOTE

We have seen that computer simulation of multiexponential decay curves which model the excitation and detection conditions is a convenient means of studying the uncertainties which are introduced into curve fitted lifetime determinations by cascade repopulation. It is interesting that this same approach was used by Frederick Soddy in 1912 [35]. Soddy constructed a mechanical analogue computer which automatically traced out multiexponential decay curves for chosen values of populations and lifetimes. A photograph of his apparatus and a diagram explaining its operation are shown in Fig. 8. However, cascade replenishment had already been studied much earlier. In 1866 William Esson solved the cascade rate equations and, together with A. Vernon Harcourt, identified examples of what we now call "growing-in" and "growing-out" cascades [36]. A portion of their paper showing the "Esson Equations" is reproduced in Fig. 9. As early as 1795, Riche de Prony fitted two, three and four exponential sums to vapour pressure data [37, 38]. An exposition of the concept of the exponential law prior to 1900 is presented in ref. [38].

BIBLIOGRAPHY

[1] H.J. Andrä, in Beam Foil Spectroscopy, eds. Sellin & Pegg (Plenum, New York, 1976) pp 835-51

[2] G. Astner, L.J. Curtis, L. Liljeby, S. Mannervik and I. Martinson, Z. Physik A279 (1976) 1-6

[3] A. Gaupp, Th. Krist and H.J. Andrä (to be published)

[4] K.X. To and R. Drouin, Can. J. Spectrosc. 21 (1976) 21-4

[5] S.L. Varghese, C.L. Cocke and B. Curnutte, Phys. Rev. A14 (1976) 1729-34

[6] E. Träbert, H. Winter, P.H. Heckmann and H.V. Buttlar, Nucl. Instr. Meth. 135 (1976) 353-7

[7] Y. Baudinet-Robinet, H.P. Garnir, P.D. Dumont and A.E. Livingston, Physica Scripta 14 (1976) 224-9

[8] H.-D. Betz, F. Bell, H. Panke. G. Kalkoffen, M. Welz and D. Evers, Phys. Rev. Lett. 33 (1974) 807-10

[9] H.G. Berry, J. Desesquelles and M. Dufay, Phys. Lett. 36A (1971) 237-8

[10] J.O. Stoner, Jr. and J.A. Leavitt, Appl. Phys. Lett. 18 (1971) 368-9; 477-9

[11] K.-E. Bergkvist, J. Opt. Soc. Amer. 66 (1976) 837-41

[12] N.A. Jelley, J.D. Silver and I.A. Armour, J. Phys. B10 (1977) 2339-46

[13] A.E. Livingston and H.G. Berry, Phys. Rev. A17, (1978) 1966-75

[14] H. Winter and M. Gaillard, Z. Physik A281 (1977) 311-5

[15] P.S. Ramanujam, Phys. Rev. Lett. 39 (1977) 1192-4

[16] R.J.S. Crossley, L.J. Curtis and C. Froese Fischer, Phys. Lett. 57A (1976) 220-7

[17] S.M. Younger and W.L. Wiese, Phys. Rev. A17 (1978) 1944-55

[18] D.J. Pegg, P.M. Griffin, B.M. Johnson, K.W. Jones, J.L. Cecchi and T.H. Kruse, Phys. Rev. A16 (1977) 2008-10

[19] J.R. Grover, Phys. Rev. 157 (1967) 832, cf. footnote 1

[20] L.J. Curtis, R.M. Schectman, H.G. Berry and A.E. Livingston (to be published)

[21] Oxford Dictionary, Vol. II (C) (Oxford Press-1933)

[22] C.A. Nicolaides, Phys. Lett. 63A (1977) 209-10

[23] D.G. Ellis (elsewhere in these proceedings)

[24] T. Andersen, K.A. Jessen and G. Sørensen, Nucl. Instr. Meth. 90 (1970) 41-6; H. Oona and W.S. Bickel, ibid, 223-7

[25] J.P. Forester, D.J. Pegg, P.M. Griffin, G.D. Alton, S.B. Elston, H.C. Hayden, R.S. Thoe, C.R.Vane and J.J.Wright, Phys.Rev.A (in press)

[26] F. Hopkins and P. von Brentano, J. Phys. B9 (1976) 775-8

[27] J.L. Kohl, Phys. Lett. 24A (1967) 125-6

[28] L.J. Curtis, R.M. Schectman, J.L. Kohl, D.A. Chojnacki and D.R. Shoffstall, Nucl. Instr. Meth. 90 (1970) 207-16

[29] L.J. Curtis, in Beam Foil Spectroscopy, ed. Bashkin (Springer, Berlin 1976) pp 63-109

[30] I. Martinson, L.J. Curtis, S. Huldt, U. Litzén, L. Liljeby, S. Mannervik and B. Jelenkovic, Physica Scripta (in press)

[31] S. Hultberg, L. Liljeby, S. Mannervik, E. Veje and A. Lindgård (elsewhere in these proceedings)

[32] A. Lindgård, L.J. Curtis, I. Martinson and S. E. Nielsen, Physica Scripta (in press)

[33] C. Froese Fischer, J. Phys. B10 (1977) 1241-51

[34] L.J. Curtis and D.G. Ellis, J. Phys. B11 (1978)

[35] F. Soddy, Proc. Roy. Inst. Great Britain 20 (1912) 399-413

[36] A.V. Harcourt and W. Esson, Phil. Trans. Roy. Soc. London 156 (1866) 193-221

[37] G.E. Bromage (to be published)

[38] L.J. Curtis, Amer. J. Phys. 46 (1978)