APPLICATION OF SEVERAL NEW METHODS FOR EXTRACTING MEANLIVES FROM DECAY CURVES

L. J. Curtis

Research Institute for Physics, Stockholm Sweden

Introduction

We shall describe here a number of analysis techniques which we have recently utilized to extract increased information from beam-foil decay curves. At the Lysekil beam-foil conference one year ago much concern was expressed over the systematic errors which can occur if exponential curve fitted decay curves are repopulated by cascades[1]. In addition, ambiguities exist in the determination of the presence of cascading, as well as in the identification of the cascade and primary contributions to the decay curve. We have attempted to develop criteria to quantitatively specify cascade errors, to devise non curve fitting techniques which incorporate several correlated decay curves into the analysis, and to refine curve fitting techniques to reduce errors due to cascades, blends, backgrounds, and statistical fluctuations.

Cascading in Single Exponential Decay Curves

Often in lifetime studies a decay curve is observed to contain only a single dominant exponential. One usually concludes from this that the level is virtually cascade free, and that this exponential lifetime corresponds to the level under study. Curiously, even such a seemingly obvious conclusion is not rigidly valid. This can be clearly demonstrated by an example. Consider a decay chain $0 \leftarrow 1 \leftarrow 2 \leftarrow 3$, with the populations, lifetimes, and branching ratios of level i denoted by $N_i(t)$, τ_i, and b_{ij}. If, by some chance, the relative initial populations were such that

$$N_3(0)b_{32}/N_2(0) = (\tau_3-\tau_2)/\tau_2 \quad , \quad (1)$$

then the coefficient of the τ_2 term in the decay curve of $N_2(t)$ becomes identically zero, and levels 2 and 3 both decay with a single exponential. An example of such a case is shown in Fig. 1. If, by some further chance,

$$N_3(0)b_{32}b_{21}/N_1(0) = (\tau_3-\tau_2)(\tau_3-\tau_1)/(\tau_3\tau_1) \quad , \quad (2)$$

then all three levels decay with τ_3, hence

$$N_3(t)/N_3(0) = N_2(t)/N_2(0) = N_1(t)/N_1(0) = \exp(-t/\tau_3) \quad , (3)$$

and it becomes impossible to observe any manifestation of τ_1, τ_2, or even the presence of cascading,in any decay curve. Such conditions are quite achievable, and many examples could be contrived (e.g., equal populations, unbranched decays, and $\tau_1 : \tau_2 : \tau_3 = 2:3:6$, etc.). Clearly a single exponential decay curve does not, therefore, preclude cascades, nor unambiguously determine lifetimes.

Fortuitous initial populations can similarly cause the exponential term containing the primary lifetime to be absent from multi-exponential decay curves. Fig. 2 shows a cascade scheme in which the primary decay curve contains only the exponentials present in its direct cascade. Notice that, although the primary lifetime is not exhibited explicitly as an exponential, it is nonetheless manifested implicitly through the dissimilarity of cascade and primary admixtures, if both decay curves are measured. In this simple two exponential case, the primary lifetime can be extracted by a simple formula as shown. We shall discuss more powerful techniques which we have developed to extract the lifetimes in more general cases.

Since these casual dissappearances of exponential terms can occur only for prescribed values for relative initial populations, one might vary the beam energy in the hope of altering initial population ratios to test for such an effect. However, recent excitation studies seem to indicate that, at beam-foil energies, excitation functions of levels within the same ionic charge state are relatively similar, and relative populations vary rather slowly with energy.

Identification of Exponential Contributions to Decay Curves

Even in situations in which the decay curve of a level contains an exponential of its own meanlife, this meanlife cannot be unambiguously selected from among the cascade lifetimes. Only if one can exclude both indirect cascading (cascades into the cascades) and growing-in exponentials (exponential terms of negative coefficient) does it become certain that the primary lifetime corresponds to the shortest (unblended) contributing meanlife. Most analyses to date neglect the possibility of indirect cascading, in which case the only ambiguity arises from a growing-in, which may correspond either to a fast primary level of low initial population, or to a cascade which is faster than the primary. However, if one includes the almost certain possibility of indirect cascading, the primary is not necessarily the fastest lifetime, irregardless of the presence or absence of growings-in. Fig. 3 shows an example of an indirect cascade scheme in which the primary level is of intermediate lifetime, but exhibits no growing-in. Thus the practice of setting the fastest fitted exponential of positive coefficient as an upper bound to the level lifetime is not rigorously valid.

Quantitative Cascade Contributions: The Replenishment Ratio

Even if it is possible to identify the primary lifetime from among the cascades, and to remove all backgrounds and blends, the presence of cascading may still introduce systematic errors. It was therefore suggested at Lysekil that authors quote the cascading present whenever measured lifetimes are reported. However, various widely divergent quantities have been used to quantify cascading, involving ratios of exponential coefficients, radiated intensities, level populations, or excitation cross sections. A properly chosen parameter should account for the relative lifetimes and strengths of the cascade and primary contributions, and the number of cascades contributing. We have suggested[2] a new quantity which derives its meaning in the context of the population equation

$$dN_1/dt = \sum_{i=2} N_i(t)A_{i1} - N_1(t)/\tau_1 = (\text{birth rate})-(\text{death rate}) \quad . \quad (4)$$

States are "born" by cascading and "die" by radiative decay. The population dynamics are well specified by the birth/death ratio, which we have defined as the replenishment ratio $R(t)$.

$$R(t) = \tau_1 \sum_{i=2} N_i(t)A_{i1}/N_1(t) \quad (5)$$

Thus $R \ll 1$ implies little cascading, $R \cong 1$ implies heavy cascading, and $R > 1$ implies strong growing in. We have denoted as $t=0$ the earliest point on the decay curve which is unobstructed by the foil, and quoted $R(0)$ for each of our recent lifetime measurements. For a curve fit to exponential lifetimes τ_i and coefficients C_i, $R(0)$ is given by

$$R(0) = \sum_{i=2} (1-\tau_1/\tau_i)C_i \Big/ \sum_{j=1} C_j \quad (6)$$

For an intensity calibrated measurement, $R(0)$ is given by the ratio of summed input intensities to summed output intensities, so it is also a useful quantity when cascades are measured directly.

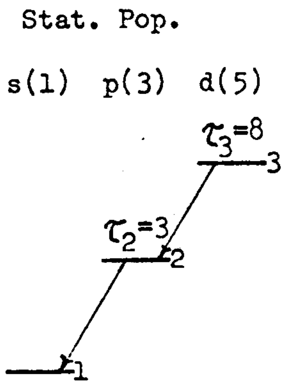


FIGURE 1. Example of a cascaded level with a single exponential decay curve. The lifetime of level 2 does not occur in the decay curve of either level 3 or 2.

$$N_2(t) = \left[N_2(0)+\frac{N_3(0)\tau_2}{\tau_2-\tau_3}\right]\exp(-t/\tau_2) + \left[\frac{N_3(0)\tau_2}{\tau_3-\tau_2}\right]\exp(-t/\tau_3)$$

$$= \underbrace{\left[3 + \frac{(5)(3)}{3-8}\right]}_{0}\exp(-t/3) + \underbrace{\left[\frac{(5)(3)}{8-3}\right]}_{3}\exp(-t/8)$$

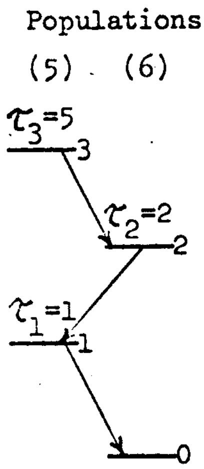


FIGURE 2. Example of a multiply cascaded level with a decay curve which does not involve its own lifetime. The lifetime of level 1 does not occur explicitly in any decay curve, but can be extracted from comparison of the admixtures.

$$\frac{N_3(t)}{N_3(0)} = (1)\exp(-t/5)$$

$$\frac{N_2(t)}{N_2(0)} = \left(\frac{7}{12}\right)\exp(-t/2) + \left(\frac{5}{12}\right)\exp(-t/5)$$

$$\frac{N_1(t)}{N_1(0)} = (0)\exp(-t) + \left(\frac{7}{10}\right)\exp(-t/2) + \left(\frac{3}{10}\right)\exp(-t/5)$$

$$\left[\frac{C_3}{C_2}\right]_{N_1} = \frac{1-\tau_1/\tau_3}{1-\tau_1/\tau_2}\left[\frac{C_3}{C_2}\right]_{N_2}$$

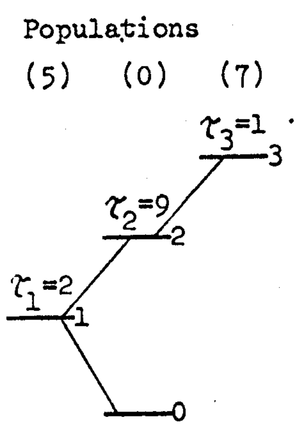


FIGURE 3. Example of a decay curve in which a cascade is the fastest contributing lifetime, but exhibits no growing in.

$$\frac{N_1(t)}{N_1(0)} = \left(\frac{7}{20}\right)\exp(-t) + \left(\frac{4}{20}\right)\exp(-t/2) + \left(\frac{9}{20}\right)\exp(-t/9)$$

INDIRECT CASCADE	PRIMARY LEVEL	DIRECT CASCADE

Integrated Decay Curve Cascade Analysis

Many of the problems discussed earlier can be eliminated if the decay curves of the contributing cascades can also be measured, and incorporated into the analysis. Since few beam-foil laboratories possess an intensity calibrated detection system over a wide wavelength range, one usually measures arbitrarily normalized decay curves (ANDC) and care must be exercised in combining them. However, since they contain no normalization information, the decay curves can be measured in any convenient branch.

We have deduced a novel analysis technique[3] by noting that the ANDC of a level k, denoted by $I_k(t)$, is proportional to its instantaneous level population

$$I_k(t) \propto N_k(t) \tag{7}$$

with a proportionality constant which depends upon the detection efficiency, the meanlife, and the branching ratio. If the ANDC of Eq(7) are substituted into Eq(4), and the constants factored together and denoted by a set of undetermined multipliers ξ_i, the equation can be written

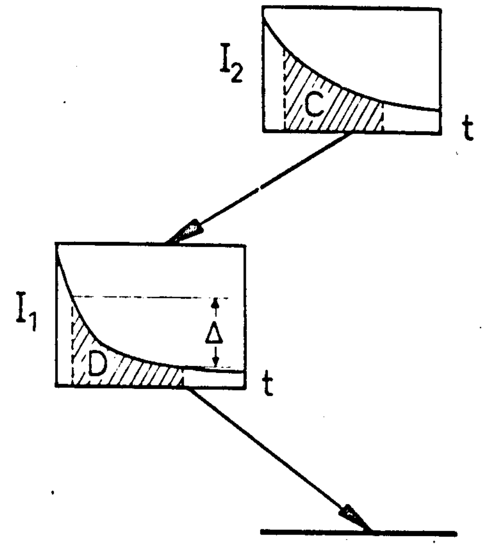
$$\tau_1 dI_1/dt = \sum_{i=2} \xi_i I_i(t) - I_1(t) \quad . \tag{8}$$

The undetermined multipliers can be related to the replenishment ratio, since clearly

$$R(0) = \sum_{i=2} \xi_i I_i(0)/I_1(0) \quad . \tag{9}$$

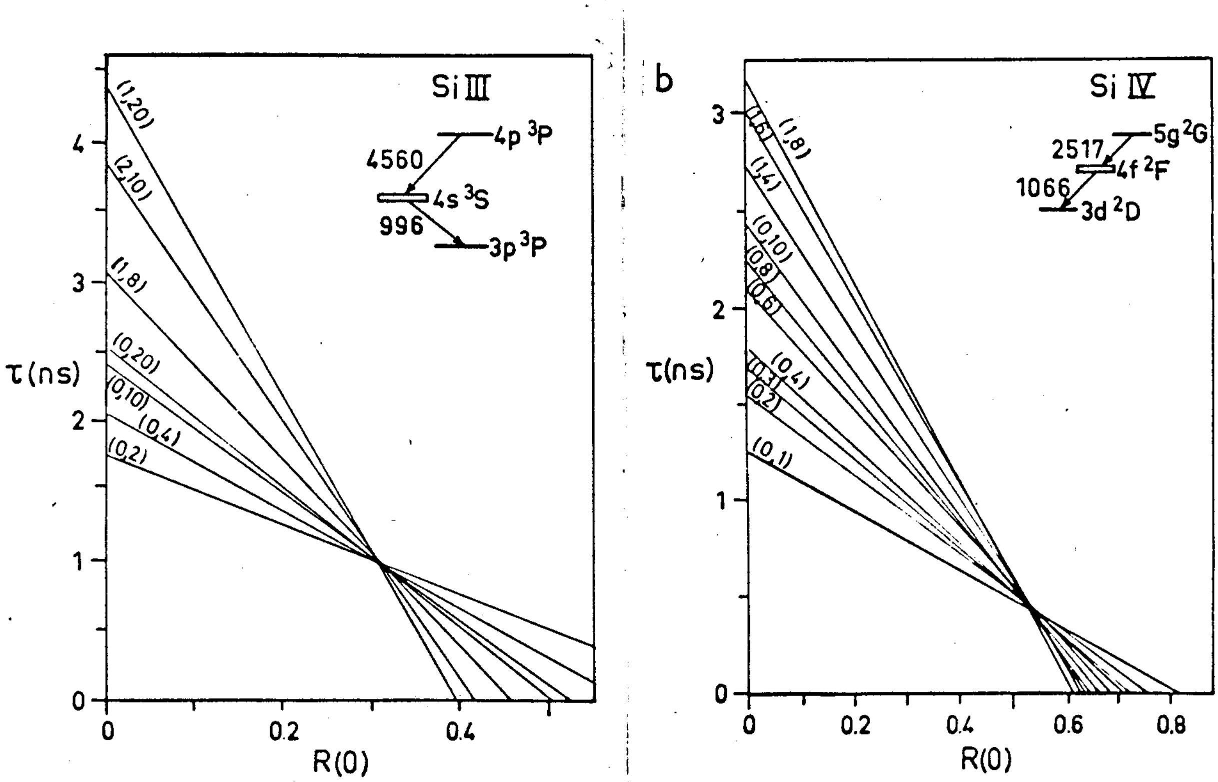
If the ANDC are all measured, Eq(7) provides a linear relationship between τ_1 and the ξ_i for each instant of time, and thus permits their determination. For ease of solution, we integrate both sides of Eq(8) between arbitrary limits t_I and t_F, which can be varied to provide a sufficient number of independent relationships. A schematic representation of the technique applied to a singly cascaded level is shown in Fig. 4. Such a simple two parameter system of equations can be solved graphically, and examples are shown in Figs. 5a and 5b. The sharpness of the intersection point verifies that the most important cascade has been correctly identified. A diffuse intersection would imply that other important cascades have been neglected, and non-intersecting lines would indicate that the cascade used is either unimportant or misidentified.

FIGURE 4. Schematic representation of the integrated decay curve cascade analysis of a singly cascaded level. The lifetime τ_1 and the relative normalization ξ of the two curves are linearly related through determinable parameters as shown.



$$\tau_1 = \left[\frac{D}{\Delta}\right] - \left[\frac{C}{\Delta}\right]\xi$$

FIGURE 5. Graphical solution of the linear relationships generated as shown in Fig. 4. The integration limits are indicated for each line, and their intersection determines their simultaneous solution.



This method can be applied to multiply cascaded levels through inversion of the resulting coupled linear equations. However, we have found that many doubly cascaded levels can also be handled graphically if each cascade dominates in a limited portion of the decay curve.[4] Fig. 6 shows schematically a situation in which a fast cascade dominates the early repopulation, then becomes depleted and a slower, initially weaker cascade becomes dominant. Separate single cascade analyses can be performed in the early and late regions, and compared for consistency.

Deconvoluting Unresolved Blends

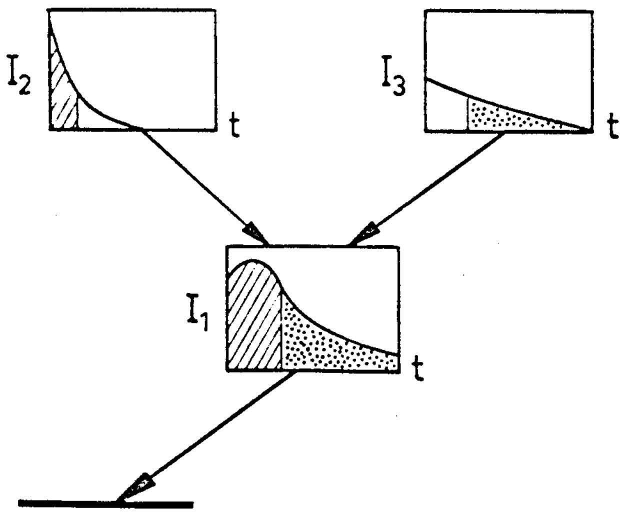
A slight variation of this method can be used to deconvolute unresolved blends in cascade decay curves, if the blending admixture can somehow be varied (e.g., by measuring at two different positions across the line profile, or in two different grating orders, or by measuring two different branches of the same level, etc.). If two different (arbitrarily normalized) admixtures of a desired and an undesired decay curve are subtracted with undetermined multipliers, there exist values of the multipliers for which the undesired contribution vanishes, and the desired contribution becomes correctly normalized. If the desired decay curve is the dominant repopulator for another measured level, the appropriate multipliers can be determined by the cascade analysis described above. An example of the application of this technique to a line masked by another line adjacent to it is shown in Fig. 7. Another example involving three degenerate lines, one measurable in another branch, is shown in Fig. 8.

Differentiation and Integration of Decay Curves

The early and late portions of a decay curve are usually quite different in information content. The early portions contain short-lived exponentials and have high statistical accuracy, while the late portions contain long-lived exponentials and have low statistical accuracy. If the decay curve is of the form

$$I_1(t) = \sum_{i=1} C_i \exp(-t/\tau_i) \qquad (10)$$

FIGURE 6, Early-late cascade analysis



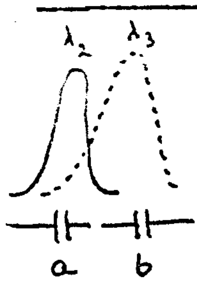


FIGURE 7. Deconvoluting a blend with two measurable admixtures.

Measured decay curves

$$I_a \propto I_2 + aI_3$$

$$I_b \propto I_2 + bI_3$$

Undetermined Multipliers

$$\xi I_a - \eta I_b \qquad (\xi - \eta)I_1 + \underset{\longrightarrow 0}{(\xi a - \eta b)}I_2$$

Determined by

$$\tau_1 dI_1/dt = \xi' I_2 - I_1 = \xi I_a - \eta I_b - I_1$$

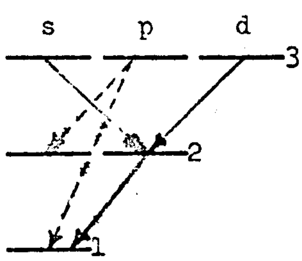


FIGURE 8. Deconvulting a blend with two branches.

Measured decay curves

$$I_{H\alpha} \propto I_{3s} + aI_{3p} + I_{3d}$$

$$I_{L\beta} \propto I_{3p}$$

Undetermined multipliers

$$\xi I_{H\alpha} - \eta I_{L\beta} \propto \xi(I_{3s}+I_{3d}) + \underset{\longrightarrow 0}{(\xi a - \eta)}I_{3p}$$

Determined by

$$\tau_{2p} dI_{2p}/dt = \xi(I_{3s}+I_{3p}) - I_{2p} = \xi I_{H\alpha} - \eta I_{L\beta} - I_{L\alpha}$$

then the derivative is

$$dI_1/dt = \sum_{i=1} (C_i/\tau_i)\exp(-t/\tau_i) \tag{11}$$

and the integral is

$$\int_t^{\infty} dt' I_1(t') = \sum_{i=1} (C_i\tau_i)\exp(-t/\tau_i) \tag{12}$$

Thus, if the early portion of the decay curve possesses sufficient statistical accuracy to permit reliable numerical differentiation (perhaps successively several times) , the resulting curve will proportionately reduce the contributions of longer lifetimes, and eliminate completely the necessity of a background subtraction.[5] Also, numerical integration of the late portion of the decay curve will proportionately increase the relative contribution of the longer lifetimes, and the integration process should smooth out statistical fluctuations[6]. Thus curve fits for the integrated and differentiated decay curves are often simpler and more reliable than those for the original decay curve. An example of our application of this technique is shown in Fig.9.

Meanlives from Logarithmic Derivatives and Replenishment Ratios

From the definition of the replenishment ratio in Eq(5), Eq(4) can be rewritten

$$dN_1/dt = [R(t)-1]N_1(t)/\tau_1 \tag{13}$$

hence we can write the meanlife as

$$\tau_1 = [1-R(t)] / [-d(\ln N_1)/dt] \quad . \tag{14}$$

For decay curves of sufficient statistical accuracy, numerically computed logarithmic derivatives near t=0 can provide an upper limit to the meanlife[7] if one assumes the primary level is the fastest contributor. Further, in many cases the value of R(t) obtained from a curve fit is only weakly sensitive to the value of τ_1 obtained in the fit, and can be corrected in an iterative fashion, using Eq(7). Thus even though both the logarithmic derivative and the replenishment ratio vary greatly as a function of distance from the foil, their use in Eq(14) may provide reliable lifetimes. Table I illustrates a measurement of a lifetime too short to be extracted by standard curve fits.

FIGURE 9. Curve fits to the measured decay curve and its numerical integral and derivative for the 4s ^{4}P level in P I. All fits contain the same exponential lifetimes, but the admixtures vary in proportion to the relative lifetimes.

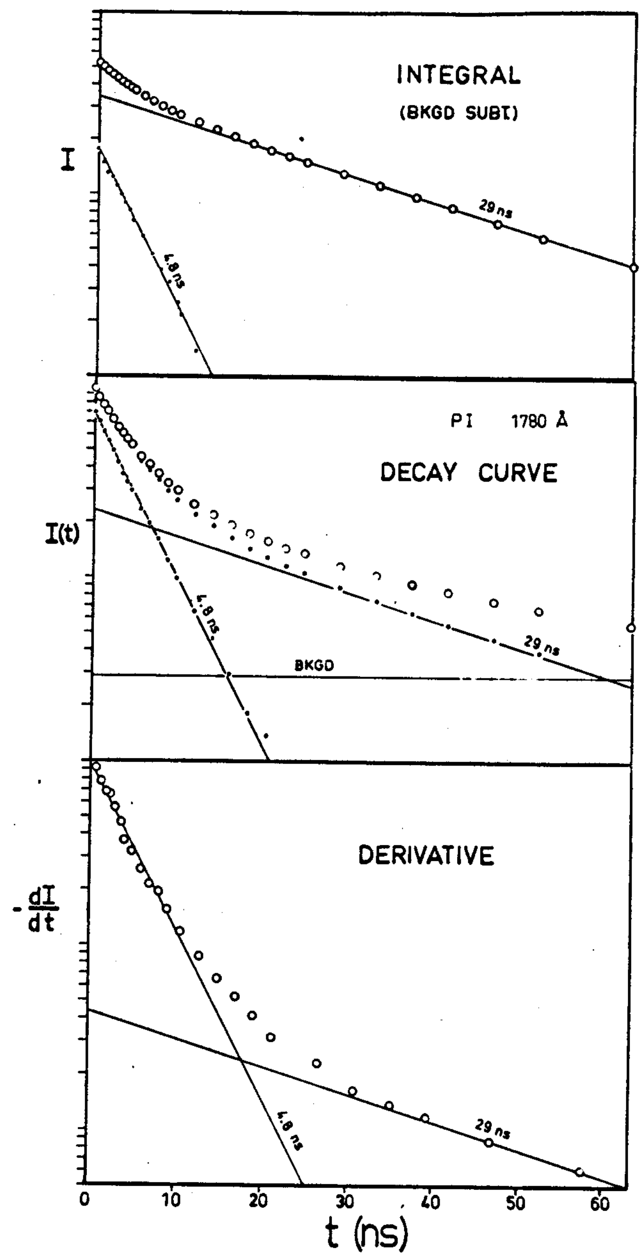


Table I. Determination of the meanlife of the 3s3p ^{1}P term in P IV from values of the logarithmic derivative and the replenishment ratio at various distances from the foil.

x	R(t)	$-d(\ln I_1)/dt$	τ_1
0.125 mm	0.222	3.58 ns^{-1}	0.217 ns
0.375	0.400	3.73	0.219
0.625	0.603	1.82	0.221
0.875	0.760	0.94	0.255
1.125	0.820	0.59	0.264
1.375	0.900	0.52	0.192
1.625	0.905	0.39	0.230
1.875	0.918	0.38	0.242
2.125	0.920	0.30	0.265

Decay of Polarization

It has been observed that radiation emitted after beam-foil excitation may exhibit appreciable polarization, which implies that magnetic substates are not equally populated by the source. It can be shown[8] that states differing only in magnetic quantum number must have the same lifetime, so the interpretation of level lifetimes does not require "natural excitation". However, decay curves $I_1^{\parallel}(t)$ and $I_1^{\perp}(t)$, viewed through polarizing filters oriented parallel and perpendicular to the beam axis, may contain different admixtures of primary and cascade exponentials. Therefore, Gordon Berry, Jean-Louis Subtil, and I have considered the use of polarization measurements to extract lifetimes from cascaded decay curves.

A field-free, selectively excited sample should have the polarization of its radiation washed out by two processes: decay, which should depopulate magnetic substates non-selectively but in proportion to their instantaneous sub-populations; and cascades, which should repopulate in some manner dependent upon higher level sub-populations and selection rules, but independent of primary level sub-populations. Thus, if we could assume either that the cascade levels are uniformly sub-populated, or that there is no coherence between the σ and π transitions in the cascades and those in the primary decays, then the cascades should affect both polarization decay curves in exactly the same way. If we then subtract the two (relatively normalized) polarization decay curves, the cascade contributions, if present, would cancel. Thus the difference between polarization decay curves would be washed out only through decay depopulation, and, unless there are fine structure complications[9], would decay as a single exponential of the primary lifetime. Thus

$$I_1^{\parallel}(t)/E^{\parallel} - I_1^{\perp}(t)/E^{\perp} \propto \exp(-t/\tau_1) + (\text{cascade cancellations} \to 0) \qquad (15)$$

The instrumental polarizations, $E^{\parallel}$ and $E^{\perp}$ can be determined either by observing an unpolarized S-state, or by comparing the unpolarized background levels on the tails of the decay curves. In preliminary studies we have measured transitions with polarization decay curves which differ by 25% and which, although heavily cascaded, yield a single exponential decay curve upon subtraction.

REFERENCES

1. Proceedings of the 2nd Internat. Conf. on BFS, Nucl. Inst. and Meth. 90, (1970), cf. papers presented by Wiese, Curtis, Gaillard, Bickel and Oona, and Kay.
2. L. J. Curtis, H. G. Berry, and J. Bromander, Physica Scripta 2,216(1970).
3. L. J. Curtis, H. G. Berry, and J. Bromander, Physics Letters 34A,169(1971).
4. H. G. Berry, J. Bromander, L. J. Curtis, and R. Buchta, to be published in Physica Scripta.
5. L. J. Curtis, I. Martinson, and R. Buchta, to be published in Physica Scripta.
6. W. R. Bennett, Jr., Advances in Quantum Electronics, p 28, Columbia University Press, 1961.
7. A. Denis, J. Desesquelles, M. Dufay, and M. C. Poulizac, Proceedings of the 1st Internat. Conf. on BFS, p 363, Gordon and Breach, 1968.
8. S. Heron, R. W. P. McWhirter and E. H. Rhoderick, Proc. Roy. Soc A 234, 565(1956) and E. U. Condon and G. H. Shortley, "The Theory of Atomic Spectra", Cambridge University Press 1935, p 72.
9. J. Macek, Phys. Rev. 1,618(1970).