Convolution of a time-dependent driven excitation into the decay curve of an arbitrarily cascaded and blended level

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An analysis is presented of the instantaneous emission rate from an atomic level that is populated by arbitrary conditions of both collisional excitation and cascade repopulation. The general solution to the rate equations is obtained in terms of transition probabilities, excitation cross sections, and a simple integral operator, and applications are given for the specific examples of impulsive, stepwise, rectangular, gaussian, and modulated excitation conditions.

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Most techniques for direct measurement of atomic (or nuclear) mean lives involve excitation of the system from a stable state by an external stimulus $Q(t)$, and observation of some quantity indicative of the instantaneous population $N_n(t)$ of an excited level n. In some cases, such as beam–foil and pulsed-beam–gas excitation, $Q(t)$ undergoes a sharp cutoff, after which the free decay can be observed. In other cases, such as the modulated-beam phase-shift technique and the stepwise approach to equilibrium, observations are made simultaneous to the excitation process. In either case, the mean-life analysis must include all effects of cascade repopulation of the level. We have shown earlier that, for the free decay, $N_n(t)$ can be decomposed into a sum of terms, grouped according to the number of steps involved in the various contributing cascade processes, which can be generated by use of a convenient mnemonic.[1] However, in studies of driven decay processes, cascade effects have generally been neglected or only partially included, a deficiency that could be particularly serious in cases where mean lives are too short to be observable in free decay. For this reason, we have generalized the cascade-decomposition procedure to include driven excitations, and have developed a formalism by which the instantaneous radiation from an arbitrarily driven, arbitrarily cascaded and blended level system can be specified.

For an arbitrarily cascaded level n that is excited with a cross section σ_n by an excitation $Q(t)$, the instantaneous population $N_n(t)$ is (neglecting collisional de-excitation and radiation trapping) governed by

$$dN_n/dt = \sigma_n Q(t) + \sum_j N_j(t) A_{jn} - N_n(t) \alpha_n, \quad (1)$$

where A_{jn} is the transition probability for a cascade from level j and α_n is the inverse mean life of level n. We assume that $N_n(-\infty) = 0$ for all levels except the ground state. Equation (1) has the integrating factor $\exp(\alpha_n t)$, and can be rewritten as

$$\frac{d}{dt}[N_n(t) \exp(\alpha_n t)] = \exp(\alpha_n t)[\sigma_n Q(t) + \sum_j N_j(t) A_{jn}]. \quad (2)$$

The solution can thus be written rather concisely in terms of an integral operator L, defined as[2]

$$L_j(t-T):f(T) \equiv \int_{-\infty}^{t} dT \exp[-\alpha_j(t-T)]f(T) \quad (3)$$

and the solution of the population differential equation is given by

$$N_n(t) = \sigma_n L_n(t-T):Q(T) + \sum_j A_{jn} L_n(t-T):N_j(T). \quad (4)$$

A similar equation also holds for each of the cascade levels. To obtain the cascade series expansion, we first neglect indirect cascades, and use Eq. (4) to compute the populations of the direct cascades, given by

$$N_j(t) \simeq \sigma_j L_j(t-T):Q(T). \quad (5)$$

We then substitute this into Eq. (4), to obtain

$$N_n(t) \simeq \sigma_n L_n(t-T):Q(T)$$
$$+ \sum_j \sigma_j A_{jn} L_n(t-t'):L_j(t'-T):Q(T), \quad (6)$$

which is the first-order cascade approximation. For the next iteration, we assume that the cascade level $N_j(t)$ has the form of Eq. (6), substitute this into Eq. (4), and obtain the second-order cascade approximation. This process is repeated until the highest-order contributing cascade level is included, at which point the expression becomes exact. Thus the population is given by the operator expression

$$N_n(t) = [\sigma_n L_n(t-T): + \sum_j \sigma_j A_{jn} L_n(t-t'):L_j(t'-T):$$
$$+ \sum_k \sum_j \sigma_k A_{kj} A_{jn} L_n(t-t'):L_j(t-t''):$$
$$\times L_k(t''-T): + \cdots]Q(T). \quad (7)$$

These nested L integral operators have a closed algebra,

and, as is demonstrated in the Appendix,

$$L_k(t-t'):L_j(t'-T):Q(T)$$

$$=\frac{L_k(t-T):Q(T)}{\alpha_j-\alpha_k}+\frac{L_j(t-T):Q(T)}{\alpha_k-\alpha_j}. \quad (8)$$

[In the special case $\alpha_k=\alpha_j$ the right-hand side becomes $-\partial(L_k:Q)/\partial\alpha_k$.] Thus the products of L operators can be rewritten as sums of the same L operators with appropriate coefficients, reducing the solution of the problem to the evaluation of a single representative integral. For added generality, we compute the radiated intensity $I_{nf}(t)=N_n(t)A_{nf}$, and sum over any levels n and f that are not resolved, thus including blending also. The final result is given by

$$I_{nf}(t)=\sum_{\text{blends}}A_{nf}\Bigg\{\sigma_nL_n:Q$$

$$+\sum_j\sigma_jA_{jn}\Bigg[\frac{L_j:Q}{(\alpha_n-\alpha_j)}+\frac{L_n:Q}{(\alpha_j-\alpha_n)}\Bigg]+\sum_k\sum_j\sigma_kA_{kj}A_{jn}$$

$$\times\Bigg[\frac{L_k:Q}{(\alpha_j-\alpha_k)(\alpha_n-\alpha_k)}+\frac{L_j:Q}{(\alpha_k-\alpha_j)(\alpha_n-\alpha_j)}$$

$$+\frac{L_n:Q}{(\alpha_k-\alpha_n)(\alpha_j-\alpha_n)}\Bigg]+\cdots\Bigg\}. \quad (9)$$

The level population $N_n(t)$ is the factor in braces. This is the most-general solution of Eq. (1), and accounts exactly for an arbitrary complexity of conditions of excitation, cascading, and blending. Explicit solution requires only the evaluation of $L:Q$ for a given analytic or empirical excitation function $Q(t)$. We list below the results for several such examples.

Example 1 Impulsive excitation,

$$Q(t)=Q_0\delta(t). \quad (10)$$

It is obvious from Eq. (3) that, for such excitation

$$L_j:Q=Q_0\exp(-\alpha_jt) \quad (t>0)$$
$$=0 \quad (t<0). \quad (11)$$

Substitution of this into Eq. (9) yields an expression that, for $t>0$, corresponds to free decay, and accordingly agrees with the results given in Ref. 1, with $N_j(0)=\sigma_jQ_0$.

Example 2 Stepwise excitation,

$$Q(t)=Q_0 \quad (t>0)$$
$$=0 \quad (t<0). \quad (12)$$

Since a step function is the integral of a δ function, it is equally obvious that, for this case

$$L_j:Q=Q_0[1-\exp(-\alpha_jt)]/\alpha_j \quad (t>0)$$
$$=0 \quad (t<0). \quad (13)$$

This expression generalizes the relationship for the approach to equilibrium under constant bombardment, which is described in many elementary textbooks[3] for the uncascaded case.

Example 3 Rectangularly pulsed excitation,

$$Q(t)=Q_0 \quad (0<t<a)$$
$$=0 \quad (0>t>a). \quad (14)$$

This is a combination of examples 1 and 2, and the solution is given by

$$L_j:Q=Q_0[\exp(\alpha_ja)-1]\exp(-\alpha_jt)/\alpha_j \quad (t>a)$$
$$=Q_0[1-\exp(-\alpha_jt)]/\alpha_j \quad (0<t<a)$$
$$=0 \quad (t<0). \quad (15)$$

Example 4 Gaussian excitation,

$$Q(t)=\frac{Q_0\exp(-t^2/2w^2)}{(2\pi)^{\frac{1}{2}}w}. \quad (16)$$

This integration yields the result

$$L_j:Q=\tfrac{1}{2}Q_0\exp[-\alpha_jt+w^2\alpha_j^2/2]\Bigg[1+\text{erf}\Bigg(\frac{t-w^2\alpha_j}{\sqrt{2}w}\Bigg)\Bigg]. \quad (17)$$

Note that this reduces to example 1 for $w\ll1/\alpha_j$.

Example 5 Modulated excitation,

$$Q(t)=Q_0+M\cos\omega t$$
$$=Q_0+M\,\text{Re}[\exp(i\omega t)]. \quad (18)$$

For this case, the integration yields

$$L_j:Q=Q_0/\alpha_j+M\,\text{Re}[\exp(i\omega t)/(\alpha_j+i\omega)]. \quad (19)$$

For this type of excitation, the measured quantity is usually the phase shift of the emitted radiation relative to that of the driver. For this reason it is not convenient to substitute the real part of this expression directly into Eq. (9), because this would yield a sum of partial phase shifts embedded in trigonometric functions. Therefore, we choose instead to retain the nested complex notation until the final equation, thus displaying the total phase shift explicitly. Since both the modulated and dc terms are separately eigenfunctions of the operator L, the solution is obtained from Eq. (7) with operators replaced by eigenvalues, so the intensity is

$$I_{nf}(t)=\sum_{\text{bl}}A_{nf}Q_0\Bigg[\frac{\sigma_n}{\alpha_n}+\sum_j\frac{\sigma_jA_{jn}}{\alpha_j\alpha_n}+\sum_k\sum_j\frac{\sigma_kA_{kj}A_{jn}}{\alpha_k\alpha_j\alpha_n}+\cdots\Bigg]$$

$$+\text{Re}\Bigg\{\sum_{\text{bl}}A_{nf}M\Bigg[\frac{\sigma_n}{(\alpha_n+i\omega)}+\sum_j\frac{\sigma_jA_{jn}}{(\alpha_j+i\omega)(\alpha_n+i\omega)}$$

$$+\sum_k\sum_j\frac{\sigma_kA_{kj}A_{jn}}{(\alpha_k+i\omega)(\alpha_j+i\omega)(\alpha_n+i\omega)}+\cdots\Bigg]$$

$$\times\exp(i\omega t)\Bigg\}. \quad (20)$$

The phase shift is given by the argument of the factor that multiplies $\exp(i\omega t)$. In the special case where only direct cascades contribute, this result agrees with that of Lawrence and Savage,[4] and permits the inclusion of arbitrary indirect cascading and blending as well.

In addition to analytic functions such as these, convolutions of experimentally measured excitation curves can be obtained by numerical integration. Lawrence[5] has developed a computer program that numerically forms the convolution of the "prompt" curve of his pulsed-electron-excitation source with a two-exponential fitting function, thus determining a singly cascaded or blended mean life, using both the excitation and free-decay data.

Thus Eq. (9), together with the convolution of the excitation, provides a surprisingly simple description of a driven atomic, molecular, or nuclear level system. This permits the determination of means lives and excitation cross sections as fitting parameters for measured driven intensities, in much the same way as is often done for free decay. This could increase the amount of data utilized in a given measurement, and might also provide access to some states too short-lived to be observable in free decay. Conversely, this description also allows the prediction of instantaneous populations and emitted intensities for level systems of known transition probabilities and excitation cross sections, under various conditions of driven excitation.

APPENDIX

Using the definition of the L operator given in Eq. (3), we consider the integral

$$L_k : L_j : Q = \int_{-\infty}^{t} dt' \exp[-\alpha_k(t-t')]$$

$$\times \int_{-\infty}^{t'} dT \exp[-\alpha_j(t'-T)]Q(T)$$

$$= \exp(-\alpha_k t) \int_{-\infty}^{t} dt' \exp[(\alpha_k-\alpha_j)t']$$

$$\times \int_{-\infty}^{t'} dT \exp(\alpha_j T)Q(T).$$

Integrating by parts, we obtain

$$L_k : L_j : Q = \exp(-\alpha_k t)$$

$$\times \left\{ \left[\frac{\exp[(\alpha_k-\alpha_j)t']}{(\alpha_k-\alpha_j)} \int_{-\infty}^{t'} dT \exp(\alpha_j T)Q(T) \right]_{t'=-\infty}^{t'=t} \right.$$

$$\left. - \int_{-\infty}^{t} dt' \frac{\exp[(\alpha_k-\alpha_j)t']}{(\alpha_k-\alpha_j)} \exp(\alpha_j t')Q(t') \right\},$$

which reduces to

$$L_k : L_j : Q = (L_j : Q - L_k : Q)/(\alpha_k-\alpha_j).$$

The special case $\alpha_k=\alpha_j$ is readily obtained by the application of l'Hopital's rule, which yields

$$L_k : L_k : Q = -\partial(L_k : Q)/\partial\alpha_k.$$

REFERENCES

[1] L. J. Curtis, Am. J. Phys. **36**, 1123 (1968).
[2] This is actually a Laplace transform with a shifted kernel, but we shall not use its inversion properties in the solution of this problem, considering instead its less-common application to multiple integrals. To make clear what each of a set of nested operators acts upon, we have adopted a notation in which a colon denotes an integral operation over a dummy variable [T in Eq. (3)] which occurs on both sides of the colon.
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[4] G. M. Lawrence and B. D. Savage, Phys. Rev. **141**, 67 (1966).
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