Derivation of the universal decay cascade distribution

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A detailed derivation of the decay cascade probability distribution stated in Eqs. (4)-(6) and (11) of Phys. Rev. Lett. 104, 186805 (2010) [arXiv:0901.4102] by Kashcheyevs and Kaestner is provided. Recurrence relations are solved explicitly and connections between solutions in different limits are demonstrated.

I. MATHEMATICAL DEFINITION OF THE DECAY CASCADE MODEL

The probability distribution $P_n(t)$ is governed by the equation

$$\frac{dP_n(t)}{dt} = -\Gamma_n(t)P_n(t) + \Gamma_{n+1}(t)P_{n+1}(t),$$  \hspace{1cm} (KK-1)

for $n \geq 0$ and with $\Gamma_0 \equiv 0$. Normalization and initial conditions are

$$\sum_{n=0}^{N} P_n(t) = 1, \hspace{1cm} (1)$$

$$P_n(t_0) = \begin{cases} 1, & n = N \\ 0, & n \neq N. \end{cases} \hspace{1cm} (KK-2)$$

Equation (KK-1) is a general kinetic equation for a birth-death Markov process for time- and population-size-dependent rates. Here we will be interested in the asymptotic values of $P_n$ as the transition rates $\Gamma_n$ gradually decrease to zero as function of time.

The original publication discussing solutions to the above model in the context of dynamic quantum dot initialization is denoted KK; equations marked here as KK-1, KK-2 etc. match the equations in KK with the corresponding numbers.

II. IMPLICIT EXACT SOLUTION

In KK, the following exact iterative solution, valid for $t > t_0$, is presented

$$P_n(t) = \int_{t_0}^{t} e^{-\int_{t_0}^{t'} \Gamma_n(\tau)d\tau} \Gamma_{n+1}(t') P_{n+1}(t') dt', \hspace{1cm} (KK-3)$$

$$P_{N+1}(t) = \delta(t-t_0)/\Gamma_{N+1}(t_0). \hspace{1cm} (2)$$

Condition (2) is introduced formally, in order for the general formula (KK-3) to accommodate the initial condition (KK-2); the delta functions are regularised as $\int_{t_0}^{t} \delta(t-t_0) = 1$ for $t > t_0$.

Equation (KK-3) is just the standard solution of a single linear first order differential equation (KK-1) for an unknown function $P_n(t)$ with $P_{n+1}(t)$ treated as known. One can verify that (KK-3) solves (KK-1) by direct substitution:

$$\frac{dP_n(t)}{dt} = e^{-\int_{t_0}^{t} \Gamma_n(\tau)d\tau} \Gamma_{n+1}(t) P_{n+1}(t) + \int_{t_0}^{t} \frac{d}{dt} \left[ e^{-\int_{t_0}^{t'} \Gamma_n(\tau)d\tau} \Gamma_{n+1}(t') P_{n+1}(t') \right] dt'$$

$$= \Gamma_{n+1}(t) P_{n+1}(t) + \int_{t_0}^{t} e^{-\int_{t_0}^{t'} \Gamma_n(\tau)d\tau} \frac{d}{dt} \left[ -\int_{t'}^{t} \Gamma_n(\tau)d\tau \right] \Gamma_{n+1}(t') P_{n+1}(t') dt'$$

$$= \Gamma_{n+1}(t) P_{n+1}(t) - \Gamma_n(t) P_n(t).$$
III. EXPLICIT SOLUTION FOR TIME-INDEPENDENT RATE RATIO \( \Gamma_n(t)/\Gamma_{n-1}(t) = \text{const} \)

The first solution described in KK corresponds to the case of

\[
\Gamma_n(t) = \frac{X_n}{X_1} \Gamma_1(t),
\]

with \( X_n = \exp \sum_{k=1}^{n} \delta_k \) being time-independent constants.

For the rates obeying the condition (3), the general solution can be constructed in the following form \(^1\)

\[
P_n(t) = \sum_{k=n}^{N} R_{nk} e^{-\int_{t_0}^{t} \Gamma_k(t') dt'},
\]

with constant coefficients \( R_{nk} \) that need to be determined.

A. Derivation of \( R_{nk} \)

For \( n = N \), the initial conditions \( P_N(t_0) = 1, P_{N+1}(t_0) = 0 \) and equation (4) for \( P_N(t) \) give

\[
P_N(t) = e^{-\int_{t_0}^{t} \Gamma_N(t') dt'} \implies R_{NN} = 1.
\]

The initial condition \( P_n(t_0) = 0 \) for \( n < N \), applied to (4), implies:

\[
\sum_{k=n}^{N} R_{nk} = 0, \quad n < N.
\]

For \( n < N \) the substituting (4) into the differential equation (4) gives

\[
\frac{dP_n(t)}{dt} = -\sum_{k=n}^{N} R_{nk} \Gamma_k(t) e^{-\int_{t_0}^{t} \Gamma_k(t') dt'}
\]

\[
-\Gamma_n(t) P_n(t) + \Gamma_{n+1}(t) P_{n+1}(t) = -\Gamma_n(t) \sum_{k=n}^{N} R_{nk} e^{-\int_{t_0}^{t} \Gamma_k(t') dt'} + \Gamma_{n+1}(t) \sum_{k=n+1}^{N} R_{n+1,k} e^{-\int_{t_0}^{t} \Gamma_k(t') dt'}
\]

Now we invoke the condition (3) which allows us to equate the coefficients of \( e^{-\int_{t_0}^{t} \Gamma_k(t') dt'} \) between (7) and (8):

\[
X_k R_{nk} = X_n R_{nk} + X_{n+1} R_{n+1,k}, \quad n < k \leq N
\]

\[
R_{n-1,k} = \frac{X_n}{X_k - X_{n-1}} R_{n,k}, \quad n \leq k \leq N
\]

Equations (6) and (9) give the sought-after recurrence relations:

\[
R_{nk} = R_{kk} \prod_{m=n+1}^{k} \frac{X_m}{X_k - X_{m-1}}, \quad n < k \leq N
\]

\[
R_{kk} = -\sum_{m=k+1}^{N} R_{km}, \quad k < N
\]

Equations (KK-5’) and (KK-6’) together with (5) are equivalent to Eqs. (5) and (6) of KK with \( C_k \equiv R_{kk} \) and \( Q_{kn} \equiv R_{kn}/R_{kk} \).

\(^1\) This form was inspired by studying explicit solutions for \( \Gamma_n(t) \sim e^{-t} \) and \( N = 1, 2, 3 \) with the means of a computer algebra system Mathematica.
B. Explicit solution of recurrence relations

The recurrence relations (KK-5) and (KK-6) admit the following explicit solution:

\[ R_{nk} = \prod_{m=n+1}^{N} X_m \prod_{\substack{m=n+1 \atop m \neq k}}^{N} \frac{1}{X_k - X_m}. \] (10)

The solution (10) can be obtained for finite \( n \) and \( k \) by means of computer algebra, and proven in general form by induction.

Since integration over time preserves the condition (3), we can choose

\[ X_n = \int_{t_0}^{t} \Gamma_n(t) \, dt \] (11)

and write down the solution explicitly:

\[ P_n(t) = \sum_{k=n}^{N} e^{-X_k} \prod_{\substack{m=n+1 \atop m \neq k}}^{N} \frac{1}{X_k - X_m}. \] (12)

The solution (12) agrees precisely with the solution for \( N = 3 \) and \( \Gamma_n = \text{const} \) obtained by Miyamoto et al. [3].

IV. SOLUTION IN THE LIMIT OF TIME-SCALE SEPARATION BETWEEN CASCADE STEPS

A more general solution that does not rely on condition (3) is derived in KK in the limit of decay time-scale separation between consecutive steps of the cascade. This is motivated as follows:

\( P_{n+1}(t) \) stops changing appreciably over a timescale on the order of \( \Gamma_{n+1}(t) \) during which \( P_n(t) \) changes only due to probability flux from state \((n+1)\), with negligible decay down the cascade to state \((n-1)\). This condition corresponds to \( \Gamma_{n+1}(t) \gg \Gamma_n(t) \) during the relevant time interval.

The mathematical part of the derivation proceeds as follows:

1. Summing equations (KK-3) for all \( dP_m/dt \) with \( m \geq n \) gives

\[ \Gamma_{n+1}(t)P_{n+1}(t) = -\frac{d}{dt} \sum_{m>n} P_m(t). \] (13)

2. On the “slow” time-scale controlled by \( \Gamma_n(t) \), the function \( \sum_{m>n} P_m(t) \) is changing rapidly from 1 to its asymptotic value \( \sum_{m>n} P_m(t \to \infty) \) and hence can be approximated by a step function in time. \( t \to \infty \) corresponds to the time when the decay transitions no longer take place.

3. The derivative of a step function is proportional to a delta function and thus the exact integral (KK-3) can be approximated as follows:

\[ P_n(t) = \int_{t_0}^{t} e^{-f_0^t \Gamma_n(\tau) \, d\tau} \left[ \frac{\Gamma_n+1(t') P_{n+1}(t')}{\Gamma_n(t') \prod_{\substack{m=n+1 \atop m \neq k}}^{N} \frac{1}{X_k - X_m}} \right] \, dt' \approx e^{\int_{t_0}^{t} \Gamma_n(\tau) \, d\tau} \left[ 1 - \sum_{m>n} P_m(t \to \infty) \right]. \] (14)

Equation (14) essentially states that the decay of all previous states (higher than \( n \)) provides an initial condition for the decay of the \( n \)-th state. Based on (14), the condition on the final probabilities \( P_n(t \to \infty) \equiv P_n \) stated in KK is formulated:

\[ P_n = e^{-X_n} \left( 1 - \sum_{m=n+1}^{N} P_m \right). \] (15)

4. Equation (15) is solved by expressing \( P_{n-1} \) in terms of \( P_n \). Using (15) for \( P_n \) we can express \( \sum_{m>n} P_m = 1 - e^{X_n}P_n \) and

\[ \sum_{m>n-1} P_m = 1 + (1 - e^{X_n})P_n. \] (16)
Substituting the sum (16) into (15) for $P_{n-1}$, we get the desired recurrence relation

$$P_{n-1} = e^{-X_{n-1}}(e^{X_n} - 1)P_n. \quad (17)$$

Starting from $P_N = e^{-X_N}$ and iterating (17) gives

$$P_{n-1} = e^{-X_N}\times(e^{X_N} - 1)\times(e^{X_{N-1}} - 1)\times(e^{-X_{N-2}}\times\ldots, \quad (18)$$

wherefrom the general form is easy to infer

$$P_n = e^{-X_n}\prod_{m=n+1}^{N}(1 - e^{-X_m}) \quad (\text{KK-11}).$$

Equation (KK-11) has also been applied to a generalised decay cascade scenario by Fricke et al. [4]. They consider the onset of decay steps at different times, $t_0 < \ldots < t_{n+1} < t_n < \ldots$, which in the notation of KK corresponds to

$$\Gamma_n(t) = \tilde{\Gamma}_n(t)\Theta(t - t_n^b), \quad (19)$$

where $\Theta(x)$ is a unit step function and $\tilde{\Gamma}_n(t)$ are smooth functions that decay to zero as for $t \to \infty$. The condition $X_n = \int_{t_0}^{\infty} \Gamma_n(t)dt \gg X_{n-1}$ justifies the sequential cascade approximation (14) and hence the probability distribution (KK-11).

[1] C. Gardiner, *Stochastic Methods: A Handbook for the Natural and Social Sciences* (Springer Verlag, 2009), 4th ed.
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[3] S. Miyamoto, K. Nishiguchi, Y. Ono, K. M. Itoh, and A. Fujwara, Appl. Phys. Lett. 93, 222103 (2008).
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