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1987 J. Phys. A: Math. Gen. 20 3367

(http://iopscience.iop.org/0305-4470/20/11/039)

J. Phys. A: Math. Gen. **20** (1987) 3367-3380. Printed in the UK

Age distributions in physical systems

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Received 17 November 1986

Abstract. Recently Vlad *et al* introduced a new stochastic description of memory effects, based on a system of age-dependent master equations (ADME). In this paper the ADME approach is applied to physical processes obeying a phenomenological master equation. The joint probabilities and the correlation functions of the ages of different fluctuation states are computed. The suggested method is extended to systems for which the transition rates are age dependent. A physical interpretation of the main results is given by means of a system of age-dependent integral equations. We prove that the ADME formalism is a generalisation of the well known continuous time random walk theory (CTRW).

1. Introduction

In statistical physics, the usual measure of the temporal development of fluctuations is the so-called correlation time τ_c (Kubo 1961). τ_c is the time necessary for the regression of a fluctuation state towards equilibrium.

Recently, Vlad *et al* (1984) and Vlad and Popa (1986, 1987) have introduced a new notion, the age of a fluctuation state, a. a measures the time interval within which a fluctuation state remains unchanged. Considering a system described in terms of S variables $\boldsymbol{X}=(X_1, \ldots, X_S)$, the age a of a state $\boldsymbol{X}$ can be viewed, together with $X_1, \ldots, X_S$, as a random variable. In Vlad *et al* (1984) and Vlad and Popa (1986) a system of age-dependent master equations (ADME) for the state probability $\mathscr{P}(\boldsymbol{X}, a, t)$ is derived and its solution for certain cases is given.

In this paper we consider a related problem—the determination of multitemporal joint probabilities $\mathscr{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)\mathrm{d}\boldsymbol{X}_1 \ldots \mathrm{d}\boldsymbol{X}_n\, \mathrm{d}a_1 \ldots \mathrm{d}a_n$. The layout of the paper is as follows. We first consider a physical problem that could motivate the investigation and then examine the case of an arbitrary system obeying a phenomenological master equation. The next step is to consider the more general case of age-dependent transition rates. As a final topic we compare our approach with the theory of continuous time random walks (CTRW, Montroll and Weiss 1965, Scher and Lax 1973, Weiss and Rubin 1983).

2. Formulation of the problem

As a simple physical example we consider the following model of an intrinsic semiconductor (Van Kampen 1976). A crystal has a nearly empty conduction band and a nearly full valence band. Denoting by N the number of electrons that by thermal fluctuations have been excited into the conduction band, the transition rate for an excitation to occur can be expressed as (Van Kampen 1976):

$$W(N \to N+1) = \beta V \tag{1}$$

where V is the volume of the crystal and β is a constant. Similarly, the probability for a recombination is given by

$$W(N \to N-1) = V\gamma(N/V)^2. \tag{2}$$

The constants β and γ are related to each other through the relationship

$$\beta/\gamma \sim e^{-\varepsilon/kT} \tag{3}$$

where ε is the energy gap between the two bands.

Making use of (1) and (2) we can derive a phenomenological master equation for the state probability $P(N, t)$:

$$\partial_t P(N, t) = \beta V[P(N-1) - P(N)] + \gamma V\{P(N+1)[(N+1)/V]^2 - P(N)(N/V)^2\}. \tag{4}$$

Applying the Van Kampen extensivity ansatz, i.e. assuming that the volume dependence of N is given by

$$N = \langle n\rangle V + V^{1/2}x \qquad n = N/V = \langle n\rangle + V^{-1/2}x \tag{5}$$

where $\langle n\rangle V$ and $V^{1/2}x$ are the deterministic and fluctuating contributions to N, with

$$\langle n\rangle, x \sim V^0 \tag{6}$$

the stationary solution of (4) can be expressed as (Van Kampen 1976)

$$P^{\mathrm{st}}(N) = \frac{1}{(2\pi\langle\Delta N^2\rangle^{\mathrm{st}})^{1/2}} \exp\left(-\frac{(N-\langle N\rangle^{\mathrm{st}})^2}{2\langle\Delta N^2\rangle^{\mathrm{st}}}\right) \tag{7}$$

where

$$\langle N\rangle^{\mathrm{st}} = V(\beta/\gamma)^{1/2} \tag{8}$$

$$\langle\Delta N^2\rangle^{\mathrm{st}} = \langle(N - \langle N^{\mathrm{st}}\rangle)^2\rangle = \tfrac{1}{2}V(\beta/\gamma)^{1/2}. \tag{9}$$

A similar relationship can be derived for the temporal correlation function of N:

$$\langle\Delta N(0)\Delta N(\tau)\rangle^{\mathrm{st}} = \tfrac{1}{2}V(\beta/\gamma)^{1/2} e^{-2\tau\sqrt{\beta\gamma}} \tag{10}$$

Then, in this case the correlation time (Kubo 1961)

$$\tau_c = \int_0^\infty \frac{\langle\Delta N(0)\Delta N(\tau)\rangle^{\mathrm{st}}}{\langle\Delta N^2\rangle^{\mathrm{st}}} \, d\tau \tag{11}$$

is equal to

$$\tau_c = 1/2\sqrt{\beta\gamma}. \tag{12}$$

Introducing the age state-probability distribution $\mathcal{P}^{\mathrm{st}}(N, a)$ from (1) and (2) we can derive the following 'balance equations':

$$\mathcal{P}^{\mathrm{st}}(N, a+\Delta a) = \mathcal{P}^{\mathrm{st}}(N, a)(1 - \beta V\Delta a - V\gamma(N/V)^2\Delta a) \qquad a \neq 0 \tag{13}$$

$$\mathcal{P}^{\mathrm{st}}(N, 0+\Delta a) = \int_0^\infty \mathcal{P}^{\mathrm{st}}(N-1, a)\beta V \, da + \int_0^\infty \mathcal{P}^{\mathrm{st}}(N+1, a) V\gamma[(N+1)/V]^2 \, da \tag{14}$$

from which, making $\Delta a \to 0$, we obtain the required age-dependent master equations

$$\partial_a \mathscr{P}^{st}(N, a) + [\beta V + V\gamma(N/V)^2]\mathscr{P}^{st}(N, a) = 0 \tag{15}$$

$$\mathscr{P}^{st}(N, 0) = \int_0^\infty \{\mathscr{P}^{st}(N-1, a)\beta V + \mathscr{P}^{st}(N+1, a)\gamma V[(N+1)/V]^2\} \, da. \tag{16}$$

Integrating (15), making use of (16) and taking into account that

$$P^{st}(N) = \int_0^\infty \mathscr{P}^{st}(N, a) \, da \tag{17}$$

we come to

$$\mathscr{P}^{st}(N, a) = V(\beta + \gamma N^2/V^2) P^{st}(N) \exp[-aV(\beta + \gamma N^2/V^2)]. \tag{18}$$

Obviously, the mean age and the dispersion of the age of a given fluctuation state N are defined in terms of the conditional probability

$$R^{st}(a \mid N) = \frac{\mathscr{P}^{st}(N, a)}{P^{st}(N)} = V(\beta + \gamma N^2/V^2) \exp[-aV(\beta + \gamma N^2/V^2)] \tag{19}$$

through the relationships

$$\langle a(N)\rangle^{st} = \int_0^\infty aR^{st}(a \mid N) \, da = V^{-1}(\beta + \gamma N^2/V^2)^{-1} \tag{20}$$

$$\langle \Delta a^2(N)\rangle^{st} = \int_0^\infty (a - \langle a\rangle^{st})^2 R^{st}(a \mid N) \, da = V^{-2}(\beta + \gamma N^2/V^2)^{-2}. \tag{21}$$

In particular,

$$\langle a(\langle N\rangle^{st})\rangle^{st} = 1/2\beta V. \tag{22}$$

We see that the mean age of a fluctuation state decreases with the increase of the system volume, while the correlation time is volume independent. This fact outlines the different nature of the two averages. It is interesting to evaluate the ratio $\tau_c/\langle a(\langle N\rangle^{st})\rangle^{st}$. Considering that $\langle N\rangle^{st} \sim 10^{14}\text{-}10^{15}$ we obtain

$$\tau_c/\langle a(\langle N\rangle^{st})\rangle^{st} = \langle N\rangle^{st} \sim 10^{14}\text{-}10^{15}. \tag{23}$$

This is an expected result, since for macroscopical systems the mean age of a fluctuation state is very small (Vlad and Popa 1986).

The next step of our analysis might be to determine the joint probabilities $\mathscr{P}_n^{st}(N_n, a_n, t_n, \ldots, N_1, a_1, t_1) \, da_1 \ldots da_n$ and the corresponding moments. We are particularly interested in the determination of the function

$$\begin{aligned} \varphi_2(a_1(N_1, t_1), a_2(N_2, t_2)) &= \langle \Delta a_1(N_1, t_1) \Delta a_2(N_2, t_2)\rangle^{st} \\ &= \int\int_0^\infty R_2^{st}(a_2, t_2; a_1, t_1 | N_2, t_2; N_1, t_1) \, (a_2 - \langle a_2(N_2, t_2)\rangle^{st}) \\ &\quad \times (a_1 - \langle a_1(N_1, t_1)\rangle^{st}) \, da_1 \, da_2 \end{aligned} \tag{24}$$

where

$$R_2^{st}(a_2, t_2; a_1, t_1 | N_2, t_2; N_1, t_1) = \frac{\mathscr{P}_2^{st}(N_2, a_2, t_2; N_1, a_1, t_1)}{\int\int_0^\infty \mathscr{P}_2^{st}(N_2, a_2, t_2; N_1, a_1, t_1) \, da_1 \, da_2}. \tag{25}$$

The physical meaning of this function is straightforward: φ_2 is a measure for the correlation between the ages $a_1(N_1, t_1)$ and $a_2(N_2, t_2)$ of the states $N_1(t_1)$ and $N_2(t_2)$.

The joint probabilities can be determined by means of a generalised ADME system. The derivation and analysis of this system is the main purpose of the following sections.

3. Joint probabilities

In this section we answer the following question. Considering a Markovian system described by a set of macrovariables $X_1, \ldots, X_s$ and supposing that the transition rates

$$W'\Delta t = W(\boldsymbol{X} \to \boldsymbol{X}')\Delta t \qquad W''\Delta t = W(\boldsymbol{X} \to \boldsymbol{X}'')\Delta t, \ldots \tag{26}$$

$$\boldsymbol{X} = (X_1, \ldots, X_s) \qquad \boldsymbol{X}' = (X_1', \ldots, X_s'), \ldots \tag{27}$$

are known, what is the probability

$$\mathscr{P}_N \,\mathrm{d}\boldsymbol{X}_1 \ldots \mathrm{d}\boldsymbol{X}_N \,\mathrm{d}a_1 \ldots \mathrm{d}a_N = \mathscr{P}_N(\boldsymbol{X}_N, a_N, t_N; \ldots; \boldsymbol{X}_1, a_1, t_1)\,\mathrm{d}\boldsymbol{X}_1 \ldots \mathrm{d}\boldsymbol{X}_N \,\mathrm{d}a_1 \ldots \mathrm{d}a_N \tag{28}$$

that the state vector has the value $\boldsymbol{X}_1$ at time $t_1, \ldots,$ and the value $\boldsymbol{X}_N$ at time t_N and that the age of the state $\boldsymbol{X}_1$ at time t_1 is $a_1, \ldots,$ and that the age of the state $\boldsymbol{X}_N$ at time t_N is a_N?

The answer to this question may be given by generalising the system of age-dependent master equations (ADME, Vlad *et al* 1984). The ADME system describes the time evolution of the probability $\mathscr{P}_1(\boldsymbol{X}, a, t)$:

$$(\partial_{t_1} + \partial_{a_1})\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1) = -\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1)\int_{\boldsymbol{X}'} W(\boldsymbol{X}_1 \to \boldsymbol{X}')\,\mathrm{d}\boldsymbol{X}' \tag{29}$$

$$\mathscr{P}_1(\boldsymbol{X}_1, 0, t_1) = \int_{\boldsymbol{X}'}\int_0^\infty W(\boldsymbol{X}' \to \boldsymbol{X}_1)\mathscr{P}_1(\boldsymbol{X}', a', t_1)\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}' \tag{30}$$

with

$$\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_0) = \mathscr{P}_1^0(\boldsymbol{X}_1, a_1). \tag{31}$$

Introducing the probability

$$P_1(\boldsymbol{X}_1, t_1) = \int_0^\infty \mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1)\,\mathrm{d}a_1 \tag{32}$$

(29) and (30) lead to the well known phenomenological master equation (PME, Van Kampen 1981, see also Vlad *et al* 1984, Vlad and Popa 1986):

$$\partial_{t_1} P_1(\boldsymbol{X}_1, t_1) = \int_{\boldsymbol{X}'} W(\boldsymbol{X}' \to \boldsymbol{X}_1) P_1(\boldsymbol{X}', t_1)\,\mathrm{d}\boldsymbol{X}' - P_1(\boldsymbol{X}_1, t_1)\int_{\boldsymbol{X}'} W(\boldsymbol{X}_1 \to \boldsymbol{X}')\,\mathrm{d}\boldsymbol{X}' \tag{33}$$

with

$$P_1(\boldsymbol{X}_1, t_0) = P_1^0(\boldsymbol{X}_1) = \int_0^\infty \mathscr{P}_1^0(\boldsymbol{X}_1, a_1)\,\mathrm{d}a_1. \tag{34}$$

The integration of the ADME system is straightforward, provided that the solution $P_1(\boldsymbol{X}_1, t_1)$ of the phenomenological master equation is known. Indeed, integrating the equation (29) and making use of (30)-(32) yields

$$\begin{aligned}\mathcal{P}_1(\boldsymbol{X}_1, a_1, t_1) = {} & h(t_1 - t_0 - a_1)\exp(-a_1\Omega(\boldsymbol{X}_1))\int_{\boldsymbol{X}'} W(\boldsymbol{X}' \to \boldsymbol{X}_1)P_1(\boldsymbol{X}', t_1 - a)\,\mathrm{d}\boldsymbol{X} \\ & + h(a_1 - t_1 + t_0)\exp[-(t_1 - t_0)\Omega(\boldsymbol{X}_1)]\mathcal{P}_1^0(\boldsymbol{X}_1, a_1 - t_1 + t_0)\end{aligned} \tag{35}$$

where

$$\Omega(\boldsymbol{X}_1) = \int_{\boldsymbol{X}'} W(\boldsymbol{X}_1 \to \boldsymbol{X}')\,\mathrm{d}\boldsymbol{X}' \tag{36}$$

and $h(t)$ is the usual Heaviside function.

To derive the equations which describe the temporal development of the joint probabilities, we assume the validity of the following inequalities:

$$t_N \geqslant t_{N-1} \geqslant \ldots \geqslant t_1 \geqslant t_0. \tag{37}$$

Taking (37) into account, we may write a chain of 'balance equations' for $\mathcal{P}_n, \mathcal{P}_{n-1}, \ldots$:

$$\begin{aligned}&\mathcal{P}_n(\boldsymbol{X}_n, a_n + \Delta t, t_n + \Delta t; \boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1) \\ &\quad = \mathcal{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)\left(1 - \int_{\boldsymbol{X}'} W(\boldsymbol{X}_n \to \boldsymbol{X}')\,\Delta t\,\mathrm{d}\boldsymbol{X}'\right) \\ &\qquad\qquad a_n \neq 0,\; n = 1, 2, \ldots, N\end{aligned} \tag{38}$$

$$\begin{aligned}&\mathcal{P}_n(\boldsymbol{X}_n, 0 + \Delta t, t_n + \Delta t; \boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)\Delta t \\ &\quad = \int_{\boldsymbol{X}'}\int_0^\infty W(\boldsymbol{X}' \to \boldsymbol{X}_n)\Delta t\,\mathcal{P}_n(\boldsymbol{X}', a', t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a' \\ &\qquad\qquad n = 1, 2, \ldots, N.\end{aligned} \tag{39}$$

For $\Delta t \to 0$, these equations become

$$\begin{aligned}&(\partial_{t_n} + \partial_{a_n})\mathcal{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1) \\ &\quad = -\mathcal{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)\int_{\boldsymbol{X}'} W(\boldsymbol{X}_n \to \boldsymbol{X}')\,\mathrm{d}\boldsymbol{X}' \qquad n = 1, \ldots, N\end{aligned} \tag{40}$$

$$\begin{aligned}&\mathcal{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1) \\ &\quad = \int_{\boldsymbol{X}'}\int_0^\infty W(\boldsymbol{X}' \to \boldsymbol{X}_n)\mathcal{P}_n(\boldsymbol{X}', a', t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a' \\ &\qquad\qquad n = 1, \ldots, N.\end{aligned} \tag{41}$$

Together with the initial conditions

$$\begin{aligned}&\mathcal{P}_n(\boldsymbol{X}_n, a_n, t_n = t_{n-1}; \boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1) \\ &\quad = \mathcal{P}_{n-1}(\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)\delta(\boldsymbol{X}_n - \boldsymbol{X}_{n-1})\delta(a_n - a_{n-1}) \\ &\qquad\qquad n = 2, \ldots, N\end{aligned} \tag{42}$$

$$\mathcal{P}_1(\boldsymbol{X}_1, a_1, t_1 = t_0) = \mathcal{P}_1^0(\boldsymbol{X}_1, a_1)$$

(40) and (41) describe completely the time evolution of $\mathcal{P}_N, \ldots, \mathcal{P}_1$. Introducing the

conditional probabilities

$$\mathscr{G}_n(\boldsymbol{X}_n, a_n, t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \boldsymbol{X}_{n-2}, a_{n-2}, t_{n-2}; \ldots; \boldsymbol{X}_1, a_1, t_1)$$
$$= \frac{\mathscr{P}_n(\boldsymbol{X}_n, a_n, t_n; \ldots; \boldsymbol{X}_1, a_1, t_1)}{\mathscr{P}_{n-1}(\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)} \qquad n=2, \ldots, N \tag{43}$$

(40) and (41) turn into a simpler form:

$$(\Omega(\boldsymbol{X}_n) + \partial_{t_n} + \partial_{a_n})\mathscr{G}_n(\boldsymbol{X}_n, a_n, t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; X_1, a_1, t_1) = 0 \qquad n=2, \ldots, N \tag{44}$$

$$\mathscr{G}_n(\boldsymbol{X}_n, 0, t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)$$
$$= \int_{\boldsymbol{X}'}\int_0^\infty W(\boldsymbol{X}' \to \boldsymbol{X}_n)\mathscr{G}_n(\boldsymbol{X}', a', t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)\, \mathrm{d}\boldsymbol{X}'\, \mathrm{d}a'$$
$$n=2, \ldots, N \tag{45}$$

with

$$\mathscr{G}_n(\boldsymbol{X}_n, a_n, t_n = t_{n-1}|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)$$
$$= \delta(\boldsymbol{X}_n - \boldsymbol{X}_{n-1})\delta(a_n - a_{n-1}) \qquad n=2, \ldots, N. \tag{46}$$

From (44)-(46) we can see that

$$\mathscr{G}_n(\boldsymbol{X}_n, a_n, t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)$$
$$= \mathscr{G}_n(\boldsymbol{X}_n, a_n, t_n|\boldsymbol{X}_{n-1}, a_{n-1}, t_{n-1})$$
$$= \text{independent of } (\boldsymbol{X}_{n-2}, a_{n-2}, t_{n-2}, \ldots, \boldsymbol{X}_1, a_1, t_1) \tag{47}$$

an expected result, since the stochastic process considered is Markovian.

Using (47) and dropping the index n, equations (44)-(46) become

$$(\Omega(\boldsymbol{X}) + \partial_t + \partial_a)\mathscr{G}(\boldsymbol{X}, a, t-t''|\boldsymbol{X}'', a'', 0) = 0 \tag{48}$$

$$\mathscr{G}(\boldsymbol{X}, 0, t-t''|\boldsymbol{X}'', a'', 0) = \int_{\boldsymbol{X}'}\int_0^\infty W(\boldsymbol{X}' \to \boldsymbol{X})\mathscr{G}(\boldsymbol{X}', a', t-t''|\boldsymbol{X}'', a'', 0)\, \mathrm{d}a'\, \mathrm{d}\boldsymbol{X}' \tag{49}$$

with

$$\mathscr{G}(\boldsymbol{X}, a, 0|\boldsymbol{X}'', a'', 0) = \delta(\boldsymbol{X} - \boldsymbol{X}'')\delta(a - a''). \tag{50}$$

Integrating (48) and (50) with respect to a, eliminating the function $\mathscr{G}(a=0)$ from (49) and introducing the probability

$$G(\boldsymbol{X}, t-t''|\boldsymbol{X}'', a'', 0) = \int_0^\infty \mathscr{G}(\boldsymbol{X}, a, t-t''|\boldsymbol{X}'', a'', 0)\, \mathrm{d}a \tag{51}$$

leads to a phenomenological master equation similar to (33):

$$\partial_t G(\boldsymbol{X}, t-t''|\boldsymbol{X}'', a'', 0)$$
$$= \int_{\boldsymbol{X}'} W(\boldsymbol{X}' \to \boldsymbol{X})G(\boldsymbol{X}', t-t''|\boldsymbol{X}'', a'', 0)\, \mathrm{d}\boldsymbol{X}'$$
$$- \int_{\boldsymbol{X}'} W(\boldsymbol{X} \to \boldsymbol{X}')G(\boldsymbol{X}, t-t''|\boldsymbol{X}'', a'', 0)\, \mathrm{d}\boldsymbol{X}' \tag{52}$$

with

$$G(\boldsymbol{X}, 0 | \boldsymbol{X}'', a'', 0) = \delta(\boldsymbol{X} - \boldsymbol{X}''). \tag{53}$$

From (52) and (53) we notice that

$$G(\boldsymbol{X}, t - t'' | \boldsymbol{X}'', a'', 0) = G(\boldsymbol{X}, t - t'' | \boldsymbol{X}'', 0) = \text{independent of } a''. \tag{54}$$

On the other side, comparing (33) and (34) with (52) and (53) we can see that $G(\boldsymbol{X}, t - t'' | \boldsymbol{X}'', 0)$ is the Green function corresponding to the equation (33), and the solution $P_1(\boldsymbol{X}, t)$ of (33) can be written as

$$P_1(\boldsymbol{X}, t) = \int_{\boldsymbol{X}''} G(\boldsymbol{X}, t - t_0 | \boldsymbol{X}'', 0) P_1^0(\boldsymbol{X}'') \, \mathrm{d}\boldsymbol{X}''. \tag{55}$$

Using the 'Trucco variables' $\xi = t - a$, $\eta = a$ (Trucco 1965), (48) may be integrated analytically, resulting in

$$\begin{aligned} &\mathscr{G}(\boldsymbol{X}, a, t - t'' | \boldsymbol{X}'', a'', 0) \\ &\quad = h(t - t'' - a) \mathscr{G}(\boldsymbol{X}, 0, t - t'' - a | \boldsymbol{X}'', a'', 0) \exp(-a\Omega(\boldsymbol{X})) \\ &\qquad + h(a - t + t'') \delta(\boldsymbol{X} - \boldsymbol{X}'') \delta(a - t + t'' - a'') \exp[-(t - t'')\Omega(\boldsymbol{X})]. \end{aligned} \tag{56}$$

Eliminating from (49) and (56) the functions $\mathscr{G}(\boldsymbol{X}, 0, t - t'' - a | \boldsymbol{X}'', a'', 0)$ and making use of (51) and (52) we obtain

$$\begin{aligned} &\mathscr{G}(\boldsymbol{X}, a, t - t'' | \boldsymbol{X}'', a'', 0) \\ &\quad = h(t - t'' - a)(G(\boldsymbol{X}, t - t'' - a | \boldsymbol{X}'', 0)\Omega(\boldsymbol{X}) \\ &\qquad + \partial_t G(\boldsymbol{X}, t - t'' - a | \boldsymbol{X}'', 0)) \exp(-a\Omega(\boldsymbol{X})) \\ &\qquad + h(a - t + t'') \delta(\boldsymbol{X} - \boldsymbol{X}'') \delta(a - t + t'' - a'') \exp[-(t - t'')\Omega(\boldsymbol{X})]. \end{aligned} \tag{57}$$

Now combining (32), (43), (55) and (57) leads to

$$\begin{aligned} &\mathscr{P}_N(\boldsymbol{X}_N, a_N, t_N; \ldots; \boldsymbol{X}_1, a_1, t_1) \\ &\quad = \prod_{q=2}^{N} [\mathscr{G}_q(\boldsymbol{X}_q, a_q, t_q - t_{q-1} | \boldsymbol{X}_{q-1}, a_{q-1}, 0)] \mathscr{P}(\boldsymbol{X}_1, a_1, t_1) \\ &\quad = \prod_{q=2}^{N} \{h(t_q - t_{q-1} - a_q) \exp(-a_q \Omega(\boldsymbol{X}_q)) \\ &\qquad \times (\Omega(\boldsymbol{X}_q) + \partial_{t_q}) G(\boldsymbol{X}_q, t_q - t_{q-1} - a_q | \boldsymbol{X}_{q-1}, 0) \\ &\qquad + h(a_q - t_q + t_{q-1}) \exp[-(t_q - t_{q-1})\Omega(\boldsymbol{X}_q)] \\ &\qquad \times \delta(\boldsymbol{X}_q - \boldsymbol{X}_{q-1}) \delta(a_q - t_q + t_{q-1} - a_{q-1})\} \\ &\qquad \times \bigg(h(t_1 - t_0 - a_1) \exp(-a_1 \Omega(\boldsymbol{X}_1)) \int_{\boldsymbol{X}''} \mathrm{d}\boldsymbol{X}'' \, P_1^0(\boldsymbol{X}'') \\ &\qquad \times [(\Omega(\boldsymbol{X}_1) + \partial_{t_1}) G(\boldsymbol{X}_1, t_1 - t_0 - a_1 | \boldsymbol{X}'', 0)] \\ &\qquad + h(a_1 - t_1 + t_0) \exp[-(t_1 - t_0)\Omega(\boldsymbol{X}_1) \mathscr{P}_1^0(\boldsymbol{X}_1, a_1 - t_1 + t_0)] \bigg) \\ &\qquad t_N \geqslant t_{N-1} \geqslant \ldots \geqslant t_1 \geqslant t_0. \end{aligned} \tag{58}$$

Thus, the determination of the joint probabilities $\mathscr{P}_N, \ldots, \mathscr{P}_1$ reduces to the integration of the phenomenological master equation (52).

By removing the constraints (37) the equations (58) may be rewritten in a more convenient form. For instance, in the case $N=2$, we have

$$
\begin{aligned}
&\mathscr{P}_2(\boldsymbol{X}_2, a_2, t_2; \boldsymbol{X}_1, a_1, t_1) \\
&\quad = h(t_2-t_1)\bigg(h(t_1-t_0-a_1)\exp(-a_1\Omega(\boldsymbol{X}_1)) \\
&\qquad \times \int_{\boldsymbol{X}''} \mathrm{d}\boldsymbol{X}''\, P_1^0(\boldsymbol{X}'')[(\Omega(\boldsymbol{X}_1)+\partial_{t_1})G(\boldsymbol{X}_1, t_1-t_0-a_1|\boldsymbol{X}'', 0)] \\
&\qquad + h(a_1-t_1+t_0)\mathscr{P}_1^0(\boldsymbol{X}_1, a_1-t_1+t_0)\exp[-(t_1-t_0)\Omega(\boldsymbol{X}_1)]\bigg) \\
&\qquad \times \{h(t_2-t_1-a_2)\exp(-a_2\Omega(\boldsymbol{X}_2))(\Omega(\boldsymbol{X}_2)+\partial_{t_2})G_2(\boldsymbol{X}_2, t_2-t_1-a_2|\boldsymbol{X}_1, 0) \\
&\qquad + h(a_2-t_2+t_1)\exp[-(t_2-t_1)\Omega(\boldsymbol{X}_2)]\delta(\boldsymbol{X}_2-\boldsymbol{X}_1)\delta(a_2-t_2+t_1-a_1)\} \\
&\qquad + h(t_1-t_2)\bigg(h(t_2-t_0-a_2)\exp(-a_2\Omega(\boldsymbol{X}_2))\int_{\boldsymbol{X}''} \mathrm{d}\boldsymbol{X}''\, P_1^0(\boldsymbol{X}'') \\
&\qquad \times [(\Omega(\boldsymbol{X}_2)+\partial_{t_2})G(\boldsymbol{X}_2, t_2-t_0-a_2|\boldsymbol{X}'', 0)] \\
&\qquad + h(a_2-t_2+t_0)\mathscr{P}_1^0(\boldsymbol{X}_2, a_2-t_2+t_0)\exp[-(t_2-t_0)\Omega(\boldsymbol{X}_2)]\bigg) \\
&\qquad \times \{h(t_1-t_2-a_1)\exp(-a_1\Omega(\boldsymbol{X}_1))(\Omega(\boldsymbol{X}_1)+\partial_{t_1})G(\boldsymbol{X}_1, t_1-t_2-a_1|\boldsymbol{X}_2, 0) \\
&\qquad + h(a_1-t_1+t_2)\exp[-(t_1-t_2)\Omega(\boldsymbol{X}_1)]\delta(\boldsymbol{X}_1-\boldsymbol{X}_2)\delta(a_1-t_1+t_2-a_2)\}.
\end{aligned}
\tag{58'}
$$

4. Correlation functions

The correlation functions

$$\langle a_N^{k_N} a_{N-1}^{k_{N-1}} \ldots a_1^{k_1}\rangle = \langle a_N^{k_N}(\boldsymbol{X}_N, t_N) a_{N-1}^{k_{N-1}}(\boldsymbol{X}_{N-1}, t_{N-1}) \ldots a_1^{k_1}(\boldsymbol{X}_1, t_1)\rangle \tag{59}$$

of the ages of fluctuation states $\boldsymbol{X}_N, \boldsymbol{X}_{N-1}, \ldots, \boldsymbol{X}_1$ are defined in terms of the conditional probabilities

$$
\begin{aligned}
&R_N(a_N, t_N; a_{N-1}, t_{N-1}; \ldots; a_1, t_1|\boldsymbol{X}_N, t_N; \boldsymbol{X}_{N-1}, t_{N-1}; \ldots; \boldsymbol{X}_1, t_1) \\
&\quad = \frac{\mathscr{P}_N(\boldsymbol{X}_N, a_N, t_N; \boldsymbol{X}_{N-1}, a_{N-1}, t_{N-1}; \ldots; \boldsymbol{X}_1, a_1, t_1)}{P_N(\boldsymbol{X}_N, t_N; \boldsymbol{X}_{N-1}, t_{N-1}; \ldots; \boldsymbol{X}_1, t_1)}
\end{aligned}
\tag{60}
$$

as follows:

$$
\begin{aligned}
&\langle a_N^{k_N}(\boldsymbol{X}_N, t_N) \ldots a_1^{k_1}(\boldsymbol{X}_1, t_1)\rangle \\
&\quad = \int_0^\infty \cdots \int_0^\infty a_N^{k_N} \ldots a_1^{k_1} \\
&\qquad \times R_N(a_N, t_N; \ldots; a_1, t_1|\boldsymbol{X}_N, t_N; \ldots; \boldsymbol{X}_1, t_1)\,\mathrm{d}a_1 \ldots \mathrm{d}a_N
\end{aligned}
\tag{61}
$$

where P_N, $N=1, \ldots,$ are age-independent joint probabilities:

$$P_N(\boldsymbol{X}_N, t_N; \ldots; \boldsymbol{X}_1, t_1) = \int_0^\infty \cdots \int_0^\infty \mathscr{P}_N(\boldsymbol{X}_N, a_N, t_N; \ldots; \boldsymbol{X}_1, a_1, t_1)\,\mathrm{d}a_N \ldots \mathrm{d}a_1. \tag{62}$$

For illustration, in the following we shall compute the bitemporal correlation functions $\langle a_1 a_2 \rangle$ in the case of a stationary state. For steady states (58′) becomes

$$
\begin{aligned}
&\mathscr{P}_2^{\mathrm{st}}(\boldsymbol{X}_2, a_2, t_2; \boldsymbol{X}_1, a_1, t_1) \\
&\quad = h(\theta) P_1^{\mathrm{st}}(\boldsymbol{X}_1)\Omega(\boldsymbol{X}_1) \exp(-a_1\Omega(\boldsymbol{X}_1)) \\
&\qquad \times [h(|\theta| - a_2) \exp(-a_2\Omega(\boldsymbol{X}_2))(\Omega(\boldsymbol{X}_2) + \partial_{|\theta|}) G(\boldsymbol{X}_2, |\theta| - a_2|\boldsymbol{X}_1, 0) \\
&\qquad + h(a_2 - |\theta|) \exp(-|\theta|\Omega(\boldsymbol{X}_2))\delta(\boldsymbol{X}_2 - \boldsymbol{X}_1)\delta(a_2 - a_1 - |\theta|)] \\
&\qquad + h(-\theta) P_1^{\mathrm{st}}(\boldsymbol{X}_2)\Omega(\boldsymbol{X}_2) \exp(-a_2\Omega(\boldsymbol{X}_2)) \\
&\qquad \times [h(|\theta| - a_1) \exp(-a_1\Omega(\boldsymbol{X}_1))(\Omega(\boldsymbol{X}_1) + \partial_{|\theta|}) G(\boldsymbol{X}_1, |\theta| - a_1|\boldsymbol{X}_2, 0) \\
&\qquad + h(a_1 - |\theta|) \exp(-|\theta|\Omega(\boldsymbol{X}_1))\delta(\boldsymbol{X}_1 - \boldsymbol{X}_2)\delta(a_1 - a_2 - |\theta|)]
\end{aligned} \tag{63}
$$

where P_1^{st} is the stationary solution of the master equation (33) and

$$\theta = t_2 - t_1. \tag{64}$$

Combining equations (60)-(63), after simple but lengthy manipulations we come to

$$
\begin{aligned}
&\langle a_1(\boldsymbol{X}_1, t_1) a_2(\boldsymbol{X}_2, t_2) \rangle \\
&\quad = h(\theta)\Bigg(2(\Omega(\boldsymbol{X}_1))^{-2} \exp(-|\theta|\Omega(\boldsymbol{X}_1))\delta(\boldsymbol{X}_2 - \boldsymbol{X}_1) \\
&\qquad + (\Omega(\boldsymbol{X}_1))^{-1} \int_0^{|\theta|} \mathrm{e}^{-\lambda\Omega(\boldsymbol{X}_2)} G(\boldsymbol{X}_2, |\theta| - \lambda|\boldsymbol{X}_1, 0)\, \mathrm{d}\lambda \Bigg)[G(\boldsymbol{X}_1, |\theta||\boldsymbol{X}_2, 0)]^{-1} \\
&\qquad + h(-\theta)\Bigg(2(\Omega(\boldsymbol{X}_2))^{-2} \exp(-|\theta|\Omega(\boldsymbol{X}_2))\delta(\boldsymbol{X}_1 - \boldsymbol{X}_2) \\
&\qquad + (\Omega(\boldsymbol{X}_2))^{-1} \int_0^{|\theta|} \mathrm{e}^{-\lambda\Omega(\boldsymbol{X}_1)} G(\boldsymbol{X}_1, |\theta| - \lambda|\boldsymbol{X}_2, 0)\, \mathrm{d}\lambda \Bigg)[G(\boldsymbol{X}_2, |\theta||\boldsymbol{X}_1, 0)]^{-1}
\end{aligned} \tag{65}
$$

5. A generalisation

The next step of our analysis is to extend the above results to systems with memory for which the transition probabilities are age dependent:

$$W'\Delta t = W(\boldsymbol{X} \to \boldsymbol{X}'; a)\Delta t \qquad W''\Delta t = W(\boldsymbol{X} \to \boldsymbol{X}''; a)\Delta t, \ldots. \tag{66}$$

In this case the evolution equations (29) and (30), and (44) and (45) become

$$(\partial_{t_1} + \partial_{a_1})\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1) = -\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1) \int_{\boldsymbol{X}'} W(\boldsymbol{X}_1 \to \boldsymbol{X}'; a_1)\, \mathrm{d}\boldsymbol{X}' \tag{67}$$

$$\mathscr{P}_1(\boldsymbol{X}_1, 0, t_1) = \int_{\boldsymbol{X}'} \int_0^{\infty} W(\boldsymbol{X}' \to \boldsymbol{X}_1; a')\mathscr{P}(\boldsymbol{X}', a', t_1)\, \mathrm{d}\boldsymbol{X}'\, \mathrm{d}a' \tag{68}$$

and

$$\left(\int_{X'} W(X_n \to X'; a_n)\,dX' + \partial_{t_n} + \partial_{a_n}\right)\mathcal{G}_n(X_n, a_n, t_n|X_{n-1}, a_{n-1}, t_{n-1}; \ldots; X_1, a_1, t_1) = 0$$

$$n = 2, \ldots, N \quad (69)$$

$$\mathcal{G}_n(X_n, 0, t_n|X_{n-1}, a_{n-1}, t_{n-1}; \ldots; X_1, a_1, t_1) = \int_{X'}\int_0^\infty W(X' \to X_n; a') \times \mathcal{G}_n(X', a', t_n|X_{n-1}, a_{n-1}, t_{n-1}; \ldots; X_1, a_1, t_1)\,dX'da'$$

$$n = 2, \ldots, N. \quad (70)$$

From (46), (69) and (70) it turns out that the relations (47) remain valid and then the equations (69) and (70) may be written in a form similar to (48) and (49):

$$(\partial_t + \partial_a)\mathcal{G}(X, a, t - t''|X'', a'', 0) + \mathcal{G}(X, a, t - t''|X'', a'', 0)\int_{X'} W(X \to X'; a)\,dX' = 0 \quad (71)$$

$$\mathcal{G}(X, 0, t - t''|X'', a'', 0) = \int_{X'}\int_0^\infty W(X' \to X; a')\mathcal{G}(X', a', t - t''|X'', a'', 0)\,dX'\,da'. \quad (72)$$

Unfortunately, in this case, the determination of $\mathcal{P}_1(X_1, a_1, t)$ and $\mathcal{G}(X', a', t' - t''|X'', a'', 0)$ cannot be reduced to the integration of a phenomenological master equation. However, by integrating (67) with the initial condition (31) we obtain

$$\mathcal{P}_1(X_1, a_1, t) = h(t_1 - t_0 - a_1)Z(X_1, t_1 - a_1)\Psi(X_1, a_1) + h(a_1 - t_1 + t_0)\mathcal{P}_1^0(X_1, a_1 - t_1 + t_0)\frac{\Psi(X_1, a_1)}{\Psi(X_1, a_1 - t_1 + t_0)} \quad (73)$$

where

$$Z(X_1, t_1) = \mathcal{P}_1(X_1, 0, t_1) \quad (74)$$

and

$$\Psi(X_1, a_1) = \exp\left(-\int_{\mu=0}^{a_1}\int_{X'} W(X_1 \to X'; \mu)\,d\mu\,dX'\right). \quad (75)$$

Substituting (73) into (68) leads to a linear integral equation for $Z(X, t)$:

$$Z(X, t) = \int_{a'=0}^{t-t_0}\int_{X'} W(X' \to X; a')\Psi(X', a')Z(X', t - a')\,da'\,dX' + \int_{a'=t-t_0}^{\infty}\int_{X'} W(X' \to X; a')\frac{\Psi(X', a')}{\Psi(X', a' - t + t_0)}\mathcal{P}^0(X', a' - t + t_0)\,da'\,dX'. \quad (76)$$

By applying a similar approach equations (46), (71) and (72) give

$$\mathcal{G}(X, a, t - t''|X'', a'', 0) = \mathcal{G}(X, 0, t - t'' - a|X'', a'', 0)\Psi(X, a)h(t - t_0 - a) + h(a - t + t_0)\frac{\Psi(X, a)}{\Psi(X, a'')}\delta(a - t + t_0 - a'')\delta(X - X'') \quad (77)$$

where the function $\mathscr{G}(\boldsymbol{X}, 0, t-t''|\boldsymbol{X}'', a'', 0)$ is the solution of the integral equation

$$\begin{aligned}
&\mathscr{G}(\boldsymbol{X}, 0, t-t''|\boldsymbol{X}'', a'', 0)\\
&\quad=\int_{\boldsymbol{X}'}\int_0^{t-t''} W(\boldsymbol{X}'\to\boldsymbol{X}; a')\Psi(\boldsymbol{X}', a')\mathscr{G}(\boldsymbol{X}', 0, t-t''-a'|\boldsymbol{X}'', a'', 0)\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'\\
&\quad\quad+W(\boldsymbol{X}''\to\boldsymbol{X}; a''+t-t'')\frac{\Psi(\boldsymbol{X}'', a''+t-t'')}{\Psi(\boldsymbol{X}'', a'')}.
\end{aligned}\tag{78}$$

Examining equations (76) and (78) we notice that $\mathscr{G}(\boldsymbol{X}, 0, t-t_0|\boldsymbol{X}_0, a_0, 0)$ is the Green function attached to the integral equation (76). Thus

$$Z(\boldsymbol{X}, t)=\int_{\boldsymbol{X}_0}\int_{a_0=0}^{\infty}\mathscr{G}(\boldsymbol{X}, 0, t-t_0|\boldsymbol{X}_0, a_0, 0)\mathscr{P}^0(\boldsymbol{X}_0, a_0)\,\mathrm{d}\boldsymbol{X}_0\,\mathrm{d}a_0. \tag{79}$$

From (43), (47), (73), (77) and (79) we can express the joint probabilities $\mathscr{P}_2, \mathscr{P}_3, \ldots, \mathscr{P}_N$ in terms of $\mathscr{G}(\boldsymbol{X}, 0, t-t_0|\boldsymbol{X}_0, a_0, 0)$:

$$\begin{aligned}
&\mathscr{P}_N(\boldsymbol{X}_N, a_N, t_N; \ldots; \boldsymbol{X}_1, a_1, t_1)\\
&\quad=\Bigg(h(t_1-t_0-a_1)\Psi(\boldsymbol{X}_1, a_1)\int_{\boldsymbol{X}''}\int_{a''=0}^{\infty}\mathscr{G}(\boldsymbol{X}_1, 0, t_1-t_0-a_1|\boldsymbol{X}'', a'', 0)\\
&\quad\quad\times\mathscr{P}_1^0(\boldsymbol{X}'', a'')\,\mathrm{d}\boldsymbol{X}''\,\mathrm{d}a''+h_1(a_1-t_1+t_0)\\
&\quad\quad\times\frac{\Psi(\boldsymbol{X}_1, a_1)}{\Psi(\boldsymbol{X}_1, a_1-t_1+t_0)}\mathscr{P}_1^0(\boldsymbol{X}_1, a_1-t_1+t_0)\Bigg)\\
&\quad\quad\times\prod_{q=2}^{N}\Bigg(h(t_q-t_{q-1}-a_q)\mathscr{G}(\boldsymbol{X}_q, 0, t_q-t_{q-1}-a_q|\boldsymbol{X}_{q-1}, a_{q-1}, 0)\Psi(\boldsymbol{X}_q, a_q)\\
&\quad\quad+h(a_q-t_q+t_{q-1})\delta(a_q-t_q+t_{q-1}-a_{q-1})\delta(\boldsymbol{X}_q-\boldsymbol{X}_{q-1})\frac{\Psi(\boldsymbol{X}_q, a_q)}{\Psi(\boldsymbol{X}_{q-1}, a_{q-1})}\Bigg)\\
&\quad\quad t_N\geqslant t_{N-1}\geqslant\ldots\geqslant t_1\geqslant t_0.
\end{aligned}\tag{80}$$

6. Comparison with the CTRW theory

To outline the analogies between the ADME formalism and the theory of continuous time random walks, we shall derive the equation for the state probability $\mathscr{P}_1(\boldsymbol{X}_1, a_1, t_1)$ avoiding the use of the ADME system. To do this, we shall introduce the following probabilities:

(i) the probability that the time between two successive transitions is between a and $a+\mathrm{d}a$ and that the state of the system after the transition is $\boldsymbol{X}'$ provided that the initial state is $\boldsymbol{X}$:

$$p\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a=p(\boldsymbol{X}\to\boldsymbol{X}'; a)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a \tag{81}$$

$$\int_{\boldsymbol{X}'}\int_0^{\infty}p(\boldsymbol{X}\to\boldsymbol{X}'; a)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a=1 \tag{82}$$

(ii) the probability that a transition was made to $\boldsymbol{X}$ between times t, $t+\mathrm{d}t$:

$$\eta_1(\boldsymbol{X}, t)\,\mathrm{d}t=\mathscr{P}_1(\boldsymbol{X}, a=0, t)\,\mathrm{d}t=Z(\boldsymbol{X}, t)\,\mathrm{d}t \tag{83}$$

(iii) the probability that in the age interval a_0, a the system rests in the state $\boldsymbol{X}$:

$$\gamma = \gamma(\boldsymbol{X}, a, a_0) \tag{84}$$

(iv) the probability that in the age interval a_0, a the system rests in the state $\boldsymbol{X}$ and that in the interval $a, a+\mathrm{d}a$ a transition occurs to $\boldsymbol{X}'$:

$$\Gamma\,\mathrm{d}a = \Gamma(\boldsymbol{X}, a, a_0, \boldsymbol{X}')\,\mathrm{d}a. \tag{84'}$$

According to their definitions, these probabilities fulfil the following relationships:

$$\gamma(\boldsymbol{X}, a, a_0)\gamma(\boldsymbol{X}, a_0, 0) = \gamma(\boldsymbol{X}, a, 0) \tag{85}$$

$$\gamma(\boldsymbol{X}, a, 0) = 1 - \int_{\boldsymbol{X}'}\int_{a'=0}^{a} p(\boldsymbol{X}\to\boldsymbol{X}', a')\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a' \tag{86}$$

$$p(\boldsymbol{X}\to\boldsymbol{X}'; a) = \Gamma(\boldsymbol{X}, a, a_0, \boldsymbol{X}')\gamma(\boldsymbol{X}, a_0, 0). \tag{87}$$

Observing that $\gamma(\boldsymbol{X}, a, 0)$ obeys the balance equations

$$p(\boldsymbol{X}\to\boldsymbol{X}'; a) = \gamma(\boldsymbol{X}, a, 0)\,W(\boldsymbol{X}\to\boldsymbol{X}'; a) \tag{88}$$

$$\gamma(\boldsymbol{X}, a+\Delta a, 0) = \gamma(\boldsymbol{X}, a, 0)\left(1 - \int_{\boldsymbol{X}'} W(\boldsymbol{X}\to\boldsymbol{X}'; a)\Delta a\,\mathrm{d}\boldsymbol{X}\right) \qquad \gamma(\boldsymbol{X}, 0, 0) = 1 \tag{89}$$

and making use of (85)-(87) we can express the functions $p(\boldsymbol{X}\to\boldsymbol{X}'; a)$ and $\gamma(\boldsymbol{X}, a, 0)$ in terms of the transition rates $W(\boldsymbol{X}\to\boldsymbol{X}', a)$. Indeed, from (85)-(87), and (88) and (89) we come to

$$\partial_a\gamma(\boldsymbol{X}, a, 0) = -\gamma(\boldsymbol{X}, a, 0)\int_{\boldsymbol{X}'} W(\boldsymbol{X}\to\boldsymbol{X}'; a)\,\mathrm{d}\boldsymbol{X}' \qquad \gamma(\boldsymbol{X}, 0, 0) = 1 \tag{90}$$

$$\gamma(\boldsymbol{X}, a, 0) = \exp\left(-\int_{\boldsymbol{X}'}\int_0^a W(\boldsymbol{X}\to\boldsymbol{X}'; \mu)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}\mu\right) = \Psi(\boldsymbol{X}, a) \tag{91}$$

$$p(\boldsymbol{X}\to\boldsymbol{X}'; a) = W(\boldsymbol{X}\to\boldsymbol{X}', a)\exp\left(-\int_{\boldsymbol{X}'}\int_0^a W(\boldsymbol{X}\to\boldsymbol{X}'; \mu)\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}\mu\right). \tag{92}$$

Making use of the initial condition (31), the probability $\mathscr{P}_1(\boldsymbol{X}, a, t)$ may be expressed as

$$\mathscr{P}_1(\boldsymbol{X}, a, t) = \begin{cases} \eta_1(\boldsymbol{X}, t-a)\gamma(\boldsymbol{X}, a, 0) & t-t_0 \geqslant a \\ \mathscr{P}_1^0(\boldsymbol{X}, a-t+t_0)\gamma(\boldsymbol{X}, a, a-t+t_0) & t-t_0 \leqslant a \end{cases} \tag{93}$$

corresponding to the following two possibilities:

(a) a transition occurred to the state $\boldsymbol{X}$ at a moment $t-a \leqslant t$ and no further transitions took place;

(b) the system remains in its initial state $\boldsymbol{X}$.

By applying a similar approach, we come to the following balance equation for the function $\eta_1(\boldsymbol{X}, t) = \mathscr{P}_1(\boldsymbol{X}, 0, t)$:

$$\eta_1(\boldsymbol{X}, t) = \int_{\boldsymbol{X}'}\int_0^{t-t_0} \eta(\boldsymbol{X}', t-a')p(\boldsymbol{X}'\to\boldsymbol{X}; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'$$
$$+ \int_{\boldsymbol{X}'}\int_{t-t_0}^{\infty} \mathscr{P}_1^0(\boldsymbol{X}', a'-t+t_0)\Gamma(\boldsymbol{X}', a', a'-t+t_0, \boldsymbol{X})\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'. \tag{94}$$

Taking into account (85)-(87), and (91) and (92), from (93) and (94) we come to (73) and (76), respectively. Thus we recover the main results of the ADME theory. The above method would be applied to derive the evolution equations for the joint probabilities ((78)-(80)) as well. The detailed calculation is tedious and is left to the reader, as an exercise.

Using (85)-(87), the evolution equations (93) and (94) may be written in an alternative form:

$$\mathscr{P}_1(\boldsymbol{X}, a, t) = \eta_1(\boldsymbol{X}, t-a)\left(1 - \int_{\boldsymbol{X}'}\int_{a'=0}^{a} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a'\right)h(t-t_0-a)$$
$$+\frac{1-\int_{\boldsymbol{X}'}\int_{a'=0}^{a} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'}{1-\int_{\boldsymbol{X}'}\int_{a'=0}^{a-t+t_0} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'}\mathscr{P}_1^0(\boldsymbol{X}, a-t+t_0)h(a-t+t_0) \qquad (95)$$

$$\eta_1(\boldsymbol{X}, t) = \int_{\boldsymbol{X}'}\int_0^{t-t_0} \eta_1(\boldsymbol{X}', t-a')p(\boldsymbol{X}'\to\boldsymbol{X}; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}' + \int_{\boldsymbol{X}'}\int_{t-t_0}^{\infty} p(\boldsymbol{X}'\to\boldsymbol{X}; a')$$
$$\times\left(1-\int_{\boldsymbol{X}''}\int_{a''=0}^{a'-t+t_0} p(\boldsymbol{X}'\to\boldsymbol{X}''; a'')\,\mathrm{d}\boldsymbol{X}''\,\mathrm{d}a''\right)^{-1}\mathscr{P}_1^0(\boldsymbol{X}', a'-t+t_0)\,\mathrm{d}a'\mathrm{d}\boldsymbol{X}' \qquad (96)$$

from which we may derive the following equation for the age-independent state probability:

$$P_1(\boldsymbol{X}, t) = \int_0^{t-t_0} \mathrm{d}a\, \eta_1(\boldsymbol{X}, t-a)\left(1-\int_{\boldsymbol{X}'}\int_{a'=0}^{a} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}\boldsymbol{X}'\,\mathrm{d}a'\right)$$
$$+\int_{t-t_0}^{\infty}\frac{1-\int_{\boldsymbol{X}'}\int_{a'=0}^{a} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'}{1-\int_{\boldsymbol{X}'}\int_{a'=0}^{a-t+t_0} p(\boldsymbol{X}\to\boldsymbol{X}'; a')\,\mathrm{d}a'\,\mathrm{d}\boldsymbol{X}'}\mathscr{P}_1^0(\boldsymbol{X}, a-t+t_0)\,\mathrm{d}a. \qquad (97)$$

Examining (96) and (97) we see that the ADME theory is a generalisation of the theory of continuous time random walks (Gartner 1986). Indeed, if the state variables $\boldsymbol{X}_i$ are discrete and

$$p(\boldsymbol{X}\to\boldsymbol{X}'; a) = p(\boldsymbol{X}'-\boldsymbol{X}; a) \qquad (98)$$

$$\mathscr{P}_1(\boldsymbol{X}, a, t=t_0) = \delta_{\boldsymbol{X}0}\varphi(a) \qquad \int_0^{\infty}\varphi(a)\,\mathrm{d}a = 1 \qquad (99)$$

the ADME formalism reduces to CTRW. To prove this assertion we shall write the unitemporal evolution equations (96) and (97) in the form

$$P_1(\boldsymbol{X}, t) = \delta_{\boldsymbol{X}0}\Psi_1(t-t_0) + \int_0^{t-t_0}\eta_1(\boldsymbol{X}, t-a)\Psi(t-a)\,\mathrm{d}a \qquad (100)$$

and

$$\eta_1(\boldsymbol{X}, t) = p_1(\boldsymbol{X}; t-t_0) + \sum_{\boldsymbol{X}'}\int_0^{t-t_0}\eta_1(\boldsymbol{X}', t-a)p(\boldsymbol{X}-\boldsymbol{X}'; a)\,\mathrm{d}a \qquad (101)$$

where

$$\Psi(a) = \sum_{\boldsymbol{X}'}\int_{a'=a}^{\infty} p(\boldsymbol{X}'; a')\,\mathrm{d}a' \qquad (102)$$

and

$$\Psi_1(t-t_0)=\int_{t-t_0}^{\infty}\varphi(a-t+t_0)\frac{\Psi(a)}{\Psi(a-t+t_0)}\,\mathrm{d}a \tag{103}$$

$$p_1(\boldsymbol{X};t-t_0)=\int_{t-t_0}^{\infty}\varphi(a-t+t_0)\frac{p(\boldsymbol{X};a)}{\Psi(a-t+t_0)}\,\mathrm{d}a. \tag{104}$$

Equations (100) and (101) are identical with the basic equations of the CTRW theory, as written by Weiss and Rubin (1983). However, the ADME formalism is more general than the CTRW, since by means of (78) and (80) we can compute the expressions for the multitemporal joint probabilities P_n, $n=2,3,\ldots$, or $\mathscr{P}_n$, $n=2,3,\ldots$. As far as we know, no such computation has been performed within the framework of CTRW.

7. Conclusions

We have considered Markovian systems described by a phenomenological master equation (PME, Van Kampen 1981). We have determined the joint probabilities of the ages of different fluctuation states in terms of the Green functions attached to the phenomenological master equation. Our approach may be applied to physicochemical processes described by a PME such as chemical fluctuations, semiconductor noise, etc. The work on chemical fluctuations is in progress and will be presented in another paper.

We have extended the method to the case of age dependent transition rates. In this case the theory may be formulated by means of a linear integral equation and the behaviour of the random variable $\boldsymbol{X}$ is generally non-Markovian. However, even if the transition rates are age dependent the time evolution of the random vector $(\boldsymbol{X}, a)$ is Markovian (see (69) and (70)). This approach may be viewed as a generalisation of the well known continuous time random walk theory and would be applied to describe the memory effects.

Acknowledgments

The author is grateful to Paul Gartner for helpful discussions and for drawing attention to the similarity between the ADME and CTRW formalisms. This work has been supported by CIMC Bucuresti.

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