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On the Stationary Distribution of a Stochastic Chemical Process without Detailed Balance

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An exact expression of the stationary distribution is found for a particular chemical model without detailed balance. An analytical approximation of this solution is obtained for small values of the concentration. It is shown that the WKB continuous approximation of the distribution is valid for all values of the concentration.

KEY WORDS: Stochastic chemical processes; master equation; exact stationary solution; WKB approximation.

1. INTRODUCTION

The birth-and-death formalism has been widely used in the stochastic theory of chemical reactions$^{(1-4)}$ as well as in many other fields of physics and natural sciences.$^{(3)}$ A master equation describes the evolution of the system; its stationary solution serves as a basis for the study of the asymptotical behavior, and for many other purposes.$^{(5,6)}$ Unfortunately the steady state is known explicitly only for step-by-step processes, and in some particular cases$^{(2)}$; otherwise one has to make use of some approximation. In fact the exact steady state is generally replaced by the stationary solution of a continuous approximation of the master equation, such as a Fokker–Planck equation$^{(7,9)}$ or the so-called WKB approximation.$^{(8,10)}$

In this paper we study a particular case of non-step-by-step chemical process where the previous problem may be solved exactly, and the stationary solution of its master equation is written explicitly.

Then this solution is compared with the distribution obtained by the WKB approximation. It is shown that this method yields a correct represen-

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tation of the steady state for all values of the concentrations at the thermodynamic limit, which confirms the conclusions of other authors(6,10) for step-by-step processes.

2. A SOLUBLE CASE OF CHEMICAL PROCESS WITHOUT DETAILED BALANCE

2.1. General Theory

Let the stochastic variable X represent the number of particles of some chemical species in a reacting system. If X is the only variable and if the mixture is supposed to remain homogeneous, the stochastic state of the system is described by the probability distribution $\mathbf{p} = (p_i)$, p_i being the probability that $X = i$.

According to the hypothesis of the birth-and-death formalism,(1,4) (p_i) obeys a master equation which may be written

$$\frac{d}{dt} p_i = \sum_{\substack{r=-n \\ r\neq 0}}^{m} (W_{i-r}^{r} p_{i-r} - W_i^r p_i) \tag{1}$$

where W_i^r is the transition rate form $X = i$ to $X = i + r$, due to one of the possible reactions. The transition amplitude r ranges from $-n$ to m, n and m being usually small positive integers.

The stationary solution of Eq. (1) is easily found for step-by-step reactions, where $s = r = 1$, since the detailed balance equation

$$\varphi_i(\mathbf{p}) \equiv W_i^{-1} p_i - W_{i-1}^{1} p_{i-1} = 0 \tag{2}$$

then determines the steady state(2,5) and allows the calculation of all p_i by recurrence.

Such a simple relation does not exist for non-step-by-step reactions. However, it may be noticed that Eq. (2) may be generalized to any chemical system, under the form

$$\varphi_i(\mathbf{p}) \equiv \sum_{r=-n+1}^{m} R_{i-r}^{r} p_{i-r} = 0 \qquad \text{for every } i \tag{3}$$

with

$$\begin{aligned} R_i^r &= \sum_{j=-n}^{r-1} W_i^j \qquad \text{if } -n \leqslant r \leqslant 0 \\ &= -\sum_{j=r}^{m} W_i^j \qquad \text{if } 1 \leqslant r \leqslant m \end{aligned} \tag{4}$$

Indeed it is easily verified that the right-hand side of (1) equals $\varphi_{i+1}(\mathbf{p}) - \varphi_i(\mathbf{p})$ and that $\varphi_0(\mathbf{p}) = 0$; thus $\varphi_i(\mathbf{p}) = 0$ for all i in the stationary state.

The recurrence relation (3) is apparently more complicated than the relation obtained from (1) in the stationary state, but its order is lower, since in φ_i the index of p runs only from $-n+1$ to m.

In fact this advantage is generally not very useful apart from numerical evaluations, since in most cases the explicit solution of (3) is not known when n or $m \neq 1$.

We now describe a case where this solution may be found exactly.

2.2. An Exactly Soluble Case

Let us consider a chemical system where $n = 1$ and $m = 2$ (or the contrary case), such as

$$\begin{aligned} A + X &\rightleftarrows Y + C \\ B + 2Y &\rightleftarrows 2X + D \end{aligned} \tag{5}$$

where the concentrations of the species A, B, C, D are kept constant. The total number N of molecules X and Y also remains constant according to (5), and the state of the system may be defined by the number of molecules X only.

Although the reverse reactions $\leftarrow$ are always present in principle, we shall assume that they may be neglected, which is a reasonable approximation if for instance the concentrations of C and D are maintained to very low values. Thus we obtain a theoretical system similar to the system studied by Görtz and Walls[11,12]:

$$\begin{aligned} A + X &\xrightarrow{W^{-1}} Y \\ B + 2Y &\xrightarrow{W^{2}} 2X \end{aligned} \tag{5'}$$

The master equation reads in this case

$$\frac{d}{dt} p_i = W_{i+1}^{-1} p_{i+1} - \left(W_i^{-1} + W_i^2\right) p_i + W_{i-2}^2 p_{i-2} \tag{6}$$

and Eq. (3) becomes

$$\begin{aligned} \varphi_i(\mathbf{p}) &\equiv -W_{i-2}^2 p_i - W_{i-1}^2 p_{i-1} + W_i^{-1} p_i = 0 \qquad (i \geqslant 2) \\ \varphi_1(\mathbf{p}) &\equiv -W_0^2 p_0 + W_1^{-1} p_1 = 0 \end{aligned} \tag{7}$$

Let us write

$$q_i = W_i^2 p_i \tag{8}$$

and

$$\alpha_i = W_i^2 / W_i^{-1} \tag{9}$$

Then the recurrence equations (7) take the simple form

$$\begin{aligned} q_1 &= \alpha_1 q_0 \\ q_{i+1} &= \alpha_{i+1}(q_i + q_{i-1}) \qquad (i \geqslant 1) \end{aligned} \tag{10}$$

which suggests that we introduce the new variables

$$\begin{aligned} s_0 &= q_0 \\ s_i &= \left(\prod_{j=1}^{i} \alpha_i \right)^{-1} q_i \qquad (i \geqslant 1) \end{aligned} \tag{11}$$

which satisfy the relations

$$s_i = s_{i-1} + \beta_{i-1} s_{i-2} \tag{12}$$

with

$$\beta_i = (\alpha_i)^{-1} = W_i^{-1} / W_i^2 \tag{13}$$

Taking $s_0 = q_0 = 1$, we obtain

$$\begin{aligned} s_1 &= 1 \\ s_2 &= 1 + \beta_1 \\ s_3 &= 1 + \beta_1 + \beta_2 \\ s_4 &= 1 + \beta_1 + \beta_2 + \beta_3 + \beta_1 \beta_3 \end{aligned}$$

and in general

$$s_i = \sum_{k=0}^{[i/2]} \textstyle\sum_{(I_n^k)} \beta_{i_1} \beta_{i_2} \cdots \beta_{i_k} \tag{14}$$

where

$$\begin{aligned} [i/2] &= i/2 && \text{if } i \text{ is even} \\ &= (i-1)/2 && \text{if } i \text{ is odd} \end{aligned} \tag{15}$$

$\sum_{(I_n^k)}$ denotes the sum over all the sets I_n^k of k increasing indices (i_1, $i_2, \ldots, i_k$) chosen among $1, 2, \ldots, n-1$, and such that the difference between two successive indices is at least 2.

For instance if $n = 6$,

$$\begin{aligned} (I_6^2) &= (1,3) + (1,4) + (1,5) + (2,4) + (2,5) + (3,5) \\ (I_6^3) &= (1,3,5) \end{aligned}$$

Formula (14) is established by recurrence, noticing that if $k \leqslant [(n-1)/2]$,

$$(I_n^k) = (I_{n-1}^k) + (I_{n-2}^k, n) \tag{16}$$

and if $[(n-1)/2] < k \leqslant [n/2]$, which implies $n = 2n'$ and $k = n'$,

$$(I_{2n'}^{n'}) = (I_{2n'-2}^{n'-1}, 2n') \tag{17}$$

Equations (8), (11), and (14) determine completely the stationary solution (p_i), apart from a normalization factor.

It remains to give them a more useful analytic form, which is done in the following section for low densities.

3. CONTINUOUS APPROXIMATIONS OF THE STEADY STATE

3.1. Analytical Approximation of the Exact Steady State for Low Concentrations

Let Ω be the volume of the system, and $x = X/\Omega$ be the concentration of X. For large Ω, x may be considered a continuous variable. By switching from sums to integrals in formula (14) one obtains

$$s_i = \sum_{k=0}^{[i/2]} \Omega^k \int_0^x dt_1\, \beta(t_1) \int_{t_1+2\epsilon}^x dt_2\, \beta(t_2) \cdots \int_{t_{k-1}+2\epsilon}^x dt_k\, \beta(t_k) \tag{18}$$

where $\epsilon = \Omega^{-1}$, and $\beta(t)$ is the continuous version of $\beta_i = W_i^{-1}/W_i^2$. For reactions (5) the general properties of the transition rates$^{(8,9)}$ imply

$$\begin{aligned} W_i^{-1} &= \Omega k^{-1} a x \\ W_i^2 &= \Omega k^2 b(n-x)(n-x-\epsilon) \end{aligned} \tag{19}$$

where k^{-1} and k^2 are the macroscopic rate constants, and $a = A/\Omega$, $b = B/\Omega$. Thus

$$\beta(x) = k^{-1}a(k^2 b)^{-1} x(n-x)^{-1}(n-x-\epsilon)^{-1} \tag{20}$$

Let us assume that x tends to 0 with ϵ, and more precisely that x is of order $\Omega^{-\nu}$ for some $\nu > 0$. We may define

$$\tilde{x} = \Omega^{\nu} x = O(1) \tag{21}$$

and

$$\tilde{\beta}(\tilde{x}) = \beta(\tilde{x}\Omega^{-\nu}) = \Omega^{-\nu}\gamma(x) \tag{22}$$

Then $\gamma(x)$ admits a finite limit as $\Omega \to \infty$ and Eq. (18) takes the asymptotic form

$$s_i = \sum_{k=0}^{[i/2]} \Omega^{k-2\nu} \int_0^{\tilde{x}} dt_1\, \gamma(t_1) \cdots \int_{t_{k-1}+2\epsilon^{1-\nu}}^{\tilde{x}} dt_k\, \gamma(t_k) \tag{23}$$

The right member of (23) may be written

$$\int_0^{\tilde{x}} dt_1\, \gamma(t_1) \int_0^{\tilde{x}} dt_2\, \gamma(t_2) \cdots \int_0^{\tilde{x}} dt_k\, \gamma(t_k)\left[1 + O(\epsilon^{1-\nu})\right]$$

$$= \frac{1}{k!}\left[\int_0^{\tilde{x}} dt\, \gamma(t)\right]^k \left[1 + O(\epsilon^{1-\nu})\right] \tag{24}$$

which gives

$$s_i = \sum_{k=0}^{[i/2]} \frac{\Omega^{k(1-2\nu)}}{k!}\left[\int_0^{\tilde{x}} dt\, \gamma(t)\right]^k \left[1 + O(\epsilon^{1-\nu})\right] \tag{25}$$

for $\nu = 1/2$, (25) yields for large i

$$s_i = \exp\left[\int_0^{\tilde{x}} dt\, \gamma(t)\right] \tag{26}$$

which, in terms of x, becomes

$$s_i = \exp\left[\Omega \int_0^{x} dt\ \beta(t)\right] \qquad (x \sim \Omega^{-1/2}) \tag{27}$$

Then (11) and (14) yield the following continuous expression for the stationary solution of the master equation when $x \sim \Omega^{-1/2}$:

$$p_{\mathrm{st}}(x) \propto \frac{1}{W^2(x)} \exp\left\{\Omega \int_0^{x} \left[-\ln \beta(t) + \beta(t)\right] dt\right\} \tag{28}$$

which may be written

$$p_{\mathrm{st}}(x) \propto \exp\left[\Omega U(x)\right] \qquad (x \sim \Omega^{-1/2}) \tag{29}$$

with

$$U(x) = \int_0^{x} \left[-\ln \beta(t) + \beta(t)\right] dt \tag{29′}$$

It should be pointed out that expressions (28) and (29) are by no means valid for finite values of x. In particular they have no maximum for the macroscopic value of x [determined by $\beta(x) = 2$] as it should be for $p_{\mathrm{st}}(x)$, but this point does not affect the validity of (28) and (29) for low concentrations.

These asymptotical expressions will be useful for comparison with the results of the WKB approximation of the master equation.

3.2. The Continuous Approximations of the Master Equation

There are two classical methods to get a continuous approximation of the master equation at the thermodynamic limit: $\Omega \to \infty$, $x \to \infty$, $x = X/\Omega = \text{const}$.

The first one is to transform the master equation into a Fokker–Planck equation(6,7): this method is satisfying for finite times. However, the stationary distribution obtained in this way may differ appreciably from the exact result(6); *a fortiori*, it should not be expected to agree with this result when $x \sim \Omega^{-1/2}$, since the method no longer applies. It may indeed be shown by working out the Fokker–Planck equation for reactions (5) that the resulting steady state does not satisfy (29). This is not surprising; the point is that the following approximation still holds in these conditions.

This method, which may be called by analogy the WKB approximation, consists in writing the probability distribution in the asymptotic form given by Kubo *et al.*(8) at the thermodynamic limit:

$$p(x) \propto \exp\left[\Omega U(x/\Omega)\right] \tag{30}$$

where $U(x)$ is supposed to be analytic.

Recently Lemarchand(10) proposed a systematic application of this method by expanding $U(x)$ in powers of $\epsilon = \Omega^{-1}$:

$$U(x) = U^0(x) + \epsilon U^1(x) + \cdots \tag{31}$$

In this way he obtained the exact steady state of step-by-step processes.

If we apply this method to reactions (5), the master equation (6) yields the following equation in the stationary case:

$$W^2(x)\exp\left(-\frac{\partial U^0}{\partial x}\right) + W^2(x) - W^{-1}(x)\exp\left(\frac{\partial U^0}{\partial x}\right) = 0 \tag{32}$$

which leads to

$$U^0(x) = \int_0^x \ln\left\{\frac{1 + \left[1 + 4\beta(t)\right]^{1/2}}{2\beta(t)}\right\} dt \tag{33}$$

and

$$p_{\text{st}}(x) \propto \exp\left[\Omega U^0(x)\right] \tag{34}$$

It is easily verified that expression (33) presents a maximum, as expected, when x equals the macroscopic stationary concentration $\hat{x}$, which is realized for $\beta = 2$. Moreover, when the scaled variable $q = (x - \hat{x})\Omega^{1/2}$ is introduced, the distribution (33) becomes Gaussian if $\Omega \to \infty$:

$$p_{\text{st}}(q) \propto \exp\left[\tfrac{1}{2} U''^0(\hat{x}) q^2\right]$$

with

$$U''^{0}(\hat{x}) = \frac{d^2U^0}{dx^2}(\hat{x}) = -\frac{1}{3}\frac{d\beta}{dx}(\hat{x})$$

in agreement with the general results of Kurtz(7) or Keizer,(15,16) which yield

$$p_{\text{st}}(q) \propto \exp\left[\tfrac{1}{2}U''(\hat{x})q^2\right]$$

with

$$U''(\hat{x}) = \frac{d^2}{d\hat{x}^2}\left[\int_0^{\hat{x}} 2\frac{2-\beta(t)}{4+\beta(t)}\,dt\right] = -\frac{1}{3}\frac{d\beta}{dx}(\hat{x})$$

In principle, the WKB method should only apply for finite values of x. However, it is seen that expression (34) does agree with the continuous representation (28) of the exact steady state when $x \sim \Omega^{-1/2}$.

Indeed, when $x \ll 1$, or equivalently $\beta \ll 1$, (33) may be written

$$U^0 = \int_0^x \left[-\ln\beta(t) + \beta(t) + O(\beta^2)\right]dt \tag{35}$$

which is identical to (29′).

Thus the WKB approximation still holds for low concentrations. In fact it will be shown in Section 4 that this approximation holds for any value of the concentration at the thermodynamic limit.

4. A GENERAL RESULT ON THE WKB APPROXIMATION

In Section 2 we have obtained the exact stationary solution of the master equation by introducing the quantity $q_i = W_i^2 p_i$, which satisfies Eq. (10):

$$q_i = \alpha_i(q_{i-1} + q_{i-2}) \qquad \text{if } i \geqslant 2 \tag{36}$$

Görtz(11) proposed that the master equation be approximated by means of the function

$$r_i = q_i/q_{i-1} \tag{37}$$

which is supposed to vary slowly with i. Thus one may write

$$r_{i+1} = r_i + O(\Omega^{-1}) \tag{38}$$

Ω being the volume of the system.

The quantity r_i verifies the exact equation

$$r_{i+1} = \alpha_{i+1}(1 + 1/r_i) \tag{39}$$

but the present approximation amounts to replace r_i by the solution r_i^0 of the equation

$$r_i = \alpha_i(1 + 1/r_i) \tag{40}$$

Thus

$$r_i^0 = (\alpha_i/2)\left[1 + (1 + 4/\alpha_i)^{1/2}\right] \tag{41}$$

and the corresponding approximation for q_i is

$$q_i^0 \propto \prod_{j=2}^{i+1} r_i^0 \tag{42}$$

but the relation between q_i and q_i^0 is not clear at the thermodynamic limit: $\Omega \to \infty$, $i \to \infty$, $i/\Omega = x$.

However it is shown in Appendix A that under reasonable assumptions the exact solution of (39) satisfies the inequalities

$$r_{i+1}^0 < r_i < r_i^0 \tag{43}$$

Thus, taking $q_i = 1$, we have

$$\frac{r_{i+1}^0}{r_1^0} q_i^0 = \prod_{j=2}^{i+1} r_i^0 < q_i = \prod_{j=1}^{i} r_i < \prod_{j=1}^{i} r_i^0 = q_i^0 \tag{44}$$

At the thermodynamic limit the hypothesis (38) leads to the WKB approximation, which thus yields a continuous estimate $q^0(x)$ of q_i^0 for $x = i/\Omega$.

Then

$$q_i \propto \exp \Omega U(x)$$

$$q_i^0 \propto \exp \Omega U^0(x)$$

and (24) implies

$$U^0(x) + \Omega^{-1} \log \frac{r^0(x)}{r^0(0)} < U(x) < U^0(x) \tag{45}$$

Therefore $U^0(x) \sim U(x)$ as $\Omega \to \infty$, which establishes the validity of the WKB method and confirms the results obtained in Section 3 by a direct comparison of the solutions.

APPENDIX A

Proof of inequalities (43): $r_{i+1}^0 < r_i < r_i^0 \cdot r_i^0$ is the positive solution of Eq. (40),

$$r_i^0 = \alpha_i(1 + 1/r_i^0) \tag{A1}$$

or

$$r_i^0 = \left[\alpha_i + (\alpha_i^2 + 4\alpha_i)^{1/2}\right]/2 \tag{A2}$$

while $r_i = q_i/q_{i-1}$ satisfies the recurrence relations

$$\begin{aligned} r_1 &= \alpha_1 \\ r_{i+1} &= \alpha_{i+1}(1 + 1/r_i) \qquad (i \geqslant 1) \end{aligned} \tag{A3}$$

α_i being defined by Eq. (9):

$$\alpha_i = W_i^2/W_i^{-1} \tag{A4}$$

It is clear that for the chemical system (5)

$$W_0^{-1} = W_N^2 = 0$$

Thus it may be reasonably assumed, without any detailed hypothesis on the transition rates, that α_i smoothly decreases from $\alpha_i \gg 1$ to 0 as i increases from 1 to N. More precisely, with the usual form of the transition rates[13,14]

$$\begin{aligned} W_i^{-1} &\propto i \\ W_i^2 &\propto (N-i)(N-i-1) \end{aligned} \tag{A5}$$

α_i is given by

$$\alpha_i \propto (N-i)(N-i-1)/i \tag{A6}$$

which indeed is a decreasing function of i.

As a consequence r_i^0 decreases from $r_1^0 \gg 1$ to $r_N^0 = 0$ when i varies from 1 to N, according to formula (A2). It is seen from (A1) and (A3) that if $r_i > r_i^0 + 1$ (respectively, $r_i < r_{i+1}^0$), then $r_{i+1} < r_{i+1}^0 < r_i$ (respectively, $r_{i+1} > r_{i+1}^0 > r_i$) and conversely. Thus a necessary and sufficient condition for r_i to be a decreasing function of i is that condition (43) hold for every i:

$$r_{i+1}^0 < r_i < r_i^0 \tag{A7}$$

These inequalities are verified for $i = 1$, since $r_1 = \alpha_i$. Indeed (A1) shows that

$$\alpha_i < r_i^0 < \alpha_i + 1 \qquad \text{for every } i \tag{A8}$$

and because of the properties of α_i,

$$\alpha_2 + 1 \ll \alpha_1$$

[with expression (A6) of α_i, $\alpha_2 \simeq \alpha_1/2 \gg 1$]. Now let us suppose that (A7) holds for one value of i: then it also holds for $i + 1$. Indeed $r_i > r_{i+1}^0$ implies $r_{i+1} < r_{i+1}^0$; $r_i < r_i^0$ implies

$$r_{i+1} > \alpha_{i+1}(1 + 1/r_i^0) = \alpha_{i+1} r_i^0/\alpha_i \tag{A9}$$

and it will be shown that

$$\alpha_{i+1} r_i^0/\alpha_i > r_{i+2}^0 \tag{A10}$$

which establishes the recurrence of property (A7). ■

Proof of (A10). Let the relation (A2) between α_i and r_i^0 be written

$$r_i^0 = \phi(\alpha_i)$$

and define α by

$$\alpha_{i+1} r_i^0/\alpha_i = \phi(\alpha) \tag{A11}$$

which gives

$$\alpha = \frac{(r_i^0)^2(\alpha_{i+1}/\alpha_i)^2}{1 + r_i^0\alpha_{i+1}/\alpha_i} = \frac{(1 + r_i^0)\alpha_{i+1}^2/\alpha_i}{1 + r_i^0\alpha_{i+1}/\alpha_i} \tag{A12}$$

Since $\phi(\alpha)$ increases with α, (A10) is equivalent to $\alpha > \alpha_{i+2}$, or

$$\frac{\alpha_i\alpha_{i+2}}{(\alpha_{i+1})^2} < \frac{1 + r_i^0}{1 + r_i^0\alpha_{i+1}/\alpha_i} \tag{A13}$$

This relation is obviously true for small values of i, since then $r_i^0 \gg 1$, and the right-hand side of (A13) (which is always greater than 1) is

$$\alpha_i/\alpha_{i+1} > (\alpha_i/\alpha_{i+1})(\alpha_{i+2}/\alpha_{i+1})$$

On the other hand if $i \simeq N$, it is seen from (A6) that $\log\alpha_i$ is a concave function of i (and this would be true for $W^2 \propto (N - i)^a$ and any positive value of a); thus

$$\alpha_i\alpha_{i+2}/(\alpha_{i+1})^2 < 1$$

and (A13) is true again.

Finally (A13) holds at the thermodynamic limit, when $x = i/\Omega$. Indeed, taking the logarithm of (A13) and writing $z = \log\alpha(x)$, one obtains

$$\frac{d^2z}{dx^2}\Omega^{-2} < -\log\left[1 + \frac{r^0(x)}{1 + r^0(x)}\frac{dz}{dx}\Omega^{-1}\right] \simeq \frac{r^0(x)}{1 + r^0(x)}\left|\frac{dz}{dx}\right|\Omega^{-1} \tag{A14}$$

which is verified for any finite value of x.

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