On a Linear Polymerisation Process

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Abstract. We consider a Markov fusion-disaggregation process of 'molecules' which are linear arrangements of 'atoms'. Molecules may fuse at the ending atoms by creating a bond in between at a given rate. Each bond joining two atoms in a molecule can break so that a molecule disaggregates into two shorter molecules at another rate. We give en explicit expression for the stationary distribution of the number of molecules and the conditional distribution of their lengths given their number. We show an asymptotic normality of the distribution of the number of molecules when the number of atoms in the system grows.

Keywords: Fusion-Disaggregation Process · Reversibility · Stationary Distribution · Markov Chain

1 Introduction

The model we consider in this article has origins in colloidal chemistry. Colloid is a mixture of molecules which stay evenly dispersed in the volume neither settling to the botton nor floating to the surface of the container. Although the geometry of molecules can be rather complex, some materials, like lacquers, have long chains of atoms in a linear molecules which combine to form a net when the lacquer sets. The molecule bonds may also break causing emergence of shorter molecules, this is the main mechanism of degradation of the material. The two competing mechanisms: fusion and disaggregation compete with each other and should be in balance in a stable system.

To mathematically model a colloid with linear molecules, we consider a system consisting of N elements called *atoms*. $n \geq 2$ atoms may form a *molecule* of size n (or an n-*molecule*) which is a linearly arranged set of atoms linked by *bonds*. We will also call a singular atom a 1-molecule.

Time evolution of the system is described by a homogeneous continuous time Markov chain (MC) determined by two competing processes: fusion (or clustering) and disaggregation (or severance). Two molecules close to each other may combine by creating a bond between terminal atoms. Ignoring spatial considerations, each n-molecule fuses to an r molecule at rate λ to form an $n+r$ molecule. In other words, each pair of molecules fuses at the rate $2\lambda > 0$ independently of their sizes. Each bond breaks with the rate 1, so that an n-molecule disaggregates to two shorter molecules at rate $n-1$.

It is clear that the longer molecule is, the more bonds it contains, the more is its rate of disaggregation. On the other hand, the more molecules are in the system, the more is the number of their terminal atoms which may fuse, so the two competing mechanisms should balance each other in the long run leading to a stationary distribution. Since the system is finite, the stationary distribution is also its asymptotic distribution which we aim to characterise.

The similar (and even more general) model has been considered by Frank Kelly who obtained a product form of the stationary distribution [3, Th.8.1]. Unfortunately, no details on how the expression was obtained is given there, the proof consists in just checking that the solution satisfies the so-called Detailed Balance Equations which is a necessary and sufficient condition for the reversibility of the corresponding Markov chain. In this short article, we fill this gap by explicitly describing a procedure which can be used to establish reversibility and complement Kelly's result with finding exact asymptotics of the stationary distribution when the number of atoms in the system grows.

In the next section we give the necessary definitions and describe the model we are working with. In Section 3 we find an explicit expression for the stationary distribution of the model and then study its asymptotic properties when the number of atoms grows in Section 4.

2 Markov Model and Reversibility

The states of the system can be described by a vector $\mathbf{n} = (n_1, n_2, \dots, n_N)$, $\sum_{k=1}^N kn_k = N$ with n_k being the number of k-molecules in the system. Let $X(t),\ t \geq 0$ denotes the corresponding MC in this state space and e_k stands for the k-th coordinate vector of $\mathbb{R}^N$. If τ is the time of the first change of the system state, then $X(\tau) - X(0)$ with positive probability may assume only the following values:

1. $v_{ij} = -e_i - e_j + e_{i+j},\ i \neq j$, when two unequal size i- and j- molecules fuse. The intensity of this jump is $2\lambda n_i n_j$.
2. $v_{ii} = -2e_i + e_{2i}$, when two i-molecules fuse. The intensity is then $\lambda n_i(n_i - 1)$.
3. $-v_{ij}, i \neq j$, when an $i+j$-molecule breaks into two uneven parts. There are two bonds in each $i+j$-molecule which breaking results in i- and j-molecules, so the intensity of this transition is $2n_{i+j}$.
4. $-v_{ii}$, when a $2i$-molecule breaks in the middle into two i-molecules, the intensity of this is n_{2i}.

Not all these transitions are possible from all the states: the ones which would lead to negative values of any coordinate are forbidden.

Obviously, any state of the system is attainable from any other state by a sequence of positive-intensity jumps: for instance, by disaggregating to singular atoms and then fusing to the other configuration. This means the associated MC is irreducible and hence geometrically ergodic, i.e. there exists a unique stationary distribution to which convergence of the distribution at a time t happens exponentially fast whatever is the starting state $X(0)$, see, e.g., [2].

An important property some MCs possess is time reversibility. Recall that a continuous time irreducible MC $X(t)$ with states $\{1, 2, 3, \dots\}$ and transition rates $q(i, j)$ is called *reversible* if $(X(t_1), \dots, X(t_n))$ has the same distribution as $(X(t_0 - t_1), \dots, X(t_0 - t_n))$ for all $t_0, t_1, \dots, t_n$. Then $X(t_0 - t)$ is a distributional copy of $X(t)$, hence the name.

A MC is reversible if and only if there is a probability distribution $(\pi(i))$ such that the following *Detailed Balance Equations* are satisfied for all pairs of states i, j:

$$\pi(i)q(i, j) = \pi(j)q(j, i), \tag{1}$$

see, e.g., [3, Th. 1.2]. Such $(\pi(i))$ is then the stationary distribution of the MC.

The Kolmogorov's criterion of reversibility states that MC is reversible if and only if for any cyclic sequence of states $i, i_1, i_2, \dots, i_n, i$, the product of the transition intensities on one and opposite directions coincide:

$$q(i, i_1)q(i_1, i_2) \dots q(i_n, i) = q(i, i_n)q(i_n, i_{n-1}) \dots q(i_1, i), \tag{2}$$

see, e.g., [3, Th. 1.8]. Both (1) and (2) allow to express the stationary distribution of a reversible MC in the form:

$$\pi(j) = \pi(i_0)\frac{q(i_0, i_1)q(i_i, i_2) \dots q(i_n, j)}{q(j, i_n)q(i_n, i_{n-1}) \dots q(i_1, i_0)} \tag{3}$$

for any chosen state i_0 and a sequence of states (a path) $i_0, i_1, \dots, i_n, j$ for which the product of the intensities above is non-zero. The value of $\pi(i_0)$ itself is then found from normalisation: $\sum_j \pi(j) = 1$.

3 Stationary Distribution

In this section we give a closed form expression for the stationary distribution of the MC modelling the molecular system described in the previous section. For this, we first establish that the MC is reversible. Property (2) is hard to check. Instead, we evaluate expressions (3) and see if the obtained sequence $(\pi(j))$ satisfies the detailed balance equations (1). Then the MC is reversible and such $(\pi(j))$ is its stationary distribution. This the way we prove the main result of this section.

Theorem 1. *The Markov chain describing evolution of the system of N atoms is reversible. Its stationary distribution satisfies, for $\mathbf{n}_0 = (N, 0, \dots, 0)$,*

$$\pi(\mathbf{n}) = \pi(\mathbf{n}_0)\lambda^{L(\mathbf{n})}\frac{N!}{\prod_{k\geq 1} n_k!}, \tag{4}$$

where $L(\mathbf{n}) = \sum_{k\geq 2}(k-1)n_k$ is the number of links in the state $\mathbf{n}$.

Proof. Take the state $\mathbf{n}_0 = (N, 0, \dots, 0)$, where all atoms are separated, to express the probability of all other states in the form of (3). We choose the following path to reach a state $\mathbf{n} = (n_1, \dots, n_N) \neq \mathbf{n}_0$ from $\mathbf{n}_0$. Let $m, m \geq 2$ be the length of the longest molecule in $\mathbf{n}$, i.e. all $n_{m+1} = \cdots = n_N = 0$. At first, the atoms join one by one to create an m-molecule, then, if $n_m > 1$, the second m-molecule, the third, etc., until all m-molecules are created. Then it comes the turn of $m-1$-molecules (or the next smaller size molecules if $n_{m-1} = 0$ until all the molecules of that size are created. Then, the next smaller size molecules, etc., until all k-molecules, $k \geq 2$, are created. $\mathbf{n}_1 = \mathbf{n}_0 - 2e_1 + e_2$. The next one is $\mathbf{n}_2 = \mathbf{n}_0 - e_1 - e_2 + e_3$ and so on. The state with one m-molecule is obtained by adding $-e_1 - e_{m-1} + e_m$ to the previous state. This creation process is exemplified on Figure 1. Each state can be represented uniquely as an array of segments representing molecules spanning the grid points $\{1, 2, \ldots, N\}$. The k-molecules are represented by disjoint segments $[j, j+m-1]$ arranged by their size, the longest on the left.

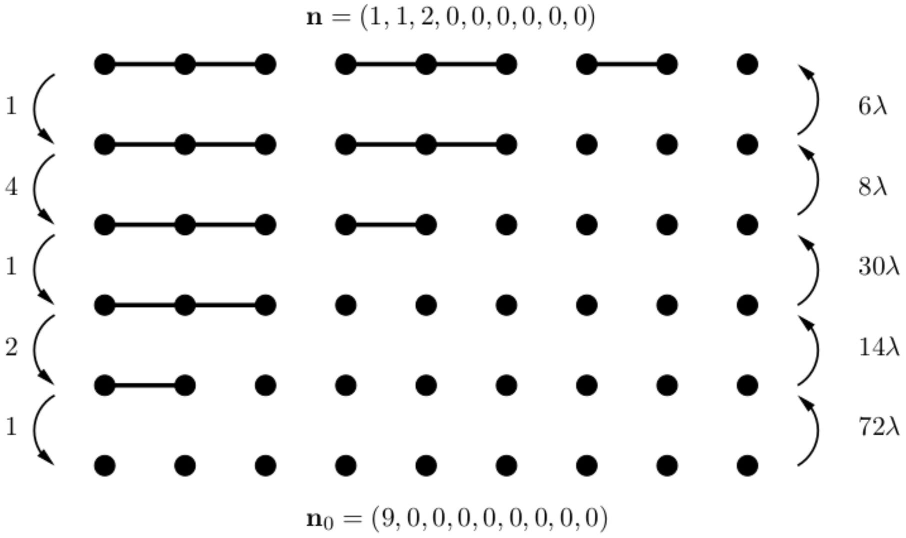

Fig. 1. The path between the starting and the terminal states in a system with 9 atoms. The molecules are represented by segments ordered by their size. The transition intensities are shown on the sides.

The rate at which the first link is established is $\lambda N(N-1)$ since there are $\binom{N}{2}$ pairs of atoms in configuration $(N, 0, \ldots, 0)$ and each pair joins at rate 2λ. The next atom joins the newly formed 2-molecule at the rate $2\lambda(N-2)$, the next one at the rate $2\lambda(N-3)$ and so on, the last atom to complete the first m-molecule joins at the rate $2\lambda(N-m+1)$. Now there are $N-m$ separate atoms in the system and one m-molecule. The product of the intensities of all the above steps is then

$$2^{m-2}\lambda^{m-1}N(N-1)\ldots(N-m+1) = 2^{m-2}\lambda^{m-1}\frac{N!}{(N-m)!}.$$

The intensities of construction of the next molecule just repeat the above expressions with N replaced by $N-m$. For instance, if $n_m > 1$, the product of intensities of all the steps until construction of the second m-molecule is given by

$$2^{m-2}\lambda^{m-1}\frac{N!}{(N-m)!}\times 2^{m-2}\lambda^{m-1}\frac{(N-m)!}{(N-2m)!}=2^{2(m-2)}\lambda^{2(m-1)}\frac{N!}{(N-2m)!}$$

After all n_m molecules are created, the product is

$$2^{(m-2)n_m}\lambda^{(m-1)n_m}\frac{N!}{(N-mn_m)!}.$$

Now there are $N-mn_m$ atoms in the system and the construction of $(m-1)$-molecules proceed similarly. Once all k-molecules for $k\geq 3$ are created, there are $N' = N-\sum_{k\geq 3} kn_k$ atoms. The first 2-molecule is created with intensity $\lambda N'(N'-1)$, the second – $\lambda(N'-2)(N'-3)$ etc. until the last n_2-nd one with intensity $\lambda(N'-2(n_2-1))(N'-2(n_2-1)-1)$. The product of intensities of the transitions involving creation of all 2-molecules is thus

$$\lambda^{n_2}\frac{N'!}{(N'-2n_2)!}=\lambda^{n_2}\frac{(N-\sum_{k\geq 3} kn_k)!}{(N-\sum_{k\geq 2} kn_k)!}=\lambda^{n_2}\frac{(N-\sum_{k\geq 3} kn_k)!}{n_1!}.$$

This gives the following product of the intensities of all the transitions from $\mathbf{n}_0$ to $\mathbf{n}$:

$$2^{\sum_{k\geq 3}(k-2)n_k}\lambda^{\sum_{k\geq 2}(k-1)n_k}\frac{N!}{n_1!}=2^{I(\mathbf{n})}\lambda^{L(\mathbf{n})}\frac{N!}{n_1!},\tag{5}$$

where $I(\mathbf{n})=\sum_{k\geq 3}(k-2)n_k$ is the number of inner atoms (i.e. connected to two other atoms in the molecules) and $L(\mathbf{n})=\sum_{k\geq 2}(k-1)n_k$ is the number of links.

The reversed moving along the same path consists, first, in one of n_2 2-molecules to break if $n_2\geq 1$. This happens at the rate n_2, because there are that many links in all 2-molecules. The next 2-molecule breaks at the rate n_2-1, etc. The last one breaks at the rate 1, so that the product of intensities of all these breaks is $n_2!$. If $n_3\geq 1$, the next step is a link of a 3-molecule to break, this happens at rate $2n_3$, and then the second link of the same, now 2-molecule, breaks at rate 1. Now the number of 3-molecules in the system is decreased by one, so if there are still 3-molecules, the next one disintegrates into single atoms at rate $2(n_3-1)$. Thus, the product of intensities of the steps from $\mathbf{n}$ to the state where all 2- and 3-molecules are disintegrated into atoms equals $n_2!\,2^{n_3}n_3!$. After all the molecules of the sizes smaller than k have disintegrated into single atoms, the first link in a k-molecule breaks at rate $(k-1)n_k$, the second link in the same molecule breaks at rate 2 (since there are two ends) and so on until the last one breaks at rate 1. So the first disintegrated k-molecule contributes $2^{k-2}n_k$ to the product of intensities. Similarly, the second contributes $2^{k-2}(n_k-1)$ and so on. Hence, the product of the intensities of all the steps from $\mathbf{n}$ back to $\mathbf{n}_0$ is given by

$$\prod_{k\geq 2}2^{n_k(k-2)}n_k!=2^{I(\mathbf{n})}\prod_{k\geq 2}n_k!\,.\tag{6}$$

This allows us to define the quantities

$$\pi(\mathbf{n}) = \pi(\mathbf{n}_0)\frac{2^{I(\mathbf{n})}\lambda^{L(\mathbf{n})}\frac{N!}{n_1!}}{2^{I(\mathbf{n})}\prod_{k\geq 2} n_k!} = \pi(\mathbf{n}_0)\lambda^{L(\mathbf{n})}\frac{N!}{\prod_k n_k!}. \tag{7}$$

The next step consists in verifying that such defined quantities satisfy the detailed balance equations (1) for the four possible type of jumps described at the beginning of Section 2.

Indeed, for the transition $\mathbf{n} \leftrightarrow \mathbf{n} + v_{ij}$, $i \neq j$, the detailed balance equation becomes

$$\pi(\mathbf{n})\, 2\lambda n_i n_j = \pi(\mathbf{n} + v_{ij})\, 2(n_{i+j} + 1).$$

Since $L(\mathbf{n} + v_{ij}) = L(\mathbf{n}) + 1$, this reduces to

$$\lambda^{L(\mathbf{n})}\frac{N!}{\prod_k n_k!} 2\lambda n_i n_j = \lambda^{L(\mathbf{n})+1}\frac{n_i n_j N!}{(n_{i+j}+1)\prod_k n_k!} 2(n_{i+j} + 1)$$

which is true. Similarly one can easily verify that for transition $\mathbf{n} \leftrightarrow \mathbf{n} + v_{ii}$, the detailed balance equation

$$\pi(\mathbf{n})\, \lambda n_i(n_i - 1) = \pi(\mathbf{n} + v_{ii})\, (n_{2i} + 1)$$

also holds. Thus by [3, Th. 1.2], the Markov chain is reversible and its stationary distribution is given by (4).

Corollary 1 *Denote by $M(\mathbf{n}) = \sum_k n_k$ the total number of molecules (including atoms) in the state $\mathbf{n}$. Under the stationary regime, the probability mass function (p.m.f.) of M is given by*

$$\pi(M(\mathbf{n}) = m) = Z^{-1}(\lambda, N)\, \frac{\lambda^{-m}}{m!}\binom{N-1}{m-1}, \quad m = 1, \ldots, N, \tag{8}$$

the normalising constant being

$$Z(\lambda, N) = \sum_{m=1}^{N} \frac{\lambda^{-m}}{m!}\binom{N-1}{m-1} = \frac{1}{\lambda N}\, L^1_{N-1}(-1/\lambda), \tag{9}$$

where $L^\alpha_n(x)$ is the generalised Laguerre polynomial of degree n, see, e.g., [4]. The distribution is unimodal with the mode at the point

$$m^* = \left\lceil \frac{-(1+\lambda) + \sqrt{(1+\lambda)^2 + 4\lambda N}}{2\lambda} \right\rceil \sim O(\sqrt{N/\lambda}) \quad \textit{as } N \to \infty. \tag{10}$$

Its Laplace transform is

$$\mathcal{L}_M(t) = \mathbf{E}\, e^{-tM} = \frac{Z(e^t\lambda, N)}{e^t Z(\lambda, N)} = \frac{e^{-t}L^1_{N-1}(-e^{-t}/\lambda))}{L^1_{N-1}(-1/\lambda)}, \tag{11}$$

the first two moments are given by

$$\mathbf{E}\, M = 1 + \frac{l_2}{\lambda l_1}; \qquad \mathbf{var}\, M = \frac{l_2}{\lambda l_1} + \frac{l_1 l_3 - l_2^2}{\lambda^2 l_1^2}, \tag{12}$$

where $l_k = L^k_{N-k}(-1/\lambda),\ k = 1, 2, 3.$

The p.m.f. of the number M of molecules under the stationary regime are shown on Figure 2.

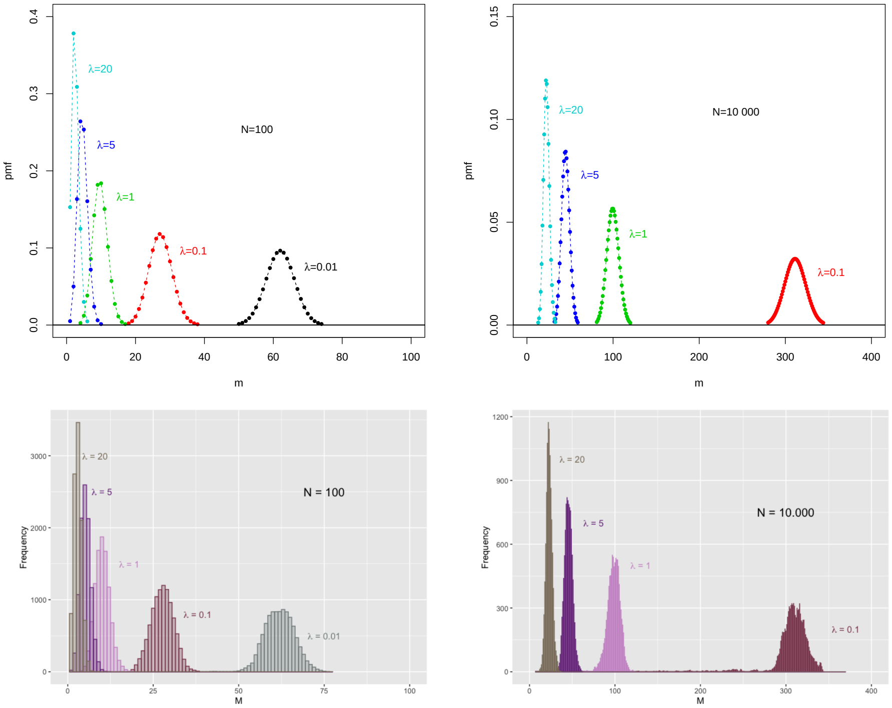

Fig. 2. Top prot: the p.m.f. of the number M of molecules under the stationary regime for the cases $N = 100$ and $N = 10\,000$. Bottom plot: the histograms of M in a computer simulation.

Proof. Taking into account that $\sum_k kn_k = N$, expression (4) gives that

$$\pi(M(\mathbf{n}) = m) = \pi(\mathbf{n}_0)\lambda^{N-m} \sum_{\substack{\mathbf{n}:\ \sum_k n_k = m \\ \sum_k kn_k = N}} \frac{N!}{\prod_k n_k!}. \tag{13}$$

Assume the system contains exactly m molecules. Number them all somehow from 1 to m and let $(l_1, \dots, l_m)$ be their sizes. If we represent each k-molecule by a closed segment of length $k-1$, then they can be put from left to right starting from the first to the last segment disjointly on $[1, N]$ so that each k-molecule spans exactly k integer points, like on Figure 1, but not ordered by their lengths, but by their number. Since $\sum_k k n_k = N$, all the integer points of $[1, N]$ are covered and there are exactly $m-1$ open segments of the form $(i, i+1)$ which separate the molecules. Thus all the sequences $(l_1, \dots, l_m)$ of the sizes of ordered molecules can be obtained by throwing away $m-1$ open unit segments out of total $N-1$ of $[1, N] \setminus \mathbb{N}$, so there are $\binom{N-1}{m-1}$ of these.

Alternatively, the sequences $(l_1, \dots, l_m)$, $l_i \geq 1$ satisfying $\sum_{i=1}^m l_i = N$ can be produced from $\mathbf{n} = (n_1, n_2, \dots)$ satisfying $\sum_k n_k = m$, $\sum_k k n_k = N$ by assigning n_1 1's, n_2 2's, etc. to places $1, \dots, m$, so that each such $\mathbf{n}$ generates the multinomial number $m!/\prod_k n_k!$ of such distinct sequences. Therefore,

$$\sum_{\substack{\mathbf{n}:\ \sum_k n_k = m \\ \sum_k k n_k = N}} \frac{m!}{\prod_k n_k!} = \binom{N-1}{m-1}$$

yielding

$$\pi(M(\mathbf{n}) = m) = \pi(\mathbf{n}_0) \lambda^{N-m} \frac{N!}{m!} \binom{N-1}{m-1}.$$

The normalising condition $\sum_{m=1}^N \pi(M(\mathbf{n}) = m) = 1$ allows to express

$$\pi(\mathbf{n}_0) = \left[\sum_{m=1}^{N} \lambda^{N-m} \frac{N!}{m!} \binom{N-1}{m-1} \right]^{-1} \tag{14}$$

that gives for $M(n)$ distribution (8) after cancelling out $N! \lambda^N$.

Notice that

$$\pi(M(\mathbf{n}) = m+1) = \frac{N-m}{\lambda m(m+1)} \pi(M(\mathbf{n}) = m).$$

The multiplicative coefficient given by the fraction is monotonely decreasing and becomes less than 1 for $m = m^*$ as in (10). Thus the distribution is unimodal.

The expression for the Laplace transform is immediate from (9) and hence the moments follow from its Taylor expansion at $t = 0$.

The obtained expression (14) for $\pi(\mathbf{n}_0)$ plugged into (4) together with the distribution (8) leads to the main result of this section:

Theorem 2. *The stationary distribution* π *is given by*

$$\pi(\mathbf{n}) = Z^{-1}(\lambda, N) \frac{\lambda^{-M(\mathbf{n})}}{\prod_k n_k!} \tag{15}$$

with $Z(\lambda, N)$ as in (9). The conditional distribution of the molecule lengths given their total number is

$$\pi(\mathbf{n} \bigm| M(\mathbf{n}) = m) = \frac{m!}{\binom{N-1}{m-1} \prod_k n_k!} = \frac{\binom{m}{n_1,\dots,n_N}}{\binom{N-1}{m-1}} \tag{16}$$

which is the symmetric Multinomial distribution $\mathsf{Mult}(m; N^{-1}, \dots, N^{-1})$ conditioned on the set $\{\mathbf{n} : \sum_k k n_k = N\}$.

4 Asymptotic Distribution for Molecule Number

In this section we consider the limit distributions for the system when the number N of molecules grow. In the previous section we expressed the Laplace transform of the stationary distribution of the total number $M = M_N$ of molecules in the system with N atoms in terms of the generalised Laguerre polynomials. In [1], the following asymptotics in n for fixed parameters α and z was obtained:

$$L_n^\alpha(-z) = \frac{e^{-z/2}}{2\sqrt{\pi}} \frac{e^{2\sqrt{(n+1)z}}}{z^{1/4+\alpha/2}(n+1)^{1/4-\alpha/2}} \left(1 + O\Big(\frac{1}{\sqrt{n+1}}\Big)\right). \tag{17}$$

Substituting this expression into (11) with $n = N - 1$ and $z = 1/\lambda$, we get

$$\mathcal{L}_{M_N}(t) = \exp\Big\{ -\frac{t}{4} - \frac{z}{2}\big(e^{-t} - 1\big) + 2\sqrt{Nz}\big(e^{-t/2} - 1\big)\Big\}\Big(1 + O\Big(\frac{1}{\sqrt{N}}\Big)\Big). \tag{18}$$

Introduce $a_N = \sqrt{Nz} = \sqrt{N/\lambda}$ and denote

$$\mu_N = \frac{M_N - a_N}{\sqrt{a_N/2}}.$$

For the Laplace transform of μ_N we have the identity

$$\mathcal{L}_{\mu_N}(t) = e^{t\sqrt{2a_N}} \mathcal{L}_{M_N}\Big(\frac{t}{\sqrt{a_N/2}}\Big)$$

and it is straightforward to check using (18) that $\lim_{N\to\infty} \mathcal{L}_{\mu_N}(t) = t^2/2$. Thus we have proved the following Central limit theorem:

Theorem 3. *The stationary distributions of the number M_N of molecules in the system with N atoms when properly centred and scaled weakly converge to the standard Normal law:*

$$\frac{M_N - a_N}{\sqrt{a_N/2}} \Longrightarrow \mathcal{N}(0,1), \quad \textit{where } a_N = \sqrt{N/\lambda}. \tag{19}$$

Note that from (12) and (17), both $\mathbf{E}\, M_N$ and $\mathbf{var}\, M_n$ under the stationary regime have order of $\sqrt{N/\lambda}$, so the centering and scaling in (19) is as could be expected.

Using the standard methods, a local limit theorem can also be established which we give without a proof, see illustrative Figure 3:

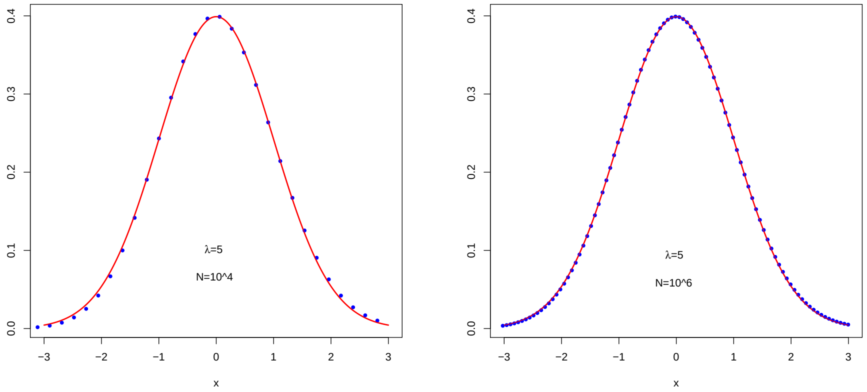

Fig. 3. The scaled p.m.f. (the dots) of the number M of molecules under the stationary regime for the case $N = 10^4$ (left plot) and $N = 10^6$ (right plot) and the density of the standard normal distribution (the curve).

Theorem 4. *The value* $\pi(M_N = m)$ *for* $m = O(\sqrt{N})$ *is approximated when* $N \to \infty$ *by the density of the Normal* $\mathcal{N}(a_N, a_N/2)$ *distribution, where* $a_N = \sqrt{N/\lambda}$*, i.e.*

$$\pi(M_N = m) = \frac{1}{\sqrt{\pi a_N}} \exp\left\{ - \frac{(m - a_N)^2}{a_N} \right\} + o(N^{-1/4}). \tag{20}$$

5 Discussion

The model we considered in this paper allows for generalisation in various directions. First of all, the linear structure of the molecules we impose can be dropped, instead, *terminal* atoms to where the molecules can attach (the molecule end-points as now) may be defined, possibly of varying accommodation capacities, see [3, Sec. 8]. It is interesting if explicit expressions and the asymptotic behaviour can also be established for the stationary distribution in these cases.

Another direction is to allow some molecules to disappear and to come into the system. This could model the diffusion, its parameters, for instance, may depend on the molecule size. One may expect a Poisson distribution to appear in the stationary regime in such an open system, as in [3, Th. 8.2].

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