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Large fluctuations of radiation in stochastically activated two-level systems

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Abstract

We study large fluctuations of emitted radiation in the system of N non-interacting two-level atoms. Two methods are used to calculate the probability of large fluctuations and the time dependence of excitation and emission. The first method is based on the large deviation principle for Markov processes. The second one uses an analogue of the quantum formalism for classical probability problems.

Particularly we prove that in a large fluctuation limit approximately half of the atoms are excited. This fact is independent on the fraction of the excited atoms in equilibrium.

Keywords: continuous-time Markov processes, large deviations, reversibility, stationary probabilities, product-formula

1. Motivations

In recent years there has been considerable interest in studying optimal realizations of large deviations in stochastic interacting particle systems, see e.g. [1, 3, 4] for optimal profiles in current large deviations of the asymmetric simple exclusion process or [10, 11] for condensation in the zero-range process. In addition to the mathematical interest in understanding how large deviations are typically realized (when they occur), motivation comes also from the fact that such large deviations are studied experimentally in order to test the validity of fluctuation

theorems such as the Gallavotti–Cohen theorem [9], the Lebowitz–Spohn results [14] or the Jarzynski relation [12]. Generally, fluctuation theorems predict the ratio of probabilities of a positive large deviation of some observable to a negative large deviation of the same absolute magnitude [5, 14, 21]. Particularly simple and therefore transparent systems where such relations can be probed experimentally are two-level systems where each microscopic entity can be either in a ground state or in an excited state. As an example we mention the optical excitation of an ensemble of single-defect centres in diamond. Measurements of the dissipated work [19] and entropy [22] agree with the expected integral fluctuation relations. Fluctuation relations for driven two-level systems have been investigated recently also in the context of biological motors [6] and quantum two-level systems [15].

Here we address the question of optimal realizations of a large deviation in an ensemble of two-level systems. We consider a scenario where every two-level system in the ensemble of (independent) two-level systems can be excited by an external source of energy and later relaxes to its ground state by the emission of radiation. Conditioning on a large deviation of the emitted radiation in a time interval $[0, T]$ we ask how the number of excited states $\Xi_N(t)$ and also how the accumulative radiation $\Upsilon_N(t)$ will typically behave during this time interval assuming that the number N of elements of the two-level system is large. In particular, we wish to answer the question whether such a large deviation is typically realized by the typical behaviour for most of the time and only some strong bursts of radiation, or by non-typical behaviour throughout $[0, T]$, or by both, i.e. by non-typical behaviour together with bursts.

For definiteness we refer to microscopic two-level systems as atoms. To keep the model simple we study the case where both excitation and emission occur after exponentially distributed random times with parameters λ and μ respectively. It will transpire that the behaviour of the ensemble of two-level systems is non-trivial: the optimal realization of a large deviation involves non-typical radiation intensity throughout $[0, T]$. If T is large then at intermediate times the radiation activity is approximately constant (and different from its typical value), and one has moderate bursts or suppression at the beginning or the end (or both) of the time-interval $[0, T]$, depending on the initial state and on the magnitude of the large deviation that is under consideration. The 'moderate bursts' are defined as an expected increase in the radiation intensity $\dot{\Upsilon}_N(t)$ during a short (exponentially bounded) interval of time. The probabilistic interpretation of this behaviour is that in order to generate a large deviation from typical behaviour in a long time interval T it is less costly to have a relatively small, but lasting, deviation from non-typical behaviour than to have almost permanent typical behaviour, punctuated by occasional strong deviations (strong bursts of radiation). The (moderate) bursts at initial and/or final times realize the transition from the initial state (and/or to the final state) to the intermediate mildly nontypical behaviour.

We address this problem both from a macroscopic Hamiltonian perspective and from a microscopic approach employing generating function techniques. The latter indicates that qualitatively similar behaviour is expected for any number of excited states per atom.

2. Macroscopic approach. Large deviations

In this section fluctuations of an ensemble of two-level atoms are described by means of the large deviation principle. The goal is to study a rare event where a very large emission is happening during time interval $[0, T]$. More exactly, we find a mean dynamics of the number of emissions and the number of excited atoms during $[0, T]$ conditioned on the rare event mentioned above.

Consider an ensemble (a sample) of N atoms where each atom can be either in a ground state or in an excited state. The sample is under a radiation by steady flux of photons. Each

atom in the ground state can absorb a photon transiting to the exited state. An atom in the excited state can emit a photon and consequently undergoes a transition to the ground state. There are no interactions between the atoms. Therefore all atom transformations are independent of each other. Given the parameters of mean times that an atom spends in the ground and the excited states it is easy to find the mean dynamics of the number of excited atoms and of the cumulative number of the photon emissions on $[0, T]$.

We show that the process path given the large emission on $[0, T]$ is close to a linear function. Moreover, under the large emission the conditional input and output flows are close to each other in the interior of $[0, T]$. This fact holds for any stationary regime of both flows.

We describe this dynamics by a two-dimensional Markov process. For any $t \in [0, T]$, the components of the process describe the number of the excited atoms at moment t and the cumulative number of emission events during $[0, t]$. In terms of the Markov process, our goal is to describe the shape of paths of the Markov process given the large cumulative number of emission events on $[0, T]$.

A tool we used for the analysis is the large deviation theory, which studies the probabilities of rare events, and also describes how these rare events evolve. The main tool of the large deviation theory is a theorem called the large deviation principle. There exists many versions of the large deviation principle for different stochastic models. Here we apply the large deviation principle to Markov processes following [8].

2.1. The model

We formalize our model as follows. Consider N independent random variables (atoms in what follows) that take two values 0 and 1. We say that the atom is in the ground state if the corresponding random variable is 0, otherwise the atom is excited. We study time dynamics of every atom. As time passes, any atom randomly changes its state. The stochastic dynamics of a single atom is a Markov process on the state space $\{0, 1\}$. The transition of the atom from the excited state to the ground state $1 \to 0$ is called an *emission*. The opposite transition is called an *excitation*.

A stochastic dynamics of the ensemble $\Sigma = (\sigma_1, ..., \sigma_N)$ of N two-level atoms where $\sigma_i \in \{0, 1\}$ is described by a two dimensional Markov process $\Gamma_N(t) = \big(\Xi_N(t), \Upsilon_N(t)\big)$, where $\Xi_N(t)$ is the number of excited atoms at time t and $\Upsilon_N(t)$ is the cumulative number of emitted photons on the interval $[0, t]$. The components $\Xi_N(t)$ and $\Upsilon_N(t)$ are strongly correlated.

2.1.1. Excitation component. The process $\Xi_N(t)$ takes its values in the set $\mathcal{N} = \{0, 1, 2, ..., N\}$. A random event $(\Xi_N(t) = n)$, $n \in \mathcal{N}$ means that the number of excited atoms is equal to n. Then $N - n$ is the number of the atoms in their ground states. The process $\Xi_N(t)$ is a jump Markov process. Every jump is either $+1$ or -1, either enlarging or reducing the number of the excited atoms by one.

An operator semigroup $(\mathbf{P}_t, \ t \in \mathbb{R}_+)$ drives the dynamics of $\Xi_N(t)$ with the initial value $\Xi_N(0) = x \in \mathcal{N}$. Namely, for any $G \in \mathcal{F}$

$$\mathbf{P}_t G(x) = \mathsf{E}_x G(\Xi_N(t)), \tag{2.1}$$

i.e. the operators $\mathbf{P}_t$ act on the space $\mathcal{F}$ of functions $\mathcal{N} \to \mathbb{R}$. The semigroup can be represented by an infinitesimal operator $\mathbf{L}$ as

$$\mathbf{P}_t G = \mathrm{e}^{t\mathbf{L}} G \tag{2.2}$$

for $G \in \mathcal{F}$. The infinitesimal operator $\mathbf{L}$ of $\Xi_N(t)$ is determined by two parameters $\lambda > 0$ and $\mu > 0$ as follows:

$$\mathbf{L}G(n) = \lambda(N-n)\left[G(n+1) - G(n)\right] + \mu n\left[G(n-1) - G(n)\right]. \qquad (2.3)$$

The constants λ and μ are intensities of the jumps $+1$ and -1 respectively.

The process $\Xi_N(t)$ is ergodic and reversible with the stationary distribution

$$\pi(n) := \Pr(\Xi_N(t) = n) = \frac{\binom{N}{n}\gamma^n}{(1+\gamma)^N}, \qquad (2.4)$$

where $\gamma = \lambda/\mu$. The distribution π can be found from the detailed balance equations

$$\lambda(N-n)\pi(n) = \mu(n+1)\pi(n+1), \text{ for } n < N. \qquad (2.5)$$

2.1.2. Emission component. The second component $\Upsilon_N(t)$ describes the number of emission events and is equal to the number of negative jumps of the process Ξ_N on the time interval $[0, t]$. Namely, let $k(t) = k_+(t) + k_-(t)$ be the total number of jumps of Ξ_N up to the moment $t \in [0, T]$, where $k_+(t)$ and $k_-(t)$ are the numbers of positive and negative jumps, respectively. Then we define

$$\Upsilon_N(t) = -k_-(t).$$

It is assumed that $k_+(0) = k_-(0) = 0$. Note that $\Xi_N(t) = \Xi_N(0) + k_+(t) - k_-(t)$.

2.1.3. The process. The two-dimensional process

$$\Gamma_N(t) = (\Xi_N(t), \Upsilon_N(t)) \qquad (2.6)$$

is a Markov process with dependent components taking their values in $\mathcal{N} \times \mathbb{Z}_-$, where $\mathbb{Z}_- = \{\dots -2, -1, 0\}$. The process $\Gamma_N(t)$ is a non-equilibrium jump process. Observe that the process has only two types of jumps $\{(1, 0), (-1, -1)\}$. The infinitesimal operator of $\Gamma_N(t)$ is

$$\mathbf{L}_\Gamma G(n, m) = \lambda(N-n)[G(n+1, m) - G(n, m)] + \mu n[G(n-1, m-1) - G(n, m)],$$
$$(2.7)$$

where $n \in \mathcal{N}, m \in \mathbb{Z}_-$, and λ, μ are positive parameters introduced in section 2.1.1, $G(n, m)$ belongs to the function space $\mathcal{F}_\Gamma$ of functions $\mathcal{N} \times \mathbb{Z}_- \to \mathbb{R}$.

2.2. Large deviations

The goal of our studies is to find the probability of a very large emission and a large deviation path of the process which produces very large emission during the interval $[0, T]$. It is assumed that the number N of the atoms is large and grows to infinity. The theory of large deviations allows one to solve both mentioned tasks: to find the asymptotics of the large emission probability as $N \to \infty$ as well as the path of the dynamics that realizes the large deviation on the interval $[0, T]$. Solving these problems by the method of large deviations we follow constructions and results in [8] (see also appendix in [7]).

In order to study the asymptotics in N it is convenient to consider the scaled process

$$\gamma_N(t) = \left(\xi_N(t) = \frac{1}{N}\Xi_N(t), \zeta_N(t) = \frac{1}{N}\Upsilon_N(t)\right) \qquad (2.8)$$

instead of (2.6). The scaled process $\gamma_N = (\xi_N, \zeta_N)$ takes values in $\mathbf{D}_N = \left(\frac{1}{N}\mathcal{N} \times \frac{1}{N}\mathbb{Z}_-\right)$. The process γ_N is a jump process with two types of jumps: $\left(\frac{1}{N}, 0\right)$ and $\left(-\frac{1}{N}, -\frac{1}{N}\right)$ with intensities λ and μ respectively. Let $(x, y) \in \mathbf{D}_N$ and $\alpha = \frac{1}{N}$. Then the infinitesimal operator of γ_N is

$$\mathbf{L}_{\gamma_N} G\,(x_N, y_N) = N\lambda(1 - x_N)[G(x_N + \alpha, y_N) - G(x_N, y_N)]$$
$$+ N\mu x_N[G(x_N - \alpha, y_N - \alpha) - G(x_N, y_N)].$$

Since $\mathbf{D}_N \subset \mathbf{D} = [0, 1] \times \mathbb{R}_-$ for every N, we will assume that the processes γ_N take their values in $\mathbf{D}$. Let $\mathcal{F}_2$ be the space of smooth functions $\mathbf{D} \to \mathbb{R}$. Assume that the sequence $(x_N, y_N) \in \mathbf{D}_N$ converges to some $(x, y) \in \mathbf{D}$, as $N \to \infty$. Then

$$\lim_{N \to \infty} \mathbf{L}_{\gamma_N} G(x_N, y_N) = \lambda(1 - x)\frac{\partial}{\partial x}G(x, y) - \mu x\left(\frac{\partial}{\partial x}G(x, y) + \frac{\partial}{\partial y}G(x, y)\right). \tag{2.9}$$

The Lagrangian plays a key role in the large deviations of the Markov processes γ_N

$$\mathcal{L}(f_1, f_2)(t) = \sup_{\varkappa_1(t), \varkappa_2(t)} \left\{\dot{f}_1(t)\varkappa_1(t) + \dot{f}_2(t)\varkappa_2(t) - H(f_1(t), f_2(t), \varkappa_1(t), \varkappa_2(t))\right\}, \tag{2.10}$$

which is the Legendre transform of the corresponding Hamiltonian

$$H(f_1, f_2, \varkappa_1, \varkappa_2) = \lambda(1 - f_1)[e^{\varkappa_1} - 1] + \mu f_1[e^{-\varkappa_1 - \varkappa_2} - 1]. \tag{2.11}$$

Thus

$$\begin{aligned}
\dot{f}_1 &= \lambda(1 - f_1)\exp\{\varkappa_1\} - \mu f_1\exp\{-(\varkappa_1 + \varkappa_2)\}, \\
\dot{f}_2 &= -\mu f_1\exp\{-(\varkappa_1 + \varkappa_2)\}.
\end{aligned} \tag{2.12}$$

Here f_1 belongs to the set $C_1[0, T]$ of differentiable functions on $[0, T]$ with values from interval $[0, 1]$, and $f_2 \in C_-[0, T]$ the set of differentiable functions on $[0, T]$ taking values in $\mathbb{R}_-$. The functions $\varkappa_1, \varkappa_2 \in C[0, T]$, where $C[0, T]$ is the space of differentiable functions on $[0, T]$ taking values in $\mathbb{R}$. The functions $\varkappa_1, \varkappa_2$ are called the *momenta* of the dynamics, and the pair (f_1, f_2) describes the dynamics of the process $\gamma_N(t)$ at large N: the probability that a trajectory of the process γ_N is close to the path $(f_1(t), f_2(t))$ is asymptotically represented by Lagrangian (2.10)

$$\mathbb{P}\big((\gamma_N(t))_{t \in [0, T]} \approx (f_1(t), f_2(t))_{t \in [0, T]}\big) \approx \exp\left(-N\int_0^T \mathcal{L}(f_1, f_2)(t)\mathrm{d}t\right).$$

To derive (2.11), we follow the scheme in [8]. First, we have to construct a non-linear Hamiltonian

$$\begin{aligned}
(\mathcal{H}_N G)(x_N, y_N) &:= \frac{1}{N}\exp\{-NG(x_N, y_N)\} \times \mathbf{L}_{\gamma_N}\exp\{NG\}(x_N, y_N) \\
&= \lambda(1 - x_N)\left[\exp\left\{N\left(G\left(x_N + \frac{1}{N}, y_N\right) - G(x_N, y_N)\right)\right\} - 1\right] \\
&\quad + \mu x_N\left[\exp\left\{N\left(G\left(x_N - \frac{1}{N}, y_N - \frac{1}{N}\right) - G(x_N, y_N)\right)\right\} - 1\right].
\end{aligned}$$

And, second, to find its limit as $N \to \infty$:

$$\lim_N(\mathcal{H}_N G)(x_N, y_N)$$
$$= \lambda(1 - x)\left[\exp\{\frac{\partial}{\partial x}G(x, y)\} - 1\right] + \mu x\left[\exp\{-\frac{\partial}{\partial x}G(x, y) - \frac{\partial}{\partial y}G(x, y)\} - 1\right]. \tag{2.13}$$

The right-hand side of (2.13) is the Hamiltonian (2.11) after the following changes of variables:

$$f_1 = x, f_2 = y, \varkappa_1 = \frac{\partial}{\partial x} G(x, y), \varkappa_2 = \frac{\partial}{\partial y} G(x, y).$$

We consider the problem of finding the optimal paths of large radiations. Namely, we consider the following boundary conditions:

$$\begin{aligned}
&f_1(0) = d \in [0, 1], \\
&f_2(0) = 0, f_2(T) = -B, \text{ where } B > 0,
\end{aligned} \tag{2.14}$$

for the Hamiltonian system corresponding to H in (2.11)

$$\begin{cases}
\dot{f_1} &= \lambda(1 - f_1) \exp\{\varkappa_1\} - \mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}, \\
\dot{f_2} &= -\mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}, \\
\dot{\varkappa_1} &= \lambda \exp\{\varkappa_1\} - \mu \exp\{-(\varkappa_1 + \varkappa_2)\} - \lambda + \mu, \\
\dot{\varkappa_2} &= 0.
\end{cases} \tag{2.15}$$

We study the system for large B.

The solution of (2.15) with the boundary conditions (2.14) presents the mean dynamics f_1 and f_2 of the occupation and radiation processes. We add the boundary condition $\varkappa_1(T) = 0$ because the value of $f_1(T)$ is free.

Remark 2.1 (On the large deviations principle). The book [8] proposes a program how to prove the large deviation principle for stochastic processes (in particular, Markov processes) in consideration. This program requires special considerations, depending on the studied process. For example, see [13], where the large deviation principle is studied for a model that is close to our model in some respect.

We, however, consider the Markov process $\Gamma_N(t)$ which is a scaled sum of N independent Markov processes. Each one of these Markov processes is concentrated on piece-wise constant paths on $[0, T]$. We can consider the Skorohod space of piece-wise continuous paths on $[0, T]$ (see [2], section 12). For our N independent paths we can use the Sanov theorem with subsequent application of the contraction principle (see [18]). This combination of the Sanov theorem and the contraction principle gives the rate function (see (2.17)) for our process Γ.

2.3. Results

Assume that the total radiation during the time interval $[0, T]$ is $\lfloor BN \rfloor$, where $\lfloor \cdot \rfloor$ is integer part, i.e. $\Upsilon_N(T) = -\lfloor BN \rfloor$, $B > 0$, with initial value $\Upsilon_N(0) = 0$. The initial number of excited atoms is $\Xi_N(0) = \lfloor dN \rfloor \in [0, N]$. For example, we can choose $d = \frac{\lambda}{\lambda + \mu}$. Then the large deviation principle yields

$$\lim_{N \to \infty} \frac{1}{N} \ln \Pr\left(\xi(0) = \frac{\lfloor Nd \rfloor}{N}, \ \zeta(0) = 0, \zeta(T) = -\frac{\lfloor NB \rfloor}{N} \right)$$
$$= -\inf_{f_1, f_2} \{ I(f_1, f_2) : f_1(0) = d, f_2(0) = 0, f_2(T) = -B \}, \tag{2.16}$$

where the rate function is, see (2.10) and (2.11),

$$\begin{aligned}
I(f_1, f_2) &= \int_0^T \mathcal{L}(f_1, f_2) \mathrm{d}t \\
&= \int_0^T \sup_{\varkappa_1, \varkappa_2} \left(\varkappa_1 \dot{f_1} + \varkappa_2 \dot{f_2} - \lambda(1 - f_1)[e^{\varkappa_1} - 1] - \mu f_1 [e^{-\varkappa_1 - \varkappa_2} - 1] \right) \mathrm{d}t.
\end{aligned}$$

Remark 2.2.   The rate function is convex therefore the infimum in (2.16) is achieved and unique. Indeed, as the functional $I(f_1, f_2)$ is the convex combination of suprema of affine functionals it is convex. So its critical point is its minimum. This minimum is unique because the corresponding boundary value problem for Euler system has a unique solution as follows from calculations.

We rewrite the rate function in an alternative way.

$$I(f_1, f_2) = \int_0^T \sup_{\varkappa_1(t), \varkappa_2(t)} \left(\varkappa_1(t)\dot{f}_1(t) + \varkappa_2(t)\dot{f}_2(t) - \rho(f_1)[\varphi(\varkappa_1(t), \varkappa_2(t)) - 1] \right) \mathrm{d}t, \qquad (2.17)$$

where $\rho(f_1) = \lambda(1 - f_1) + \mu f_1$, and

$$\varphi(\varkappa_1, \varkappa_2) = \frac{\lambda(1 - f_1)}{\rho(f_1)} \exp\{\varkappa_1\} + \frac{\mu f_1}{\rho(f_1)} \exp\{-(\varkappa_1 + \varkappa_2)\}. \qquad (2.18)$$

The supremum over $\varkappa_1(t), \varkappa_2(t)$ in (2.17) can be found from the equations

$$\begin{aligned} \dot{f}_1 &= \lambda(1 - f_1) \exp\{\varkappa_1\} - \mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}, \\ \dot{f}_2 &= -\mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}. \end{aligned} \qquad (2.19)$$

The infimum over f_1, f_2 can be found from Euler equations

$$\begin{aligned} \dot{\varkappa}_1 &= \lambda \exp\{\varkappa_1\} - \mu \exp\{-(\varkappa_1 + \varkappa_2)\} - \lambda + \mu, \\ \dot{\varkappa}_2 &= 0. \end{aligned} \qquad (2.20)$$

In other words, we have to solve the Hamiltonian system of equation (2.15)

$$\begin{cases} \dot{f}_1 &= \lambda(1 - f_1) \exp\{\varkappa_1\} - \mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}, \\ \dot{f}_2 &= -\mu f_1 \exp\{-(\varkappa_1 + \varkappa_2)\}, \\ \dot{\varkappa}_1 &= \lambda \exp\{\varkappa_1\} - \mu \exp\{-(\varkappa_1 + \varkappa_2)\} - \lambda + \mu, \\ \dot{\varkappa}_2 &= 0. \end{cases}$$

under boundary conditions $f_1(0) = d$, $f_2(0) = 0$, $f_2(T) = -B$ and $\varkappa_1(T) = 0$ with the Hamiltonian (2.11). We have derived the system of equations (2.15) with boundary conditions (2.14) as a solution of optimization problem. The solution of the boundary problem is unique and gives an extremal path for the radiation problem.

2.3.1. Solutions of (2.15).   Two last equations of (2.15) do not depend on $f_i, \dot{f}_i$, $i = 1, 2$. These equations can be solved as follows. The function $\varkappa_2$ does not depend on t. Having $\varkappa_2$ fixed we look for $\varkappa_1$ by means of the change of variable $y(t) = \mathrm{e}^{\varkappa_1(t)}$. Then the third equation of (2.15) is

$$\dot{y} = \lambda y^2 - (\lambda - \mu)y - \mu \mathrm{e}^{-\varkappa_2}. \qquad (2.21)$$

Using notations

$$\begin{aligned} \gamma &= \tfrac{\mu}{\lambda}, \\ a &= 1 - \gamma, \\ b &= \gamma \mathrm{e}^{-\varkappa_2} \end{aligned} \qquad (2.22)$$

we reduce (2.21) to the form

$$\dot{y} = \lambda(y^2 - ay - b). \qquad (2.23)$$

The value of a is upper bounded, $a \leqslant 1$, since $\gamma \geqslant 0$, however a may be negative and may have a large absolute value. We represent the expression in the parentheses of (2.23) as

$$y^2 - ay - b = (y - r_1)(y - r_2), \tag{2.24}$$

where

$$r_{1,2} = \frac{a}{2} \mp \sqrt{\left(\frac{a}{2}\right)^2 + b}. \tag{2.25}$$

Since $b > 0$, the roots r_1 and r_2 have different signs: $r_1 < 0 < r_2$. The equation (2.23) can be represented as

$$\frac{\dot{y}}{y - r_2} - \frac{\dot{y}}{y - r_1} = \lambda(r_2 - r_1). \tag{2.26}$$

The solution of (2.24) is

$$y(t) = e^{\varkappa_1(t)} = \frac{r_2 - r_1 C_1 \exp\{t\lambda(r_2 - r_1)\}}{1 - C_1 \exp\{t\lambda(r_2 - r_1)\}}, \tag{2.27}$$

where C_1 is a constant within the interval $\left(\frac{r_2}{r_1}, 1\right)$. The value of C_1 cannot be out of this interval because $0 < y(t) < \infty$. Remark that $\frac{r_2}{r_1} < 0$.

Note that $\varkappa_1(T) = 0$ since there are no constraints on the value of $f_1(T)$. Then

$$C_1 = \frac{r_2 - 1}{r_1 - 1} \exp\{-T\lambda(r_2 - r_1)\} \tag{2.28}$$

and

$$e^{\varkappa_1(t)} = \frac{r_2 + r_1 \frac{r_2 - 1}{1 - r_1} \exp\{(t - T)\lambda(r_2 - r_1)\}}{1 + \frac{r_2 - 1}{1 - r_1} \exp\{(t - T)\lambda(r_2 - r_1)\}}. \tag{2.29}$$

The energy conservation law implies (see (2.11)) that *there exists a constant K such that, for any $t \in T$,*

$$H(f_1, f_2, \varkappa_1, \varkappa_2) = K.$$

Thus the path f_1 corresponding to the density of excited atoms is

$$f_1(t) = \frac{\lambda(e^{\varkappa_1} - 1) - K}{\lambda(e^{\varkappa_1} - 1) + \mu(1 - e^{-\varkappa_1 - \varkappa_2})}. \tag{2.30}$$

The constant K is sought from the initial condition $f_1(0) = d$. That is,

$$K = \lambda(e^{\varkappa_1(0)} - 1)(1 - f_1(0)) - f_1(0)\mu(1 - e^{-\varkappa_1(0) - \varkappa_2}). \tag{2.31}$$

Next we have to evaluate the emission path (see (2.15))

$$f_2(t) = -\mu e^{-\varkappa_2} \int_0^t f_1(s) e^{-\varkappa_1(s)} ds, \quad t \in [0, T]. \tag{2.32}$$

To this end we evaluate the integral

$$J(t) = \int_0^t \frac{\lambda(e^{\varkappa_1(s)} - 1) + K}{\lambda(e^{\varkappa_1(s)} - 1) + \mu(1 - e^{-\varkappa_1(s)} e^{-\varkappa_2})} e^{-\varkappa_1(s)} ds. \tag{2.33}$$

The denominator of this integral multiplied by $e^{\varkappa_1(s)}$ can be written as

$$\lambda e^{2\varkappa_1(s)} - (\lambda - \mu)e^{\varkappa_1(s)} - \mu e^{-\varkappa_2} = \lambda(e^{\varkappa_1(s)} - r_1)(e^{\varkappa_1(s)} - r_2),$$

see (2.23)–(2.25). Hence

$$
\begin{aligned}
J(t) &= \int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_2} + \int_0^t \frac{\lambda(r_1 - 1) + K}{\lambda(e^{\varkappa_1(s)} - r_1)(e^{\varkappa_1(s)} - r_2)}\mathrm{d}s \\
&= \int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_2} + \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}\left(\int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_2} - \int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_1}\right) \\
&= \left(1 + \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}\right)\int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_2} - \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}\int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_1}.
\end{aligned}
$$

Since

$$e^{\varkappa_1(s)} - r_1 = \frac{r_2 + r_1 \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}} - r_1 = \frac{r_2 - r_1}{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}$$

then

$$
\begin{aligned}
J_1(t) &= \int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_1} = \int_0^t \frac{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}{r_2 - r_1}\mathrm{d}s \\
&= \frac{t}{r_2 - r_1} + \frac{r_2 - 1}{1 - r_1} \cdot \frac{e^{-\lambda(r_2 - r_1)T}}{\lambda(r_2 - r_1)^2}\left(e^{\lambda(r_2 - r_1)t} - 1\right).
\end{aligned}
$$

Since

$$e^{\varkappa_1(s)} - r_2 = \frac{r_2 + r_1 \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}} - r_2 = \frac{(r_1 - r_2)\frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}$$

then

$$
\begin{aligned}
J_2(t) &= \int_0^t \frac{\mathrm{d}s}{e^{\varkappa_1(s)} - r_2} = \int_0^t \frac{1 + \frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}{(r_1 - r_2)\frac{r_2 - 1}{1 - r_1}e^{\lambda(r_2 - r_1)(s - T)}}\mathrm{d}s \\
&= -\frac{t}{r_2 - r_1} - \frac{1 - r_1}{r_2 - 1} \cdot \frac{e^{\lambda(r_2 - r_1)T}}{\lambda(r_2 - r_1)^2}\left(1 - e^{-\lambda(r_2 - r_1)t}\right).
\end{aligned}
$$

Thus

$$J(t) = \left(1 + \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}\right)J_1(t) - \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}J_2(t) \tag{2.34}$$

and

$$f_2(t) = -\mu\exp\{-\varkappa_2\}I(t). \tag{2.35}$$

Therefore $\varkappa_2$ is a solution of

$$-B = f_2(T) = -\mu\exp\{-\varkappa_2\}\left[\left(1 + \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}\right)J_1(T) - \frac{\lambda(r_1 - 1) + K}{\lambda(r_2 - r_1)}J_2(T)\right]. \tag{2.36}$$

Remark that $e^{-\varkappa_2}$ is present in the definition of constants r_1, r_2 and K. Therefore (2.36) is not a linear equation with respect to $e^{-\varkappa_2}$.

3. Emergence of chaos in large fluctuations

3.1. Equations

Here we derive a surprising feature of the asymptotic 'quasi-equilibrium' behaviour of the emission and excitation rates within the interval $[0, T]$. We will prove that these rates become equal in the limit and do not depend on λ and μ.

For the hamiltonian system (2.15) with boundary conditions (2.14), let us make a change of variables $y(t) = e^{\varkappa_1(t)}$ and $z(t) = e^{-\varkappa_1(t) - \varkappa_2(t)}$. Recall that $\varkappa_2(t) = \varkappa_2$ is a constant. From (2.27), we get

$$y(t) = \frac{r_2(1 - r_1) + r_1(r_2 - 1)\varphi(t)}{1 - r_1 + (r_2 - 1)\varphi(t)}, \tag{3.1}$$

where

$$\varphi(t) = e^{(t-T)\lambda(r_2 - r_1)}. \tag{3.2}$$

We also have

$$r_2 - r_1 = \frac{\sqrt{(\lambda - \mu)^2 + 4\mu\lambda e^{-\varkappa_2}}}{\lambda},$$

(hence $0 < \varphi(t) \leqslant 1$ for $0 \leqslant t \leqslant T$) and

$$r_2 = \frac{\lambda - \mu + \sqrt{(\lambda - \mu)^2 + 4\mu\lambda e^{-\varkappa_2}}}{2\lambda}. \tag{3.3}$$

Note that the values of r_1, r_2 and $\varkappa_2$ depend on the parameter $B > 0$ and the values of $\lambda > 0$, $\mu > 0$, and $T > 0$ are fixed.

3.2. Some bounds

First, note that the right-hand side of (3.1) is monotone decreasing as ϕ changes from 0 to 1 if $r_2 > 1$ and monotone increasing in ϕ at the same interval if $r_2 < 1$. Indeed, we have

$$y(t) = \frac{r_1(1 - r_1) + (r_2 - r_1)(1 - r_1) + r_1(r_2 - 1)\varphi(t)}{1 - r_1 + (r_2 - 1)\varphi(t)}$$

$$= r_1 + \frac{(r_2 - r_1)(1 - r_1)}{1 - r_1 + (r_2 - 1)\varphi(t)},$$

where $(r_2 - r_1)(1 - r_1) > 0$ and $1 - r_1 > 0$. Hence,

$$y(t) = r_1 + \frac{c_1}{c_2 + c_3\varphi(t)},$$

where $c_1, c_2 > 0$ and where $c_3 > 0$ if $r_2 > 1$ and $c_3 < 0$ if $r_2 < 1$. Moreover, the denominator $1 - r_1 + (r_2 - 1)\varphi(t)$ is positive for all $\varphi(t) \in [0, 1]$. Then the monotonicity follows.

We have $y(t) = r_2$ if $\varphi(t) = 0$ and $y(t) = 1$ if $\varphi(t) = 1$. Since $0 \leqslant \varphi(t) \leqslant 1$, we conclude that

$$y(t) \leqslant \max\{1, r_2\} \leqslant r_2 + 1, \quad 0 \leqslant t \leqslant T. \tag{3.4}$$

Integrating the second equation in (2.15), we get

$$\mu \int_0^T z(t) f_1(t) \mathrm{d}t = B. \tag{3.5}$$

Since $0 \leqslant f_1(t) \leqslant 1$, we conclude from (3.5) that

$$\int_0^T z(t)\mathrm{d}t \geqslant \frac{B}{\mu}. \tag{3.6}$$

From the third equation in (2.15) and from the boundary condition $\varkappa_1(T) = 0$ we get the equality

$$\varkappa_1(0) = \mu \int_0^T z(t)\mathrm{d}t - \lambda \int_0^T y(t)\mathrm{d}t + (\lambda - \mu)T.$$

Next, we use (3.1) and (3.6) and derive the bound

$$\varkappa_1(0) \geqslant B - \lambda(r_2 + 1)T + (\lambda - \mu)T. \tag{3.7}$$

Since $\varkappa_1(0) = \ln y(0)$ and, hence, $\varkappa_1(0) \leqslant \ln(r_2 + 1)$, we conclude from (3.7) that

$$\ln(r_2 + 1) \geqslant B - \lambda(r_2 + 1)T + (\lambda - \mu)T. \tag{3.8}$$

In order to derive a lower bound for r_2, we denote $r = \lambda T(r_2 + 1)$ and $b = B + (\lambda - \mu)T + \ln(\lambda T)$ and reduce (3.8) to

$$\ln(r) + r \geqslant b. \tag{3.9}$$

We assume that $b > 1$ (this is true for B large enough). Let us consider two cases: $r > b$ and $r \leqslant b$. One verifies that (3.9) implies

$$r \geqslant b - \ln(b). \tag{3.10}$$

This inequality in the first case follows from $\ln b > 0$, and in the second case it follows from $\ln r \leqslant \ln b$. Hence, we have

$$r_2 \geqslant cB - d, \tag{3.11}$$

for some $c, d > 0$ and all sufficiently large $B > 0$.

A lower bound of the same form holds for $-r_1$ since $r_1 + r_2 = 1 - \mu/\lambda$. Now, let us turn to (3.2). Fix some $\alpha > 0$, $\alpha < T$ and note that

$$\varphi(t) \leqslant \mathrm{e}^{\alpha\lambda(r_2 - r_1)} \tag{3.12}$$

for all $t \in [0, T - \alpha]$. It follows that both products $r_1(r_2 - 1)\varphi(t)$ and $(r_2 - 1)\varphi(t)$ vanish uniformly for all $t \in [0, T - \alpha]$ as $B \to \infty$ (the exponent dominates polynomials). Therefore, from (3.1) we get

$$\lim_{B \to +\infty} \max_{0 \leqslant t \leqslant T - \alpha} |y(t) - r_2| = 0. \tag{3.13}$$

Now, we find the asymptotics of $z(t)$ for $t \in [0, T - \alpha]$ as $B \to +\infty$. To this end we express $\mathrm{e}^{-\varkappa_2}$ via r_2 from the equality (3.3):

$$(2\lambda r_2 - \lambda + \mu)^2 = (\lambda - \mu)^2 + 4\mu\lambda\mathrm{e}^{-\varkappa_2}. \tag{3.14}$$

Namely, we get

$$(2\lambda r_2 + (\mu - \lambda))^2 = (\mu - \lambda)^2 + 4\mu\lambda\mathrm{e}^{-\varkappa_2} \tag{3.15}$$

and then

$$4\lambda^2 r_2^2 + 4\lambda r_2(\mu - \lambda) + (\mu - \lambda)^2 = (\lambda - \mu)^2 + 4\mu\lambda e^{-\varkappa_2},$$

that is,

$$\lambda r_2^2 + r_2(\mu - \lambda) = \mu e^{-\varkappa_2}. \tag{3.16}$$

Since $z(t) = \frac{e^{-\varkappa_2}}{y(t)}$, we conclude that

$$\lim_{B \to +\infty} \frac{z(t)}{r_2} = \frac{\lambda}{\mu} \tag{3.17}$$

uniformly for all $t \in [0, T - \alpha]$.

3.3. The turnpike theorem

Let us turn to the first equation in (2.15) and write it in the form

$$\dot{f}_1(t) = \lambda y(t)(1 - f_1(t)) - \mu z(t) f_1(t), \tag{3.18}$$

where, as follows from (3.17) and (3.13),

$$\lim_{B \to +\infty} \frac{\lambda y(t)}{\lambda r_2} = 1 \quad \text{and} \quad \lim_{B \to +\infty} \frac{\mu z(t)}{\lambda r_2} = 1 \tag{3.19}$$

uniformly for all $t \in [0, T - \alpha]$.

We are ready to state and prove the main result of this section. There is a striking resemblance with a series of turnpike theorems in mathematical economics, see [17].

Theorem 3.1. *For any $\alpha > 0$ and any $\varepsilon > 0$, there exists $B_0 > 0$ such that inequality*

$$\left| f_1(t) - \frac{1}{2} \right| < \varepsilon$$

holds for all $t \in [\alpha, T - \alpha]$ whenever $B \geqslant B_0$.

Proof. We make a linear time change $\tau = \lambda r_2 t$ and derive the required assertion from the explicit solutions of resulting linear differential equations.

Rewrite (3.18) on $[0, T - \alpha]$ as

$$\dot{f}_1(t) = \lambda r_2(1 + \varepsilon_1(t))(1 - f_1(t)) - \lambda r_2(1 + \varepsilon_2(t)) f_1(t), \tag{3.20}$$

where $|\varepsilon_i(t)| \leqslant \varepsilon$ for $i = 1, 2$, and for all $t \in [0, T - \alpha]$. Because of (3.19), this is true for all B large enough. After the time change, we have

$$\frac{d}{d\tau} f_1(\tau) = (1 + \varepsilon_1(\frac{\tau}{\lambda r_2}))(1 - f_1(\tau)) - (1 + \varepsilon_2(\frac{\tau}{\lambda r_2})) f_1(\tau), \tag{3.21}$$

or, equivalently,

$$\frac{d}{d\tau} f_1(\tau) = 1 - 2f_1(\tau) + \varepsilon_0(\tau), \tag{3.22}$$

where $|\varepsilon_0(\tau)| \leqslant \varepsilon$ for all $\tau \in [0, \lambda r_2(T - \alpha)]$. It remains to note that $0 \leqslant f_1(0) \leqslant 1$ for all $B > 0$ and, hence, $f_1(\tau)$ converges to $1/2$ exponentially and uniformly in $B > 0$. Indeed, this

follows from the explicit solution

$$x(\tau) = e^{-2\tau}x(0) + \int_0^\tau \varepsilon_0(s)e^{2(s-\tau)}\mathrm{d}s,$$

where we denoted $x = f_1 - 1/2$.   □

4. Microscopic approach. Quantum Hamiltonian formalism

In this section we study the model described in the previous sections by the so-called quantum Hamiltonian formalism [16, 20]. This is somewhat a misnomer as no quantum physics is used. The name comes from certain formal analogies with linear operators used in quantum mechanics. In essence, the fundamental idea is to write the generator as a matrix and then to use linear and multi-linear algebra for explicit computations of expectations.

4.1. Notation and definitions

The first step is to consider not only the number of the excited atoms at moment t and the cumulative number of emissions during $[0, t]$, but the state $\eta_i(t)$ of each atom $i, 1 \leqslant i \leqslant N$, at moment t. These atoms form a particle system. A microstate, i.e. a microscopic configuration of the particle system, is denoted by $\eta = \{\eta_1, \ldots, \eta_N\}$ where $\eta_i \in \{0, 1\}$ denotes the state of atom i. The value 0 corresponds to the ground state, while 1 corresponds to the excited state. The state space of the particle system is therefore $\mathbb{S}_N = \{0, 1\}^N$. The number $\Upsilon_N(t)$ of emissions up to time t is a non-negative integer so that the state space S_N of the process $(\eta(t), \Upsilon_N(t))$ is $S_N = \mathbb{S}_N \times \mathbb{N}_0$. The number of excited atoms at time t is given by $\Xi_N(t) = \sum_{i=1}^N \eta_i(t)$.

For a given configuration η we shall use the shorthand η^k for the switched configuration with state variables

$$\eta_i^k = \eta_i + (1 - 2\eta_k)\delta_{k,i} \tag{4.1}$$

which corresponds to a switch of the state of atom k in the configuration η. The generator for the process then reads

$$\mathcal{L}_N f(\eta, \Upsilon_N) = \sum_{k=1}^N \left[\lambda(1 - \eta_k)\left(f(\eta^k, \Upsilon_N) - f(\eta, \Upsilon_N)\right) + \mu\eta_k\left(f(\eta^k, \Upsilon_N + 1) - f(\eta, \Upsilon_N)\right)\right]. \tag{4.2}$$

It is easy to see that one recovers (2.7) for functions $f(\Xi_N, \Upsilon_N)$ with $\Xi_N = \sum_{i=1}^N \eta_i$.

In order to apply the quantum Hamiltonian formalism we map the states of a single atom to the basis vectors $\mathfrak{e}_0 := (1, 0)^T$ (corresponding to the ground state) and $\mathfrak{e}_1 := (0, 1)^T$ (corresponding to the excited state), forming the canonical basis of $\mathbb{C}^2$. The superscript T denotes transposition, i.e. these vectors are considered to be column vectors. For the N-particle system a state η is then mapped to the tensor basis $|\eta\rangle = \mathfrak{e}_{\eta_1} \otimes \mathfrak{e}_{\eta_2} \otimes \cdots \otimes \mathfrak{e}_{\eta_N}$ of $(\mathbb{C}^2)^{\otimes N}$. Analogously we define row vectors $\langle\eta| = |\eta\rangle^T$ with orthogonality relation $\langle\eta|\eta'\rangle = \delta_{\eta,\eta'}$ where $\langle\cdot|\cdot\rangle$ denotes the usual scalar product. A Bernoulli product measure on $\mathbb{S}_N$ with density ρ_0 is thus given by the vector

$$|x\rangle = ((1 - \rho_0, \rho_0)^T)^{\otimes N} = (1 - \rho_0)^N((1, x)^T)^{\otimes N} \tag{4.3}$$

where $x = \rho_0/(1 - \rho_0)$ is the fugacity. Normalization is reflected by the property $\langle s|x\rangle = 1$ where $\langle s| = \sum_{\eta \in \mathbb{S}_N} \langle\eta| = (1, 1, \ldots, 1)$ is the so-called summation vector.

In order to consider also the number Υ_N of radiation events we define infinite-dimensional vectors $|\Upsilon_N\rangle$ with components $|\Upsilon_N\rangle_n = \delta_{n,\Upsilon_N}$ for $n \geqslant 0$. Then a state (η, Υ_N) is mapped to the tensor product $|\eta, \Upsilon_N\rangle\rangle := |\eta\rangle \otimes |\Upsilon_N\rangle$ which is a vector in $\mathbb{V} := (\mathbb{C}^2)^{\otimes N} \otimes \mathbb{C}^{\mathbb{N}_0}$ denoted by the double-ket symbol $|\cdot\rangle\rangle$. In complete analogy to above we define also $\langle\langle\eta, \Upsilon_N| = |\eta, \Upsilon_N\rangle\rangle^T$ with the orthogonality relation $\langle\langle\eta, \Upsilon_N|\eta, \Upsilon_N'\rangle\rangle = \delta_{\eta,\eta'}\delta_{\Upsilon_N,\Upsilon_N'}$. A probability measure μ on $S_N = \{0,1\}^N \times \mathbb{N}_0$ is thus represented by a vector

$$|\mu\rangle\rangle = \sum_{(\eta,\Upsilon_N)\in S_N} \mu(\eta,\Upsilon_N)|\eta,\Upsilon_N\rangle\rangle. \tag{4.4}$$

Normalization is reflected by the property $\langle\langle s|\mu\rangle\rangle = 1$ where $\langle\langle s| = \sum_{\eta,\Upsilon_N}\langle\langle\eta,\Upsilon_N| = \langle s|\otimes\langle s'|$ with $\langle s'| = \sum_{\Upsilon_N\in\mathbb{N}_0}\langle\Upsilon_N|$.

Following the quantum Hamiltonian formalism we first introduce the two-dimensional unit matrix $\mathbf{1}$ and the Pauli matrices $\sigma^{x,y,z}$. In order to build operators for the N-particle system we define the unit operator $\hat{\mathbf{1}} := \mathbf{1}^{\otimes N}$ on $(\mathbb{C}^2)^{\otimes N}$ and the unit operator $\mathbb{1}$ on $\mathbb{C}^{\mathbb{N}_0}$. In order to describe the evolution of the number of excited atoms we next introduce the diagonal number operator $\hat{N} = \sum_{i=1}^N \hat{n}_i$ which is composed of the the sum of local operators $\hat{n}_i$ with the property $\hat{n}_i|\eta\rangle = \eta_i|\eta\rangle$ and therefore $\hat{N}|\eta\rangle = \Xi_N|\eta\rangle$. The subscript i at a matrix a_i denotes the ith position in the tensor product $a_i = \mathbf{1}\otimes\cdots\otimes\mathbf{1}\otimes a\otimes\mathbf{1}\cdots\otimes\mathbf{1}$. In terms of Pauli matrices the single-atom number operator takes the form $\hat{n} = 1/2(\mathbf{1} - \sigma^z)$. We also need the operator $\hat{V} = \sum_{i=1}^N(\mathbf{1} - \hat{n}_i)$ and the (non-diagonal) flipping operators $\hat{S}^\pm = \sum_{i=1}^N \sigma_i^\pm$, where $\sigma^\pm = 1/2(\sigma^x \pm \mathrm{i}\sigma^y)$ with imaginary unit i. The matrix σ_i^+ turns excited atom i into its ground state, while σ_i^- corresponds to excitation of atom i.

For the radiation process we define the diagonal counting operator $\hat{K}$ where $\hat{K}|\Upsilon_N\rangle = \Upsilon_N|\Upsilon_N\rangle$. We also introduce the matrix $\hat{K}^+$ where $\hat{K}^+$ acts as raising operator on $\mathbb{C}^{\mathbb{N}_0}$, i.e. $\hat{K}^+|\Upsilon_N\rangle = |\Upsilon_N + 1\rangle$ with $\Upsilon_N \in \mathbb{N}_0$. We note the commutation relation

$$[\hat{K}, \hat{K}^+] = \hat{K}^+, \tag{4.5}$$

which will play a role in the computation of the radiation activity.

Finally, we lift the action of these operators to $\mathbb{V}$ by tensor multiplication with the respective unit operators: $\hat{\mathbf{N}} = \hat{N}\otimes\mathbb{1}$, $\hat{\mathbf{V}} = \hat{V}\otimes\mathbb{1}$, $\hat{\mathbf{S}}^\pm = \hat{S}^\pm\otimes\mathbb{1}$, $\hat{\mathbf{K}} = \hat{\mathbf{1}}\otimes\hat{N}$, $\hat{\mathbf{K}}^+ = \hat{\mathbf{1}}\otimes\hat{K}^+$. The generator of the process can now be written in matrix form as

$$\mathbf{H} = -\mu(\hat{\mathbf{S}}^+\hat{\mathbf{K}}^+ - \hat{\mathbf{N}}) - \lambda(\hat{\mathbf{S}}^- - \hat{\mathbf{K}}) = \sum_{i=1}^N \mathbf{A}_i, \tag{4.6}$$

with

$$\mathbf{A}_i = \mu(\hat{n}_i\otimes\mathbb{1} - \sigma_i^+\otimes\hat{K}^+) + \lambda(\tilde{\mathbf{1}} - \hat{n}_i - \sigma_i^-)\otimes\mathbb{1}. \tag{4.7}$$

For any initial measure $|\mu\rangle\rangle$ the measure at time $t \geqslant 0$ is given by

$$|\mu(t)\rangle\rangle = \mathrm{e}^{-\mathbf{H}t}|\mu\rangle\rangle. \tag{4.8}$$

Notice that the $\mathbf{A}_i$ commute among themselves and therefore $\mathrm{e}^{-\mathbf{H}t} = \prod_{i=1}^N \mathrm{e}^{-\mathbf{A}_i t}$.

Consider now the initial measure $|x\rangle\rangle = |x\rangle \otimes |0\rangle$ with the Bernoulli product measure $|x\rangle$ for the state of the atoms defined in (4.3) and define

$$F_T(x, y, z) := \mathsf{E}_x\left(y^{\Xi_N(T)} z^{\Upsilon_N(T)}\right) \tag{4.9}$$

J. Phys. A: Math. Theor. **50** (2017) 455203

E Pechersky *et al*

which is the generating function for the probability to arrive at $\Xi_N = M$ and $\Upsilon_N = K$ at time T, starting at time 0 from $K = 0$ (number of radiation events) and Bernoulli product measure $|x\rangle$ for the state of the atoms.

Moreover define for $0 \leqslant t \leqslant T$ the two-time expectations

$$G_T(x, y, z, t) := \mathsf{E}_x \left(\Xi_N(t) y^{\Xi_N(T)} z^{\Upsilon_N(T)} \right) \tag{4.10}$$

$$H_T(x, y, z, t) := \mathsf{E}_x \left(\Upsilon_N(t) y^{\Xi_N(T)} z^{\Upsilon_N(T)} \right) \tag{4.11}$$

and the scaled and normalized quantities

$$g_T(x, y, z, t) := \frac{1}{N} G_T(x, y, z, t) / F_T(x, y, z) \tag{4.12}$$

$$h_T(x, y, z, t) := \frac{1}{N} H_T(x, y, z, t) / F_T(x, y, z). \tag{4.13}$$

They can be interpreted as conditional expectations of the scaled variables $\xi_N(t)$ and $\zeta_N(t)$ of the scaled process (2.8). Thus $g_T(x, y, z, t)$ is the conditional mean number of excited atoms at time t (normalized by the total number of atoms) and $h_T(x, y, z, t)$ is the conditional number of radiation events up to time t per atom. In the following we show that these functions are the microscopic analogs of $f_1(t)$ and $f_2(t)$ of the previous section. The case when $f_1(T)$ is free corresponds to $y = 1$.

4.2. Computation of the generating functions

In the quantum Hamiltonian formalism we have by definition $F_T(x, y, z) = \langle\langle s | y^{\hat{N}} z^{\hat{K}} \mathrm{e}^{-\mathbf{H}T} | x \rangle\rangle$. Using the fact that $y^{\hat{N}}$ and $z^{\hat{K}}$ are diagonal and factorize we get that

$$F_T(x, y, z) = \langle\langle s | \mathrm{e}^{-\hat{\mathbf{H}}t} | xy \rangle\rangle \tag{4.14}$$

where $\hat{\mathbf{H}} = y^{\hat{N}} z^{\hat{K}} \mathbf{H} y^{-\hat{N}} z^{-\hat{K}} =: \sum_{i=1}^N \hat{\mathbf{A}}_i$ with $\hat{\mathbf{A}}_i = y^{\hat{n}_i} \otimes z^{\hat{K}} \mathbf{A}_i y^{-\hat{n}_i} \otimes z^{-\hat{K}}$. Next we expand the exponential of $\hat{\mathbf{H}}$ into its Taylor series and use the fact that $\langle s | \hat{\mathbf{K}}^+ = \langle s |$. It follows that $\langle s | \mathrm{e}^{-\hat{\mathbf{H}}T} = \langle s | \mathrm{e}^{-\tilde{H}T} \otimes \mathbb{1} = \langle s | \mathrm{e}^{-\tilde{H}T} \otimes \langle s' |$ with the weighted generator $\tilde{H} = \sum_{i=1}^N \tilde{A}_i$ (acting on $(\mathbb{C}^2)^{\otimes N}$) and

$$\tilde{A}_i = \mu(\hat{n}_i - y^{-1} z \sigma_i^+) + \lambda(\mathbf{1} - \hat{n}_i - y \sigma_i^-). \tag{4.15}$$

Observe that the action of $\tilde{H} \otimes \mathbb{1}$ on the subspace $\mathbb{C}^{\mathbb{N}_0}$ is trivial. Hence the inner product in (4.14) in this subspace can be factorized and is trivially equal to one. Therefore $F_T(x, y, z) = \langle s | \mathrm{e}^{-\tilde{H}T} | xy \rangle$ which is an inner product only in $(\mathbb{C}^2)^{\otimes N}$. Moreover, due the factorization of both the weighted generator and the initial measure we arrive at

$$F_T(x, y, z) = (1 - \rho_0)^N \left[(1, 1) \mathrm{e}^{-\tilde{A}T} (1, xy)^T \right]^N \tag{4.16}$$

with the 2×2 matrix

$$\tilde{A} = \begin{pmatrix} \lambda & -\mu y^{-1} z \\ -\lambda y & \mu \end{pmatrix}. \tag{4.17}$$

A general 2×2 matrix

$$M = \begin{pmatrix} a & b \\ c & d \end{pmatrix} \tag{4.18}$$

has eigenvalues $\lambda_1 = (a + d - \delta)/2$ and $\lambda_2 = (a + d + \delta)/2$ with $\delta = \sqrt{(a-d)^2 + 4bc}$. For $\delta \neq 0$ its exponential takes the form

$$\mathrm{e}^{-M\tau} = \mathrm{e}^{-\frac{(a+d)\tau}{2}} \begin{pmatrix} \cosh\frac{\delta}{2}\tau + \frac{a-d}{\delta}\sinh\frac{\delta}{2}\tau & \frac{2b}{\delta}\sinh\frac{\delta}{2}\tau \\ \frac{2c}{\delta}\sinh\frac{\delta}{2}\tau & \cosh\frac{\delta}{2}\tau - \frac{a-d}{\delta}\sinh\frac{\delta}{2}\tau \end{pmatrix}. \tag{4.19}$$

For $\tilde{A}$ we have $\delta = \sqrt{(\mu - \lambda)^2 + 4\mu\lambda z}$ and for convenience we define

$$f_T(x, y, z) := \frac{\mathrm{e}^{\frac{(\lambda+\mu)T}{2}}}{1 - \rho_0} \left(F_T(x, y, z) \right)^{\frac{1}{N}}. \tag{4.20}$$

This yields

$$f_T(x, y, z) = (1, 1) \begin{pmatrix} \cosh\frac{\delta}{2}T + \frac{\mu-\lambda}{\delta}\sinh\frac{\delta}{2}T & \frac{2y^{-1}z\mu}{\delta}\sinh\frac{\delta}{2}T \\ \frac{2y\lambda}{\delta}\sinh\frac{\delta}{2}T & \cosh\frac{\delta}{2}T - \frac{\mu-\lambda}{\delta}\sinh\frac{\delta}{2}T \end{pmatrix} \begin{pmatrix} 1 \\ xy \end{pmatrix}$$
$$= (1 + xy)\cosh\frac{\delta}{2}T + \frac{1}{\delta}\left[2y\lambda + 2xz\mu + (\mu - \lambda)(1 - xy) \right]\sinh\frac{\delta}{2}T. \tag{4.21}$$

Next we consider $G_T(x, y, z, t)$. According to definition one has

$$G_T(x, y, z, t) = \sum_{i=1}^{N} \langle\langle s | y^{\hat{N}} z^{\hat{K}} \mathrm{e}^{-\mathbf{H}(T-t)} \hat{\mathbf{N}} \mathrm{e}^{-\mathbf{H}t} | x \rangle\rangle. \tag{4.22}$$

In a similar fashion to above one finds

$$G_T(x, y, z, t) = N(1 - \rho_0)^N \mathrm{e}^{\frac{-(\lambda+\mu)(N-1)T}{2}} f_T^{N-1}(x, y, z) \left[(1, 1)\mathrm{e}^{-\tilde{A}(T-t)}\hat{n}\mathrm{e}^{-\tilde{A}t}(1, xy)^T \right], \tag{4.23}$$

and, using (4.19),

$$g_T(x, y, z, t) = \frac{1}{f_T(x, y, z)} \left[y\cosh\frac{\delta}{2}(T - t) + \frac{2z\mu - y(\mu - \lambda)}{\delta}\sinh\frac{\delta}{2}(T - t) \right]$$
$$\times \left[x\cosh\frac{\delta}{2}t + \frac{2\lambda - x(\mu - \lambda)}{\delta}\sinh\frac{\delta}{2}t \right] \tag{4.24}$$

with initial condition

$$g_T(x, y, z, 0) = \frac{x}{f_T(x, y, z)} \left[y\cosh\frac{\delta}{2}T + \frac{2z\mu - y(\mu - \lambda)}{\delta}\sinh\frac{\delta}{2}T \right] \tag{4.25}$$

parametrized by the fugacity $x = \rho_0/(1 - \rho_0)$.

Next we note that

$$H_T(x, y, z, t) = \langle\langle s | y^{\hat{N}} z^{\hat{K}} \mathrm{e}^{-\mathbf{H}(T-t)} \hat{\mathbf{K}} \mathrm{e}^{-\mathbf{H}t} | x \rangle\rangle. \tag{4.26}$$

In order to compute this quantity it is convenient to consider its derivative w.r.t. t. Using $\mathrm{d}/\mathrm{d}t\, \mathrm{e}^{\mathbf{H}t} \hat{\mathbf{K}} \mathrm{e}^{-\mathbf{H}t} = \mathrm{e}^{\mathbf{H}t}[H, \hat{\mathbf{K}}]\mathrm{e}^{-\mathbf{H}t}$ and (4.5), steps analogous to those that gave rise to (4.21) lead to

$$
\dot{H}_T(x,y,z,t) = \mu y^{-1} z \sum_{i=1}^{N} \langle\langle s \,|\, \mathrm{e}^{-\hat{\mathbf{H}}(T-t)} \hat{\mathbf{S}}^+ \hat{\mathbf{K}}^+ \mathrm{e}^{-\hat{\mathbf{H}}t} \,|\, xy \rangle\rangle
$$
$$
= N(1-\rho_0)^N \mathrm{e}^{\frac{-(\lambda+\mu)(N-1)T}{2}} f_T^{N-1}(x,y,z) \left[(1,1)\mathrm{e}^{-\tilde{A}(T-t)} \sigma^+ \mathrm{e}^{-\tilde{A}t}(1,xy)^T \right].
$$
(4.27)

Using (4.19) this yields

$$
\dot{h}_T(x,y,z,t) = \frac{z\mu}{f_T(x,y,z)} \left[\cosh\frac{\delta}{2}(T-t) + \frac{2y\lambda+\mu-\lambda}{\delta} \sinh\frac{\delta}{2}(T-t) \right]
$$
$$
\times \left[x\cosh\frac{\delta}{2}t + \frac{2\lambda-x(\mu-\lambda)}{\delta} \sinh\frac{\delta}{2}t \right].
$$
(4.28)

Integration of this quantity w.r.t. t is straightforward and yields $h(t)$ with initial condition $h(0)=0$. The final values $g_T(x,y,z,T) \in [0,1]$ and $h_T(x,y,z,T) \geqslant 0$ are parametrized by the variables y, z.

Remark 4.1. In the limit $T \to \infty$ straightforward computation for intermediate times $t \propto cT$ shows that

$$
g^*(z) := \lim_{T\to\infty} g_T(x,y,z,cT) = \frac{1}{2}\left(1 + \frac{\lambda-\mu}{\delta} \right)
$$
(4.29)

$$
\dot{h}^*(z) := \lim_{T\to\infty} \dot{h}_T(x,y,z,cT) = \frac{z\lambda\mu}{\delta}
$$
(4.30)

are constants that depend neither on the initial fugacity x nor on the atypical density of excited atoms parametrized by y. Moreover, for any choice of λ and μ the radiation activity $\dot{h}^*(z)$ is monotone in z, i.e. one has $\dot{h}^*(z) \neq \dot{h}^*(1)$ for $z \neq 1$. If $\lambda = \mu$ where radiation and excitation are in balance one finds somewhat surprisingly that, despite the conditioning on a strongly atypical number of excited atoms at time T and a strongly atypical number of radiation events up to time T, the mean number of excited atoms at intermediate times $t = O(T)$ is 1/2, which for $\lambda = \mu$ is the typical value without conditioning.

4.3. The case $\lambda = \mu = 1$

The expressions derived above simplify for $\lambda = \mu = 1$ corresponding to $\gamma = 1$ in (2.22). We present this case in more detail. One gets $\delta = 2\sqrt{z}$ and it is convenient to introduce $\tilde{y} := y/\sqrt{z}$ and $\tilde{x} := x\sqrt{z}$. With this the expression obtained above for the general case reduce to

$$
f_T(x,y,z) = \frac{1}{2}\left[(1+\tilde{y})(1+\tilde{x})\mathrm{e}^{\sqrt{z}T} + (1-\tilde{y})(1-\tilde{x})\mathrm{e}^{-\sqrt{z}T} \right]
$$
(4.31)

$$
g_T(x,y,z,t) = \frac{1}{2}\left(1 - \frac{(1+\tilde{y})(1-\tilde{x})\mathrm{e}^{\sqrt{z}(T-2t)} + (1-\tilde{y})(1+\tilde{x})\mathrm{e}^{-\sqrt{z}(T-2t)}}{(1+\tilde{y})(1+\tilde{x})\mathrm{e}^{\sqrt{z}T} + (1-\tilde{y})(1-\tilde{x})\mathrm{e}^{-\sqrt{z}T}} \right)
$$
(4.32)

$$
\dot{h}_T(x,y,z,t) = \frac{\sqrt{z}}{2}\left(h^* - \frac{(1+\tilde{y})(1-\tilde{x})\mathrm{e}^{\sqrt{z}(T-2t)} - (1-\tilde{y})(1+\tilde{x})\mathrm{e}^{-\sqrt{z}(T-2t)}}{(1+\tilde{y})(1+\tilde{x})\mathrm{e}^{\sqrt{z}T} + (1-\tilde{y})(1-\tilde{x})\mathrm{e}^{-\sqrt{z}T}} \right)
$$
(4.33)

with

$$h^* = \frac{(1+\tilde{y})(1+\tilde{x})e^{\sqrt{z}T} - (1-\tilde{y})(1-\tilde{x})e^{-\sqrt{z}T}}{(1+\tilde{y})(1+\tilde{x})e^{\sqrt{z}T} + (1-\tilde{y})(1-\tilde{x})e^{-\sqrt{z}T}}. \tag{4.34}$$

For large time frame T one gets with the definition $\tau = T - t$ the following asymptotic behaviour for $g(x, y, z, t) := \lim_{T\to\infty} g_T(x, y, z, t)$:

$$g(x, y, z, t) = \begin{cases} \frac{1}{2}\left(1 - \frac{1-\tilde{x}}{1+\tilde{x}}e^{-2\sqrt{z}t}\right) & \text{for } t = o(T) \\ \frac{1}{2} & \text{for } t = O(T) \\ \frac{1}{2}\left(1 - \frac{1-\tilde{y}}{1+\tilde{y}}e^{-2\sqrt{z}\tau}\right) & \text{for } \tau = o(T) \end{cases}. \tag{4.35}$$

The corresponding asymptotics for $h(x, y, z, t) := \lim_{T\to\infty} h_T(x, y, z, t)$ are:

$$h(x, y, z, t) = \begin{cases} \frac{\sqrt{z}}{2}t - \frac{1}{4}\frac{1-\tilde{x}}{1+\tilde{x}}e^{-2\sqrt{z}t} & t = o(T) \\ \frac{\sqrt{z}}{2}t & t = O(T) \\ \frac{\sqrt{z}}{2}t - \frac{1}{4}\frac{1-\tilde{y}}{1+\tilde{y}}e^{-2\sqrt{z}\tau} & \tau = o(T) \end{cases}. \tag{4.36}$$

Thus, for T large, the radiation intensity $\dot{h}_T(x, y, z, t)$ is approximately constant except initially and towards the end of the observation period T.

In order to make contact with the macroscopic results of the previous sections we take T fixed, choose $y = 1$, and impose the initial condition $g_T(x, y, z, t) = 1/2$. This requires choosing the initial fugacity x such that $(1+\tilde{y})(1-\tilde{x})e^{\sqrt{z}T} + (1-\tilde{y})(1+\tilde{x})e^{-\sqrt{z}T} = 0$, i.e. for $\tilde{y} = 1/\sqrt{z}$ one chooses

$$\frac{1-\tilde{x}}{1+\tilde{x}} = \frac{1-\sqrt{z}}{1+\sqrt{z}}e^{-2\sqrt{z}T} =: C. \tag{4.37}$$

Thus one obtains for $t \in [0, T]$

$$g_T(x, 1, z, t) = \frac{1}{2} + \frac{C}{1 - C^2}\sinh(2\sqrt{z}t) \tag{4.38}$$

$$h_T(x, 1, z, t) = \frac{\sqrt{z}}{2}\left(1 - \frac{2}{1 - C^2}\right)t + \frac{C}{2(1 - C^2)}\sinh(2\sqrt{z}t). \tag{4.39}$$

With the choice $z = e^{-\varkappa_2}$ (see (2.22)) one finds $f_1(t) = g_T(x, 1, z, t)$ and $f_2(t) = h_T(x, 1, z, t)$ (see (2.30) and (2.35) together with (2.31), (2.34) and (2.36) for $\lambda = \mu = 1$), thus relating the macroscopic constant $\varkappa_2$ appearing in the Hamiltonian (2.11) to the generating function parameter z introduced in (4.9).

We point out two other special cases. (a) For initial fugacity $x = 1/\sqrt{z}$ and arbitrary y one has

$$f_T(1/\sqrt{z}, y, z, t) = (1 + \tilde{y})e^{\sqrt{z}T} \tag{4.40}$$

$$g_T(1/\sqrt{z}, y, z, t) = \frac{1}{2}\left(1 - \frac{1-\tilde{y}}{1+\tilde{y}}e^{-2\sqrt{z}(T-t)}\right) \tag{4.41}$$

$$h_T(1/\sqrt{z}, y, z, t) = \frac{\sqrt{z}}{2}t + \frac{1}{4}\frac{1-\tilde{y}}{1+\tilde{y}}\left(e^{-2\sqrt{z}(T-t)} - e^{-2\sqrt{z}T}\right). \tag{4.42}$$

Hence for initial times such that $T - t \gg 1/\sqrt{z}$ both the number of excited atoms and the radiation activity are essentially constant. (b) On the other hand, for $y = \sqrt{z}$ and arbitrary initial fugacity x the expressions (4.31)–(4.33) reduce to

$$f_T(x, \sqrt{z}, z, t) = (1 + \tilde{x})\mathrm{e}^{\sqrt{z}T} \tag{4.43}$$

$$g_T(x, \sqrt{z}, z, t) = \frac{1}{2}\left(1 - \frac{1 - \tilde{x}}{1 + \tilde{x}}\mathrm{e}^{-2\sqrt{z}t}\right) \tag{4.44}$$

$$h_T(x, \sqrt{z}, z, t) = \frac{\sqrt{z}}{2}t + \frac{1}{4}\frac{1 - \tilde{x}}{1 + \tilde{x}}\left(\mathrm{e}^{-2\sqrt{z}t} - 1\right). \tag{4.45}$$

In this case the number of excited atoms and the radiation activity approach a constant for times $t \gg 1/\sqrt{z}$.

Remark 4.2. In all cases the radiation intensity is not constant, but has deviations from a constant in time windows of length $1/(2\sqrt{z})$, i.e. the intensity approaches a constant (or deviates from it) exponentially with a decay rate $1/(2\sqrt{z})$. We refer to this behaviour as the occurence of 'moderate bursts'.

Remark 4.3. The quantum Hamiltonian approach makes clear that the behaviour of a two-level system is generic. In a m-level system the functions g and h (and their analogs for similar observables) will always be of the factorized form $g_T(\ldots, t) = a(\ldots, T - t)b(\ldots, t)$ and the functions a and b will be sums of exponentials of the eigenvalues of the single-atom generator A.

Acknowledgments

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