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On the statistical background of quantum mechanics: generalities and a concrete example

Martine Le Berre[1]* and Yves Pomeau[1]

*Correspondence: martine.le-berre@universite-paris-saclay.fr
[1]Laboratoire d'Hydrodynamique, Ladhyx, (CNRS UMR 7646), Ecole Polytechnique, 91128 Palaiseau, France

Abstract

We revisit our description of randomness in quantum processes that began in collaboration of Jean Ginibre. The calculations were performed on a worked example: the fluorescence of a single two-level atom pumped by a resonant laser field. This pump laser is described classically (by a function, not an operator). Our aim is first to built a Kolmogorov-type equation (K-equation) for the atomic state, so that the two parameters θ, φ that define this density matrix are random functions of time, therefore the atomic density matrix is a random density matrix. Such an approach, initiated for gas kinetics, was not yet applied to quantum phenomena, whereas it is especially tailored to very quick events well separated (in time) like the quantum jumps observed in spontaneous emission of photons by an atom. Here, we try to clarify the basis of our statistical approach leading to the K-equation below, and we present the main results deduced from it. We explain finally that our approach can be interpreted in terms of Everett's theory of many-worlds, because at every emission a new history begins for the atom, with two nonoverlapping wave functions.

Keywords: Statistical background of quantum mechanics; Quantum jumps; Everett's interpretation of spontaneous emission

1 Introduction

Explaining randomness and irreversibility in quantum processes remains difficult because the equations of quantum mechanics (Schrödinger and Dirac) are formally deterministic and the Hamiltonian is a unitary time operator, which seems to forbid the introduction of a fundamental uncertainty in the predictions one can make from given initial data. However, in the case of open systems interacting with surroundings and with pump fields, there are dissipation (via the surroundings) and refeeding mechanisms (via the pump fields), both playing a role when correctly describing a physical system.

For that reason, we began in collaboration with Jean Ginibre to study a concrete example, the fluorescence of a single two-level atom [1], pumped continuously by a quasiresonant monomode laser. The atom interacts with the pump and surroundings, that involves infinitely many degrees of freedom. Although this problem is a worked example already treated in many well-recognized publications and textbooks since 1969, see references in

the paper of this issue by Reynaud [2], we aimed to solve it in another way. Taking into account the very short duration of each photoemission (named quantum jumps) with respect to other time scales we proposed to treat this type of phenomena by a Kolmogorov equation for the dynamics of the quantum atomic state.

As shown by Dirac [3] the duration of a quantum jump is about a few periods of the laser in resonance with the atomic transition, $\tau_{q.j} \approx \text{few}\frac{2\pi}{\omega}$, whereas in between two successive "rare" quantum jumps, the atomic state undergoes Rabi oscillations of period $2\pi/\Omega$ assumed to be much longer than $\tau_{q.j.}$. Additionally, the interaction with the surroundings is responsible for the decay time $1/\gamma$, a quantity also derived by Dirac, and supposed to be much longer than $\tau_{q.j.}$. In summary, we have

$$\tau_{q.j} \ll \frac{1}{\Omega}, \frac{1}{\gamma}. \tag{1}$$

Notwithstanding its shortness this quantum jump changes by a finite amount the state of the atom.

The atom is partly maintained above the ground state by an external drive (or pumping). In our description we treat the pump laser classically, i.e., the electromagnetic field is a complex function of space and time, not an operator. Therefore, our description is different from the dressed-atom representation presented by Serge Reynaud in this issue. We shall discuss this point below. Our goal is to put together the principles of quantum physics and nonequilibrium concepts described by a statistical picture involving a Kolmogorov-type equation because this problem has a structure similar to the kinetic theory of dilute gases: there the long scale is that of a nonequilibrium process like the mean free flight time and the short one is the duration of a binary collision. The whole process (quick collisions) and slow macroscopic phenomena are all described by Boltzmann kinetic theory. In the case of the fluorescent atom satisfying Eq. (1) the pumping puts the atom in a "pure" state in between two successive quantum jumps, which is described [4] by the wave function

$$\Psi_{at}(t) = \cos\theta(t)|g\rangle - ie^{-i(\omega_L t-\varphi)}\sin\theta(t)|e\rangle, \tag{2}$$

where the two atomic states are the ground state $|g\rangle$ and the excited state $|e\rangle$ and $\omega_L = \omega_e - \omega_g$ at exact resonance. The phase φ is constant between two successive quantum jumps, but changes randomly at each jump. The phase values in successive time intervals are supposed to be independent variables and the atomic state is described at this stage by a probability distribution $p(\theta, \varphi, t)$. After integration over the variable φ, the Kolmogorov equation for $p(\theta, \varphi, t)$ reduces to what we call below the K-equation for the probability distribution $p(\theta, t)$ of the atomic state indexed by a single random function $\theta(t)$, see Eq. (12). An important consequence of our statistical picture is to solve the logical difficulty associated to the "collapse of the wave packet" that has existed from the very beginning of quantum mechanics. This is done by our description of the atomic density matrix that becomes a two-by-two *random matrix* with the associated probability $p(\theta, t)$.

Here, we revisit our K-equation already presented in [1, 4, 5] and the statistical quantities related to the emission process. We detail in Sect. 2 the basis of our statistical theory. Section 3 is devoted to our K-equation. We explain first how our K-equation was built up, then we present our main results, namely the statistics of the quantum jumps (probability distribution of the time intervals between two successive jumps) that is compared with the

expression derived in [2, 6] via the dressed-atom method, then we recall how to derive the spectrum of the spontaneous emission. In Sect. 5 we use our method to put into evidence the irreversible character of the fluorescence because the quantum jumps are essentially nonequilibrium processes. Finally, in order to emphasize the key point of our approach leading to the evolution of the atomic random matrix, we explain in Sect. 6 how our description of the fluorescence is compatible with Everett's many-worlds interpretation of quantum mechanics.

2 Return to Dirac's work

Our problem is to mingle the random emission of photons by an atom and the continuous pumping of this atom by a quasiresonant monomode laser field. Before laying out our method, let us make some historical recap that motivated our work. The first quantum theory of the interaction between light and matter was by Einstein and Planck. In modern terms, it is about the balance of energy of an atom with two quantum levels interacting with black-body radiation. The atom jumps back and forth between the two levels in such a way that the ratio of population of the two levels follows the Boltzmann distribution. We have (after many others but differently!) considered a different problem where the excitation is not by black-body radiation but by a laser wave of amplitude large enough to act like a classical field with negligible quantum fluctuations.

After Einstein's model, Dirac in 1927 [3] at the age of 25 imagined the perturbation calculus in quantum mechanics, which is nearly the only task accessible. He derived a Hamiltonian describing the interaction of an atom with EM waves (which can be the vacuum field, also named the bath, or surroundings in our words) and showed that his theory leads to the correct expressions for the Einstein coefficients A and B for emission and absorption of radiation. He derived in Sect. 7 the rate of emission, denoted as γ below, which he interpreted in a probabilistic way. This makes what is called the "Fermi golden rule" although it was left basically unchanged by Fermi [7]. In modern script it gives

$$\Gamma_{i\to f} = \frac{2\pi}{\hbar}\left|\langle f|H'|i\rangle\right|^2 \rho(E_f), \tag{3}$$

where $\langle f|H'|i\rangle$ is the matrix element (in bra–ket notation) of the perturbation H' between the final and initial states, $\hbar$ is the reduced Planck constant, and $\rho(E_f)$ is the density of states (number of continuum states divided by dE in the infinitesimally small energy interval E, $E + dE$ at the energy E_f of the final states).

The next order of Dirac's calculation yields (part of) the "radiative" corrections to the energy levels, the small parameter being the coupling constant between radiation and electrons, the so-called fine-structure constant

$$\alpha = \frac{e^2}{4\pi\,\varepsilon_0 \hbar c}, \tag{4}$$

where e is the election charge, ε_0 is the vacuum permittivity, and c is the light velocity in vacuum. Dirac, without insisting much on the meaning of what he does, introduces clearly the idea that he computes the probability per unit time of a transition of the atom, initially in the excited state, to the ground state. Let us set $|i\rangle = |e\rangle$ and $|f\rangle = |g\rangle$. For an interaction Hamiltonian $H' = \vec{\mathbf{E}}.\vec{\mathbf{D}}$, (where $\vec{\mathbf{E}}$ and $\vec{\mathbf{D}} = e\mathbf{r}$ are the EM field and transition dipolar moment operators) and a final state at resonance with the atomic transition, $E_e =$

$E_g + \hbar\omega$, there are a continuum of possible final states, those associated to the polarization and direction of this mode. After integration over these final states one obtains an explicit formula

$$\gamma = \frac{\omega^3 |d|^2}{3\pi\varepsilon_0 \hbar c^3}, \tag{5}$$

where d is the dipolar transition moment, $|d|^2 = (\pi\varepsilon_0\hbar c)4\alpha|\langle g|\mathbf{r}|e\rangle|^2$. Therein, Dirac implicitly associates the probabilistic character of the atomic transition with the infinite number of degrees of freedom of the quantum-electrodynamics vacuum.

Contrary to Fermi and many others since, Dirac limits himself to the initial stage of the decay process (γt small) of the (initially excited) atom coupled to the EM-field. The expression for the decay of the amplitude of the excited state is correct only if $t\gamma \ll 1$. According to Dirac, such a secular term (proportional to t) has a statistical meaning. It cannot be used to define a change of the wave function in the ordinary sense. The exponentially decaying amplitude of the excited state, proportional to $exp(-\gamma t)$, is obviously correct if the excited state is not continuously reinitialized by external pumping at the resonant frequency, but it is not a fair picture of the fairly complex process where the atom decays by spontaneous emission, whereas it is continuously pumped back to the excited state. In this case there is no reason in general to assume an exponential decay of the excited state so that the full picture should include both the spontaneous decay and the pumping without assuming a particular time dependence of the fields involved, a dependence that must follow from the solution of the dynamical problem. This produces the theory we shall present below.

3 Our approach

The need for a statistical theory to describe the randomness of the emission of photons requires some explanation. The formalism of quantum mechanics makes it abundantly clear that a complete knowledge of the state of "the system" follows from the knowledge of its density matrix. In the present case, because of the coupling between the atom and the field of photons, this density matrix must include the atom itself and the quantum states of infinitely many degrees of freedom of EM field in free space (emitted photons, plus vacuum and pump laser), not an easy task. However, the problem is made tractable by observing that the interaction between the atom and the modes of the vacuum field is very quick and so change the state of the atom almost instantaneously with respect to other processes, like the Rabi oscillations. The way to describe such phenomena with a very different time scale has been looked at by Kolmogorov [8]. We propose a Kolmogorov equation for the dynamics of the probability distribution of the variable $\theta(t)$ (depending on time) that is actually a Markov random function when the nerly instantaneous quantum jumps are included.

As written in the introduction, the shortest time is the period of the emitted EM wave, also the order of magnitude of the duration of the emission process as noted by Dirac. The two other much longer time scales are $1/\gamma$ defined in (5), and $1/\Omega$, where

$$\Omega = -\frac{d\mathcal{E}}{\hbar} \tag{6}$$

is the rate of pumping, or frequency of the Rabi optical oscillations induced by the single-mode laser (pump) field of amplitude $\mathcal{E}$. Our statistical theory is valid in the range of Eq. (1)

but otherwise makes no assumption on the relative values of the "long time scales" and is therefore correct for any time scales much longer than the period of the emitted photon. This is reminiscent of Boltzmann kinetic theory [9] for a dilute gas, where he only assumes that the time scales under consideration are much longer than the duration of two-body collisions, which is the shortest time scale.

In this situation two phenomena take place:

1) The atom oscillates between its two states because of its interaction with the quasiresonant laser field. This is the phenomenon of Rabi oscillations.

2) Spontaneous quantum jumps occur randomly from the excited to the ground state with a photon emitted. This is the phenomenon of fluorescence.

Our physical picture of the emission process including these two phenomena is actually fairly simple.

The quantum jumps performed by the atom from an excited state to the ground state are assumed to be instantaneous. After each jump the atom begins a new Rabi oscillation under the effect of the driving field, an oscillation starting from the ground state. In this step of its dynamics, its quantum state can be seen as a "random pure state" made of the addition of the ground-state wave function and of the excited wave function of the excited state with random amplitudes. After some (random) time the atom jumps back to the ground state by emitting a photon and the process continues. There is a slightly nonobvious point to describe this sequence because, as compared with the situation studied by Dirac, the initial state before the jump is not a pure exited state, but a linear superposition of the excited and ground state. This leads to a change in the rate of relaxation by emission of a photon that depends on the parameter θ in Eq. (2), associated to the population of the two states. Additionally, the phase φ, which is constant in phase (1), changes at each jump. This picture of the dynamics of the atomic state between two consecutive jumps differs from what is usually in the literature where the evolution between two successive jumps depends explicitly on a damping term due formally to the emission of photons, an assumption seemingly contradicting the starting point, namely that one is considering an interval of time between two jumps and so without emission of a photon.

Let $a_g(t)$ and $a_e(t)$ be the complex amplitudes of the two atomic states. As was done by Rabi [10], one can write the coupled equations for the amplitudes when an atom is pumped at the nearly resonance frequency. In the realistic limit of a "small" atom–pump interaction, with a pump field large enough to be classical, the coupled Rabi equations at exact resonance take a simple form

$$\partial_t a_g = -i\frac{\Omega}{2} a_e e^{-i\varphi}; \qquad \partial_t a_e = -i\frac{\Omega}{2} a_g e^{i\varphi}. \tag{7}$$

Let us evaluate the validity of the semiclassical description we used, although the light beam illuminating the atom is made of photons. Those photons are supposed to be sufficiently numerous to make the quantum fluctuation of the pumping beam negligible. Following the general principle of quantum mechanics this is correct if the beam is made of a large enough number of photons. The obvious question is how to compute this number in a given situation with a given intensity of the light beam? The knowledge of the intensity of this light beam yields a flux of power. Let Φ be this flux, it has the dimension of an energy times c, speed of light per unit area. This yields also a number of photons per unit area by dividing by the energy quantum $\hbar\omega$. In experiments one tries to focus as much as possible

of this flux in a narrow surface, this being limited by the effect of diffraction to an area σ of order of the square of the wavelength. Let r_0^3 be the approximate volume of the atom (of order of the cube of the de Broglie wavelength of the bound electron). Therefore, an approximation of the number of photons of the light beam inside the atom is $N \sim (\frac{\Phi}{\hbar\omega c r_0^3 \sigma})$. This number must be large to make the assumption of a classical pump beam valid. Even if this number is large, as it always is, there are presumably effects due to its finiteness. Without going into a detailed theory of what happens when N is large but finite, as is always the case, we guess that the quantum fluctuations of N yield random time fluctuations of the Rabi frequency Ω. Such fluctuations should yield a slight departure of the exactness of the splilting of the wave function of the atom into a sum of the ground state and excited wave function and so yield a slight departure of the density matrix from its simple form valid for a coherent superposition of the two eigenfunctions. The effect of such a decay of the coherence of the density matrix could perhaps be detected by decreasing the intensity of the pumping field in Dehmelt-like [11–13] experiments.

Note that the Rabi Eqs. (7) for the amplitudes a_g and a_e are reversible (under complex conjugation) and linear. The total norm $|a_g|^2 + |a_e|^2$ is a constant of the motion. This norm is the Noether invariant associated to the invariance under a global phase shift of the equations. Those properties are trivial in the two-level case, unfortunately they are far less trivial in the three-level case with two pump fields.

Neglecting a global phase (identical for the two amplitudes) that plays no role, and setting

$$\partial_t \theta = \frac{\Omega}{2} \tag{8}$$

the solution of Eqs. (7) is

$$a_g(t) = \cos\theta(t) \qquad a_e(t) = -ie^{i\varphi}\sin\theta(t), \tag{9}$$

in agreement with Eq. (2). This periodic solution describes the Rabi oscillations, but a fundamental physical process is absent from the picture, the random emission of photons occurring when the atom is in the excited state.

It remains to put into a coherent framework this phenomenon of a spontaneous jump from the excited to the ground state. As it is random, it is monitored by a time-dependent probability distribution of the parameters (θ, φ), which will be be interpreted below. As explained in our previous papers [1–4], after integration over the variable φ, we have reduced the K-equation for $p(\theta, \phi, t)$ to the probability $p(\theta, t)$ presented just below. We shall explain in Sect. 6 that this probability is a way to measure the distribution of the states of the atom in all Everett universes at any given time.

4 K-equation for the random function $\theta(t)$

To describe briefly what is the generic K-equation, let us introduce a parameter Θ (which may differ from the angle θ introduced above) that changes following two physical process. There is first a deterministic evolution such that Θ changes continuously and smoothly at a velocity $\nu(\Theta)$, whereas it changes abruptly by quantum jumps of finite amplitude occurring randomly at a rate depending on Θ.

The generic K-equation gives the rate of change of the probability distribution of Θ, denoted as $p(\Theta, t)$. It is [8]

$$\partial_t p(\Theta, t) + \partial_\Theta\big(\nu(\Theta)p(\Theta, t)\big) = \int \mathrm{d}\Theta_1 \Gamma(\Theta|\Theta_1)p(\Theta_1, t) - p(\Theta, t)\int \mathrm{d}\Theta' \Gamma\big(\Theta'|\Theta\big), \quad (10)$$

where $\partial_t \Theta = \nu(\Theta)$ and $\Gamma(\Theta'|\Theta)$ is the positive probability of transition per unit time from Θ to Θ' (the jump). Note that the very existence of the probability transition $\Gamma(\cdot)$ implies that we are considering a Markov process where the transition rate depends on the present state of the system only. On the right-hand side the first (positive) term (or gain term) describes the increase of the probability of the Θ-state due to jumps from other states to Θ. The second term represents the loss of probability because of jumps from Θ to any other state Θ'.

In the case of a fluorescent atom, we have to define the velocity function $\nu(\Theta)$ and the transition probability depending on the variable $\Theta = \theta$. The function $\nu(\Theta)$ in (10) is now the time derivative of $\theta(t)$, which is equal to $\Omega/2$ at exact resonance. The randomness of θ is a consequence of the randomness of the time of jump, which requires to define the transition kernel $\Gamma(\theta|\theta')$, which is the probability that the atom in state θ' jumps to state θ. This probability is proportional to $\delta(\sin\theta)$, δ is the Dirac delta function, because all jumps land on the $\theta = 0$ ground state. Moreover, the probability of a jump is proportional to $\sin^2\theta'$ because the excited state a_e has amplitude $\sin^2\theta'$, and lastly is proportional to γ, the jump frequency from a pure excited state, the one calculated by Dirac when the atom is in the excited state characterized by $\theta' = \pi/2$.

Therefore, one has

$$\nu(\Theta) = \frac{\Omega}{2}, \qquad \Gamma\big(\theta|\theta'\big) = \gamma \sin^2\theta'\delta(\sin\theta). \quad (11)$$

Finally, the K-equation for a two-state single atom submitted to a resonant wave field becomes

$$\partial_t p + \frac{\Omega}{2}\partial_\theta p = \gamma\left(\delta\big(\sin(\theta)\big)\left(\int_{-\pi/2}^{\pi/2} \mathrm{d}\theta' p\big(\theta', t\big)\sin^2\big(\theta'\big)\right) - p(\theta, t)\sin^2(\theta)\right). \quad (12)$$

The left-hand side of this equation describes the Rabi oscillation, which amounts to a uniform drift in time of the angle θ. The right-hand side represents the effect of the spontaneous decay of the excited state toward the ground state; it has a gain term for the ground state $\theta = 0$ and a loss term for any other value of θ. Our K-equation satisfies the constraints that a probability remains positive if it is initially so, and that its L^1 norm, $\int_{0_-}^{\pi} \mathrm{d}\theta p(\theta, t)$, is conserved for any periodic distribution of the variable θ.

The calculation of any averaged physical quantity requires the knowledge of both the stationary probability distribution $p_{st}(\theta)$ and the conditional probability $p(\theta, t|\theta_0)$.

4.1 Stationary distribution

The stationary distribution was derived in Sect. 3.4 of Ref. [1]. As it is an exact formula, and plays a crucial role in the following, let us give a flavor of its derivation from the K-equation. It results from the formal integration of

$$\partial_\theta \hat{p}_{st}(\theta) = -\gamma' \sin^2(\theta)\hat{p}_{st}(\theta), \quad (13)$$

where $\gamma' = 2\gamma/\Omega$. The solution is

$$\hat{p}_{st}(\theta) = \hat{p}_{st}(0_+)e^{-\frac{\gamma'}{4}(2\theta - \sin(2\theta))}. \tag{14}$$

It can be checked that the solution of Eq. (13) is a periodic function of θ of period π because by integrating the equation from 0 to π one obtains zero on the right-hand side, whereas the left-hand side is proportional to the difference $p_{st}(\pi/2) - p_{st}(-\pi/2)$, which is also zero. This solution is formally not convenient because the exponent is not periodic with respect to θ. The periodicity is restored by noting that the solution has a jump at $\theta = 0$. This jump is such that the value of $p_{st}(\theta)$ for $\theta = 0_-$ is equal to $\hat{p}_{st}(\theta)$ for $\theta = \pi_-$. The constant of integration $\hat{p}_{st}(0_+)$

$$\hat{p}_{st}(0_+) = (\mathcal{I}_\pi)^{-1} \tag{15}$$

is derived from the norm constraint, where

$$\mathcal{I}_x = \int_0^x \mathrm{d}\theta\alpha(\theta) \tag{16}$$

and

$$\alpha(\theta) = e^{-\frac{\gamma'}{4}(2\theta - \sin(2\theta))}. \tag{17}$$

The stationary probability distribution is the wrapped periodic function built by translating the solution $\hat{p}_{st}(\theta)$, with a jump included

$$p_{st}(\theta) = \sum_{k=-\infty}^{\infty} \hat{p}_{st}(\theta - k\pi). \tag{18}$$

We plot in Fig. 1 this π-periodic function that is discontinuous for $\theta = k\pi$, and smooth elsewhere.

Note that formally Eq. (12) is solvable in Laplace transform but the inverse Laplace transform requires to find poles in the complex plane, a difficult task. Therefore, we turned to solve directly the K-equation. The smart derivation of a quasianalytical solution performed by Jean Ginibre is wholly retrieved here in the Appendix as a token of

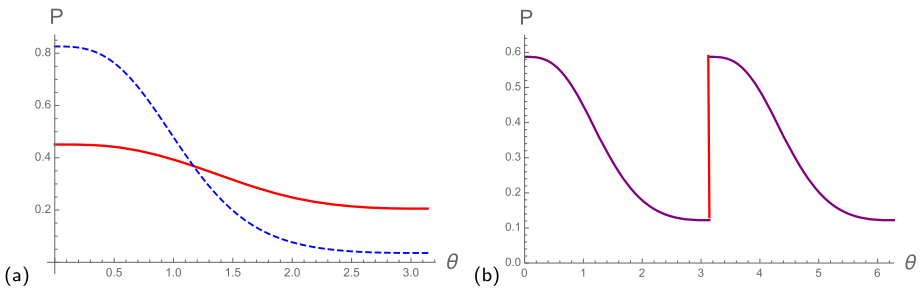

Figure 1 Stationary probability distribution $p_{st}(\theta)$, solution of Eq. (13). (**a**) In the interval $[0,\pi]$, solid line for $\gamma' = 0.5$, dashed line for $\gamma' = 2$. The periodicity of the wrapped distribution is illustrated in (**b**) for $\gamma' = 1$ on the interval $[0,2\pi]$

gratitude (first published in Ref. [1]). The result is the conditional probability $p(\theta, t|\theta_0)$ in Eq. (50) below, where $\tilde{t} = \Omega t/2$. It requires to solve an implicit function for the function $b(\tilde{t}) = \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta' p(\theta', \tilde{t}) \sin^2(\theta')$.

Let us now present a nontrivial result, very important to derive the intensity of the point process formed by the emitted photons in the observer word (for a discussion on this topic, see Sect. 6). Considering the atomic wave function in Eq. (2), the populations of the two levels, or probabilities for the atom to be in the excited or in the ground state at time t, are, respectively,

$$\rho_e(t) = \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta p(\theta, t) \sin^2\theta \tag{19}$$

and

$$\rho_g(t) = \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta p(\theta, t) \cos^2\theta. \tag{20}$$

Their sum is one, as it should be, if $p(\theta, t)$ is normalized to one. From (12) one can derive an equation for the time derivative of $\rho_1(t)$ and $\rho_0(t)$ by multiplying (12) by $\sin^2\theta$ and by $\cos^2\theta$, respectively, and integrating the result over θ. It gives,

$$\dot{\rho}_e = -\frac{\Omega}{2} \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta \sin^2\theta \frac{\partial p}{\partial \theta} - \gamma \left(\int_{-\pi/2}^{\pi/2} \mathrm{d}\theta p(\theta, t) \sin^4\theta \right) \tag{21}$$

and

$$\dot{\rho}_g = -\frac{\Omega}{2} \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta \cos^2\theta \frac{\partial p}{\partial \theta} + \gamma \left(\int_{-\pi/2}^{\pi/2} \mathrm{d}\theta p(\theta, t) \sin^4\theta \right). \tag{22}$$

In the r.h.s of the rate equations (21) and (22), the first term, proportional to the Rabi frequency Ω, describes the effect of the Rabi oscillations, whereas the second term, proportional to γ, displays the effect of the quantum jumps responsible for the *observable* photoemission. The integrand of this latter term indicates that the intensity of the spontaneous-emission field is proportional to $\sin^4\theta$, see below Eq. (24).

At this stage we emphasize that the right-hand sides of Eqs. (21) and (22) represent the new history beginning at each step as $p(\theta, t)$ includes both the fluctuations due to the quantum jumps and the streaming term.

4.2 Statistics of photoemission

Due to the Markovian nature of the function $\theta(t)$, and its behavior in the two steps labeled (1) and (2) in Sect. 3, we showed in Sect. 3.3.2 [4] that the probability distribution of the time interval between successive emission of photons, is given by the expression

$$\ell(\tau) = \lambda(\tau) e^{-\int_0^\tau \lambda(t)\, dt}, \tag{23}$$

where $\lambda(t)$ is the so-called density (or intensity) of the nonstationary point process formed by the times of photoemission, which depends on $\theta(t)$ only, not on the past values of θ.

From Eq. (21) we deduce this intensity of the point process at time t, conditionally on the knowledge of $\theta(t)$,

$$\lambda(t) = \gamma \sin^4 \theta(t). \tag{24}$$

This result, which is not so obvious, is of prime importance to derive the statistical properties of the radiated field. In this relation, the exponent 4 comes from two conditions, one that the atom is in the excited state, and the other one that it emits an observable photon. One may understand this by using the following argument. The probability for the atom to be in the excited state is $P(e) = \gamma \sin^2 \theta$. This is the sum of two terms, one is the joint probability that the atom is in the excited state AND the event "one observable photon" is emitted, $P(e, 1\,ph)$. The other term, $P(e, 0\,ph)$, is the joint probability that the atom is in the excited state AND "zero observable photon" is emitted. We have

$$P(e) = P(e, 1\,ph) + P(e, 0\,ph). \tag{25}$$

The conditional probability that one observable photon is emitted if the atom is in the excited state is given by

$$P(1\,ph|e) = \sin^2 \theta \tag{26}$$

as it yields the relation

$$P(e, 1ph) = P(1\,ph|e)P(e) = \gamma sin^4(\theta), \tag{27}$$

in agreement with Eq. (24). The two possibilities of emitting one detectable photon or zero at each quantum jump, are at the heart of the link we make below in Sect. 6 with the Everett's many-worlds interpretation.

Assuming an observable photon is emitted at time $t = 0$, the atom undergoes Rabi oscillations in between two successive emission times, which gives $\theta(t) = \Omega t/2$. Therefore, the interemission time distribution for an atom driven by a resonant pump is

$$\ell(\tau) = \gamma \sin^4\left(\frac{\Omega}{2}\tau\right) e^{-\gamma \int_0^\tau \mathrm{d}t \sin^4(\frac{\Omega}{2}t)}, \tag{28}$$

which gives $\int_0^\infty \ell(\tau)\, d\tau = 1$, as expected. The result is shown in Fig. 2 for large, middle, and small values of the ratio Ω/γ. Our curves in solid lines are compared to those derived in [2–6] by the picture of the dressed-atom radiative cascade. For the case of weak damping, or a strong input field, $\gamma \ll \Omega$, the two methods agree approximately, see Fig. 2-a. In this case, the mean interemission time is of order $1/\gamma$, which says that the atom undergoes many Rabi oscillations in between two successive emissions, therefore the large damping time drives the emission.

However, our results progressively differentiate (from the radiative cascade ones) as the ratio γ/Ω increases, until they noticeably differ, as shown in Fig. 2-c. In the latter case ($\gamma \gg \Omega$), an analytical expression of width of $\ell(\tau)$ can also be derived. Our K-derivation gives a mean delay between successive photons of order

$$\tau_K = \left(\Omega^4 \gamma\right)^{-1/5} = \frac{1}{\Omega}\left(\frac{\gamma}{\Omega}\right)^{1/5},$$

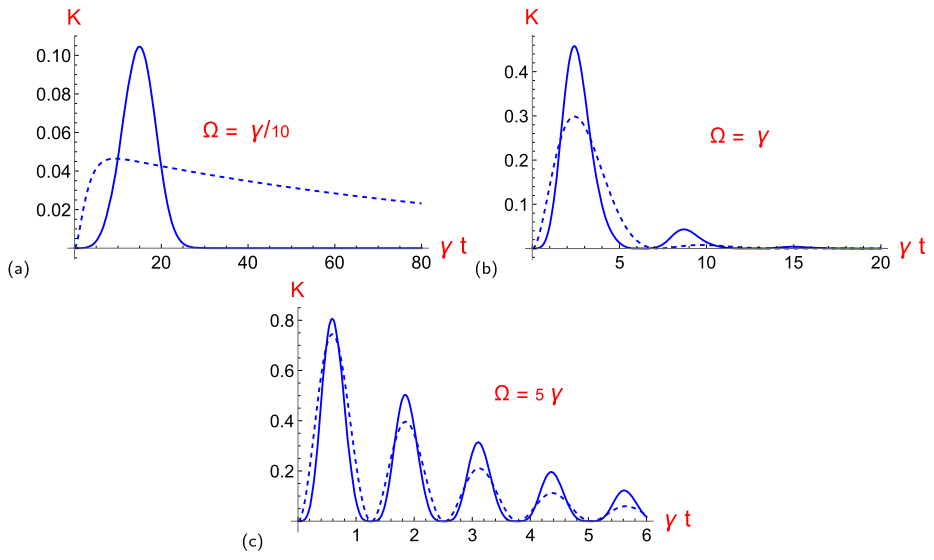

Figure 2 Probability of emission of the next fluorescence photon at time t after an emission at time 0, denoted $\ell(\tau)$ in the text. Solid lines for our Kolmogorov model, dashed lines for the radiative cascade description, Eq. (4) in Ref. [2], or Eq. (2.14) in Ref. [6]. (**a**) strong-damping regime, $\gamma = 10\Omega$. (**b**) $\gamma = \Omega$. (**c**) strong-pumping regime, $\gamma = 0.2\Omega$. The characteristic times of the distributions approximately agree in (**c**), whereas they noticeably diverge in (**a**), see a possible explanation in the text. Our coefficient γ corresponds to Γ defined in [6] and [2]

which is mainly of order of the Rabi oscillation time. This seems reasonable since the short time $1/\gamma$ makes a large probability of emission as soon as the atom is driven in its excited state via the Rabi oscillation. In summary, we find that the larger time drives the emission of photons, in the two limits.

However, the authors of Refs [2–6] find a very wide and flat interemission distribution in this limit, with mean interemission time

$$\tau_Q = \gamma/\Omega^2 = \frac{1}{\Omega}\left(\frac{\gamma}{\Omega}\right),$$

a time scale much longer than the Rabi period. A possible explanation for this difficult to understand result, is the following. The description of fluorescence as a radiative cascade in the dressed-atom picture starts with the master equation 2.1 in Ref. [6]. This picture is very evocative, but unfortunately very difficult to solve rigorously. Indeed, solving this equation for the reduced density matrix of the system atom + laser field (after tracing over the bath) leads to an infinite number of coupled equations describing what happens in each multiplicity correlated with its two adjacent ones (see Eqs. (2.2) of [6]). Hence, the authors make an uncontrolled simplification leading to their Eq. (2.3), which amounts to forgeting the feeding process schematized by the waving arrow joining each multiplicity with the upper one in their diagram (Fig. 1 of [2] and [6]). By doing this, the authors succeed in deriving simple analytical expressions for the statistical behavior of the fluorescent atom that hopefully agree with experiments as quoted in [2]. However, we conjecture that the above-mentioned simplifications could be especially questionable in the case of large damping where precisely the feeding of each multiplicity is expected to play a noticeable role and then cannot be neglected. In our approach we include the dissipation and refeed-

ing, by using tractable calculations. We hope this discrepancy will be elucidated by an experiment.

4.3 Fluorescence spectrum

The Markovian property of the fluorescence was used in our paper [4] to derive the interemission time distribution $\ell(\tau)$ and the correlation function of the fluorescence field

$$\mathcal{C}(\tau) = \gamma^2 \left\langle \sin^2\theta_0 \sin^2\theta(\tau) e^{2i(\varphi_0 - \varphi(\tau)} \right\rangle e^{-i\omega_L \tau}, \tag{29}$$

where the quantity inside the brackets must be weighted by the probability $p(\theta, \varphi, t)$ with i.c. θ_0, φ_0. We briefly mentioned in Sect. 3 that the phase φ changes abruptly at each jump occurring at times $t_j, t_{j+1}, \ldots t_{j+n}, \ldots$. Assuming that the phase differences $(\varphi_{t_{j+1}} - \varphi(j))$ are independent random variables, uniformly distributed in an interval $(-\delta\varphi, +\delta\varphi)$, the correlation function vanishes if the two times $(0, \tau)$ are separated by one (or more) quantum jump. Within this frame, we obtain the simple expression

$$\begin{aligned} \mathcal{C}(\tau) = e^{-i\omega_L \tau} \int_0^\pi \mathrm{d}\theta_0 p_{st}(\theta_0) \sin^2\theta_0 \sin^2(\theta_0 + \Omega t/2) \\ \times e^{-\gamma \int_0^\tau \mathrm{d}t \sin^4(\theta_0 + \Omega t/2)}, \end{aligned} \tag{30}$$

where the exponential

$$P^{(no)}(\tau, \theta_0) = e^{-\gamma \int_0^\tau \mathrm{d}t \sin^4(\theta_0 + \Omega b t/2)} \tag{31}$$

implements the constraint that zero photons are emitted in the time interval $[0, \tau]$. The interested reader can find the proof in Sect. 3.3.4 of [4] and the corresponding figures.

Here, we show another example of the shift spectrum of the EM field radiated by the pumped atom,

$$S(\omega - \omega_L) = 2Re\left[\int_0^\infty \mathrm{d}\tau \mathcal{C}(\tau)\right], \tag{32}$$

for large pumping, see Fig. 3. This three-peaked spectrum displays two sidebands, with amplitude about one third of the central peak one. We emphasize that our results agree with those deduced by usual quantum theory (Bloch equations and dressed-atom formalism).

Let us precise that the above expression (30) has been derived by using the Markovian property of the emission process having a density $\lambda(t)$ deduced from the K-equation. A direct calculation from the solution of our K-equation is also possible. The two methods give slightly different spectra (see Fig. 5 of [1]). This difference comes from the fact that Eq. (30) is for correlation function (or spectra) in the universe of the observer, although the direct method yields the same quantities averaged over all universes, see Sect. 6.

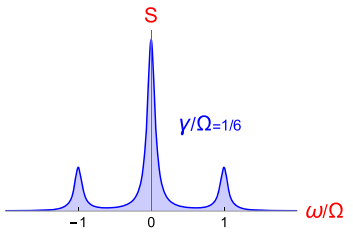

Figure 3 Three-peak spectrum versus the shifted frequency $\omega - \omega_L$, for small damping, $\gamma/\Omega = 1/6$

5 Irreversibility of quantum jumps

In the mathematical literature, irreversibility and reversibility are defined by reference to the equations of motion. For instance, Newtonian mechanics is well known to be reversible because, by inverting velocities and keeping the same positions, the trajectories of a set of interacting particles will trace back exactly their history. Seemingly, this property was already known to Newton himself: he spared computing work by calculating the trajectories of a mass around a center by inverting the speed at the apex to obtain the next part of the orbit. This definition of reversibility does not help much in real life because it cannot be used "practically". Consider for instance fluctuations in a turbulent fluid. One cannot reverse the speeds of all molecules at some time to check if the turbulent flow is in a state of reversible dynamics or not. Therefore, another definition of reversibility should be used to have measurable consequences. This was done by one of us [14]. In this paper, the idea was introduced that by analyzing some time correlation functions one can decide if a fluctuating signal is invariant or not under time reversal.

For a stationary (in time) random function, this excludes pair correlations of the same observable, like $\langle F(t)F(t+\tau)\rangle$, because this is an even function of τ, invariant under the exchange of τ and $-\tau$. When picking-up *different* random functions at time t and $(t+\tau)$, like

$$C_{FG}(\tau) = \big\langle F(t)G(t+\tau)\big\rangle$$

this symmetry $(\tau, -\tau)$ is no longer guaranteed, in general. Various examples of invariance breaking under time reversal are given in ref. [14]. As pointed out there, generally speaking this invariance is absent in out-of-equilibrium systems like a model of shear flow or turbulent flows, which are then irreversible. On the contrary, equilibrium fluctuations have the very special property of time-reversal symmetry, as had been shown by Onsager.

Let us apply this method to investigate whether the fluorescent intensity is reversible or not under time reversal, by using the following "test function"

$$\Psi_{1,2}(\tau) = \big\langle \lambda(t)\lambda^2(t+\tau) - \lambda^2(t)\lambda(t+\tau)\big\rangle, \tag{33}$$

where $\lambda(t)$ is the intensity of emitted fluorescence light at time t, a stationary random function defined in Eq. (24). The expression (33) is a test function because it is exactly zero if the signal is invariant under time reversal.

Let us consider the first term of Eq. (33)

$$C_{I,I^2} = \big\langle \lambda(t)\lambda^2(t+\tau)\big\rangle. \tag{34}$$

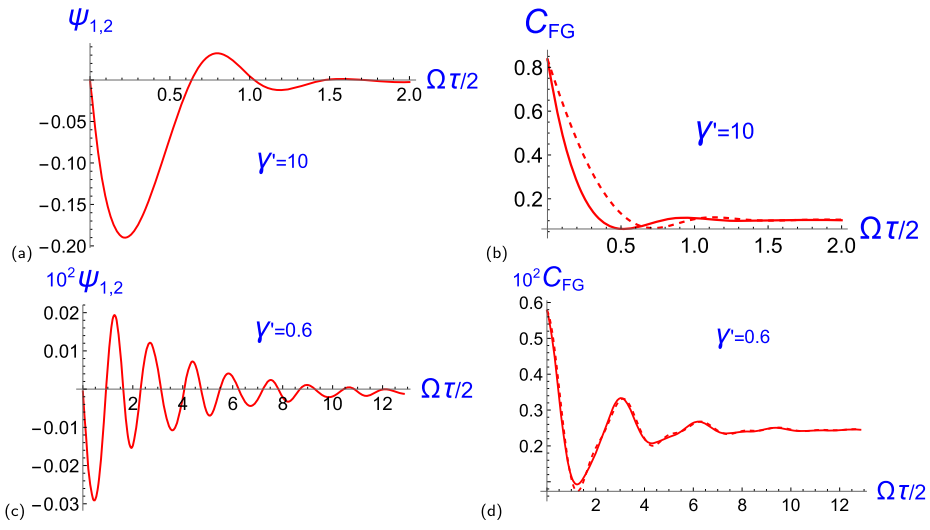

Figure 4 Test function $\Psi_{1,2}(\tau)$ defined in Eq. (33). (**a**) for $\gamma' = 10$ and (**c**) for $\gamma' = 0.6$. The role of the parameter $\gamma' = 2\gamma/\Omega$ appears when comparing (**b**) and (**d**) that display the two intercorrelation functions $C_{FG} = \mathcal{C}_{I,I^2}(\tau)$ and $C_{GF} = \mathcal{C}_{I^2,I}(\tau)$. For strong damping the irreversible character is clearly visible in (**b**) where the solid and dashed curves are well separated. In (**c**) and (**d**) the damping is weaker, and so is the ratio Ψ_{12}/C_{FG}, see text

The phase angle φ does not appear in the latter expression, which involves a product of intensities at a given time, insensitive to phase differences of the quantum states. Let us derive the test function from $\mathcal{C}_{I,I^2}(\tau)$ the K-equation solution (50) which yields a result averaged over all universes, see Sect. 6. The conditional probability $p(\theta, \tau|\theta_0, 0)$ allows us to calculate

$$\mathcal{C}_{I,I^2}(\tau) = \gamma^3 \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta_0 p_{st}(\theta_0) \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta p(\theta, \tau|\theta_0, 0) \sin^4(\theta_0) \sin^8\big(\theta(\tau)\big), \tag{35}$$

where the initial condition is $p(\theta, 0) = \delta(\sin(\theta - \theta_0))$ and $p_{st}(\theta_0)$ is the steady solution of Kolmogorov equation computed in Sect. 4.1.

Numerically, one finds a clear proof of the irreversible character of the two-level atom fluorescence. The test function is shown in Figs. 4(a) and (c) for large and small damping rates, respectively. In these figures, the difference $\Psi_{1,2} = C_{FG} - C_{GF}$, where $F = I(t)$, $G = I(t)^2$, displays damped oscillations in a time interval of order of the correlation time of the fluorescence field, which is the width of the correlation function $\mathcal{C}(\tau)$. For small damping the difference between C_{FG} and C_{GF} is hardly visible on curve (d) of Fig. 4 because it is small, with a ratio $\Psi_{1,2}/C_{FG}$ of order 6 per cent. This ratio becomes about 55 per cent for $\gamma' = 10$ (Figs. 4(a) and (b)). The important result is that this ratio increases as γ' increases, as expected, since the irreversible character of the spontaneous emission comes from quantum jumps (step (2) of the process described in Sect. 3).

6 Interpretation by Everett's theory of many-worlds

Let us explain how Everett's ideas are appropriate to understand the statistical effects observed in fluorescence, although Everett's theory of quantum physics [15] is sometimes considered as philosophical speculations without connection with real physics. When applied to the fluorescence by a single atom, the existence of many-worlds relies on the following image: each time a photon is emitted, the universe splits in two. One is the universe

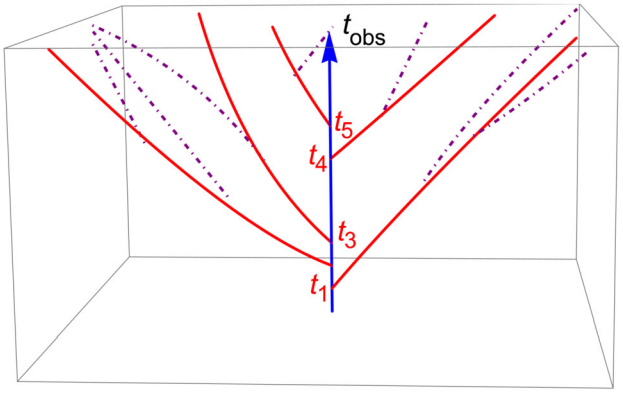

Figure 5 Schema of the possible trajectories of the atom emitting photons at times t_i, $\{i, 1, 5\}$ in the universe of the observer. The vertical blue line with an arrow is the trajectory seen by the observer, where the atom makes Rabi oscillations between t_i and t_{i+1}. The solid red lines stemming from each t_i, illustrate the successive splitting of the observer trajectory (universe) in two parts. On a red trajectory (virtual for the observer) no photon is emitted at t_i, but Rabi oscillations persist, until a photon is emitted in this universe. This occurs at the crossing points of red curves with purple dot-dashed curves. At these crossing points the "virtual red trajectory" splits into two parts, one (red) with an emitted photon and another one (purple) with no photon emitted

of the observer where the photon has been emitted and can be detected by him, the other one is a new universe created at this time (virtual for the observer), in which no photon is emitted. In this new universe the atom continues to undergo Rabi oscillations independently of what happens in the all others created before or after it. Each universe differs from the others by the time of emission of the photons. A 3D schema illustrating a possible set of trajectories coming from successive emission times t_i, is drawn in Fig. 5, see the caption, with the aim of illustrating that the various universes do not overlap.

By different universes one implies two related things. First, the histories of the two universes are a priori different after the emission event. This does not imply a large difference of course between the two universes because their initial condition at the instant of the emission are almost the same, but for the absence or presence of a single photon. Mathematically, the two universes are separated because their density matrices do not overlap: in one of the universes the photon number for the emitted photon is one, although it is zero in the other. Therefore, one can define in each universe a density matrix that will evolve in the future without any relationship with the density matrix of the other universe. Actually, the emission of photons occurs in a very short time, of the order of the period of the atomic motion, which is also the period of the EM waves emitted by the atom in its excited state. Therefore, there is a continuous emission of photons and so a continuous creation of new Everett's universes. In the case of fluorescence, what happens in all universes can be described only statistically, the statistics being carried over all universes existing at a given time. This defines a kind of super statistics because probability distributions are defined themselves over an object with a statistical meaning, namely the density matrix for the quantum state in the universe under consideration. In the case of a pumped two-level atom, this density matrix depends on the angle θ and the phase φ, so that the probability distribution is a probability depending on these two variables only. Our classical K-equation describing the dynamics of this random process takes into account all the

various states existing in different Everett universes that cannot interfere with each other because the nondiagonal elements of the density matrix of the full system are zero.

Contrary to other theories of fluorescence of a single atom, such a statistical theory has a built-in statistical structure that is, we believe, necessary to describe the randomness of the emission process. Such a randomness is intrinsic to the emission process, and it represents a bifurcation from one Everett universe to two, every time a photon is emitted. Let us note that from an experimental point of view, it would be impossible to make averages over all possible universes, because of the lack of overlap of the density matrices attached to the different universes. Therefore, one is practically restricted to making time averages in the universe where the observer lies, which poses the question of ergodicity: are these two averages identical? The case of a pumped atom could be interesting to investigate from the point of view of ergodic theory.

7 Conclusion

To conclude, our theory for the photo-emission by an atom continuously excited, relies on a single assumption, namely the smallness of the coupling between the EM field and the electrons in the atom. Because of this smallness the transition (or quantum jump) is quasi instantaneous compared to the duration of any other relevant processes like the Rabi oscillations and the intervals between two photo-emissions. Thanks to this difference of time scales we use the Kolmogorov equations to describe the statistics of the fluorescence without any supplementary assumption. This shows that a mathematical structure may emerge for describing a given problem by looking at the pertinent limit for the parameters involved.

We emphasize that Kolmogorov like equations are the only correct way to represent the dynamics of a process of finite jumps at a given rate of occurence, although Lindblad equation has been mostly used [16]-[17]. Contrary to our approach, Lindblad inspired analysis of the random process of emission of photons by an atom derive a time dependent differential equation for the reduced density matrix of the atom, with a damping coefficient representing the effect of the random emission. On time scales much longer than the period of the emitted photon we do not believe that such a deterministic equation can be derived from the basic equations of the problem, namely from the equation for the full density matrix (atom plus EM field) because the emission of photon (which is random and almost instantaneous as shown by Dirac) brings a change of the atomic state of *finite amplitude with a finite rate*. Such finite change of the atomic state is incompatible with the use of a Lindblad equation which is appropriate for describing only continuous and differentiable changes of the reduced density matrix. The description of quantum jumps requires to "put a probability on the density matrix" (in J. Ginibre's words) that leads to derive a Kolmogorov-like equation for this probability.

Kolmogorov approach of dynamical problems has been used forever in theoretical description of random processes including rare events with finite amplitude (named "stochastically determined process" by the author [8]. But Kolmogorov approach has not been yet applied to the class of problems where such randomness shows up in quantum phenomena. We have had the good luck of working on this class of problems with Jean Ginibre to whom we remain very indebted. Unfortunately we cannot thank him now for his kindness and help until the very end of a life devoted to science.

Appendix: Exact solution of the K-equation

Let us introduce the auxiliary function

$$b(t) = \int_{-\pi/2}^{\pi/2} \mathrm{d}\theta' p(\theta', t) \sin^2 \theta'. \tag{36}$$

The equation we are trying to solve becomes

$$\partial_t p + \frac{\Omega}{2} \partial_\theta p + \gamma \sin^2 \theta p = \gamma \delta(\sin\theta) b(t). \tag{37}$$

Let us take $\frac{2}{\Omega}$ as the unit of time and introduce the dimensionless parameter $\gamma' = \frac{2\gamma}{\Omega}$. Equation (37) becomes

$$\partial_t p + \partial_\theta p = g(\theta, t) - f(\theta) p \tag{38}$$

with

$$g(\theta, t) = \gamma' \delta(\sin\theta) b(t), \tag{39}$$

where $b(t)$ is still given by Eq. (36) and

$$f(\theta) = \gamma' \sin^2 \theta. \tag{40}$$

Let us derive from Eq. (38) the differential equation satisfied by the function $h(\theta, t) = p(\theta + t, t)$, which takes the form

$$\partial_t h(\theta, t) = g(\theta + t, t) - f(\theta + t)) h(\theta, t). \tag{41}$$

This can be solved as an initial value problem as follows. Take $s(\theta, t) = h(\theta, t) e^{\int_0^t \mathrm{d}t' f(\theta + t')}$. Therefore, $s(\theta, 0) = h(\theta, 0)$. The auxiliary function $s(\theta, t)$ is a solution of

$$\partial_t s = g(\theta + t, t) e^{\int_0^t \mathrm{d}t' f(\theta + t')}.$$

This has the solution

$$s(\theta, t) = s(\theta, 0) + \int_0^t \mathrm{d}t' g(\theta + t', t') e^{\int_0^{t'} \mathrm{d}t'' f(\theta + t'')}.$$

The equivalent result for the function $h(\theta, t)$ reads

$$h(\theta, t) = h(\theta, 0) e^{-\int_0^t \mathrm{d}t' f(\theta + t')} + \int_0^t \mathrm{d}t' g(\theta + t', t') exp\left(\int_0^{t'} \mathrm{d}t'' f(\theta + t'') - \int_0^t \mathrm{d}t'' f(\theta + t'') \right).$$

Tracing back the path from this explicit solution to the original equation, one finds the general solution of Eq. (38):

$$p(\theta, t) = p(\theta - t, 0) \alpha(\theta, t) + \int_0^t \mathrm{d}t' \alpha(\theta, t') g(\theta - t', t - t'), \tag{42}$$

where

$$\alpha(\theta,t) = e^{-\int_0^t \mathrm{d}t' f(\theta - t')}, \tag{43}$$

and $g(\cdot)$ is given by Eq. (39). Multiplying both sides of this equation by $f(\theta) = \gamma' \sin^2\theta$ and integrating the result over one period for θ, one finds the following Fredholm integral equation for $b(t)$,

$$b(t) = m(t) + \int_0^t \mathrm{d}t' b(t') l(t - t'). \tag{44}$$

In (44) we have

$$m(t) = \int_p \mathrm{d}\theta p(\theta, 0) f(\theta + t)\alpha(\theta + t, t), \tag{45}$$

where p stands for the period of the function f (here $p = \pi$), and

$$l(t) = f(t)\alpha(t), \tag{46}$$

where $\alpha(t)$ is the reduction to $\theta = t$ of the function of two variables $\alpha(\theta, t)$ (hopefully no confusion will arise from the use of the same notation, α for $\alpha(\theta, t)$ and $\alpha(t, t) = \alpha(t)$):

$$\alpha(t) = \alpha(t, t) = e^{-\int_0^t \mathrm{d}t' f(t')}. \tag{47}$$

Note the relation

$$l(t) = -\partial_t \alpha(t). \tag{48}$$

For $m(t)$ given, $b(t)$ can be derived from Eq. (44) either by iterations or by Laplace transforming both sides. In the numerics, we use the iteration with the initial condition

$$p(\theta, t = 0) = \delta(\theta - \theta_0), \tag{49}$$

in view of the derivation of correlation functions, see Sects. 4.3 and 5. It follows that any mean value calculated by using the probability (42) with initial condition (49), is actually a conditional average and should depend on the parameter θ_0 and should be labeled $b(t, \theta_0)$, as illustrated in Fig. 2 of [1]. With i.c. (49), the solution of the K-equation becomes the conditional probability,

$$p(\theta, t|\theta_0, 0) = \delta(\theta - t - \theta_0)\alpha(\theta_0, t) + \int_0^t \mathrm{d}t' \alpha(\theta, t') g(\theta - t', t - t'), \tag{50}$$

where $g(\theta, t)$ is given by Eq. (39).

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Declarations

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