Linear Response and Fluctuation Dissipation Theorem for non-Poissonian Renewal Processes.

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Abstract. - The Continuous Time Random Walk (CTRW) formalism is used to model the non-Poisson relaxation of a system response to perturbation. Two mechanisms to perturb the system are analyzed: a first in which the perturbation, seen as a potential gradient, simply introduces a bias in the hopping probability of the walker from on site to the other but leaves unchanged the occurrence times of the attempted jumps ("events") and a second in which the occurrence times of the events are perturbed. The system response is calculated analytically in both cases in a non-ergodic condition, i.e. for a diverging first moment in time. Two different Fluctuation-Dissipation Theorems (FDTs), one for each kind of mechanism, are derived and discussed.

Introduction. – Complex physical systems provide a number of challenges that cannot be addressed with the traditional approaches of equilibrium statistical physics. One phenomenon that reveals a striking departure from traditional statistical mechanics is Blinking Quantum Dots (BQD) [1]. BQD systems jump between two states, "light on" and "light off", thereby being a realization of dichotomous fluctuations and this two-state model of BQD proves to be sufficient to satisfactorily explain the observed data. There is consensus that these fluctuations are simultaneously non-Poissonian and renewal [2, 3]. This property makes them a paradigmatic example of weak ergodicity breakdown [4]. The Kubo-Anderson spectral diffusion theory [5] is violated [6], and a new theoretical approach has to be created to study the emission and absorption processes [7].

Note that the theoretical approach behind the work of Refs. [4, 6, 7] is inspired to the Continuous Time Random Walk (CTRW) theory [8]. However, it is not yet clear if alternative approaches based on the adoption of a Hamiltonian prescription are possible. Under the simplifying assumption that the "light on" and the "light off" states have the same waiting time distribution $\psi(t)$ the theoretical CTRW prescriptions are equivalent to the following Generalized Master Equation (GME):

$$\frac{d}{dt}\mathbf{p}(t) = -\int_0^t \Phi(t-t')\mathbf{K}.\mathbf{p}(t')dt', \qquad (1)$$

where $\mathbf{p}(t)$ is a two-dimensional vector, whose components $p_+(t)$ and $p_-(t)$ denote the probability for the system to be in the "on" and "off" state, respectively. The matrix $\mathbf{K}$ is defined by

$$\mathbf{K} = \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \qquad (2)$$

and the memory kernel function $\Phi(t)$ is connected to $\psi(t)$ by means of their Laplace Transform as: $\hat{\Phi}(u) = u\,\hat{\psi}(u)/(1-\hat{\psi}(u))$. The formal equivalence between CTRW and GME was originally established by Kenkre et al. [9] and extended to include renewal aging by Allegrini, Aquino et al. [10, 11].

Is the GME-CTRW equivalence substantial as well as merely formal? This is a question of great importance for statistical physics. It is well known [12] that starting from a Hamiltonian description of a system plus environment, that a Liouville equation formalism can be used to project the dynamics onto that of the system alone. This projected representation is equivalent to the GME.

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We are convinced that the question of the physical equivalence between GME and CTRW is still not yet settled, and in this letter, by deriving an exact linear response result, using only CTRW arguments, a step towards solving this controversy is taken. We notice in fact that the Fluctuation-Dissipation Theorem (FDT) of the first kind [13], which has a Hamiltonian origin, can be generalized [14], within an approach compatible with a Hamiltonian origin, to a non-stationary condition with the following expression:

$$R_{AB}(t,t') = \beta \frac{d}{dt'} \langle A(t)B(t') \rangle \qquad t > t', \tag{3}$$

connecting the response function $R_{AB}(t,t')$ to the correlation function $\langle A(t)B(t')\rangle$.

In Eq.(3) A denotes the variable of interest, B is the variable of the system through which the external perturbation affects its dynamics and β the inverse temperature.

To adapt this theoretical prediction to the BQD process, we assume $A = B = \xi(t)$, with $\xi = +1$ and $\xi = -1$ in the state $|+\rangle$ and $|-\rangle$, respectively and $\beta = 1$. Consequently, the FDT attains the form

$$R(t,t') = \frac{d}{dt'} C(t,t'), \tag{4}$$

where

$$C(t,t') = \langle \xi(t)\xi(t') \rangle. \tag{5}$$

It is important to remark that the response of the two-state system, compatible with the CTRW approach, has been studied by Sokolov [15] as well as Sokolov and Klafter [16], with a treatment that, as we shall see, leads to the adoption of Eq. (4). Thus, we may conclude that the formal equivalence between CTRW and GME indeed corresponds to physical equivalence. However, we notice that the theoretical predictions of these authors all rest on the same assumption, i.e. that the perturbation seen as a potential gradient, simply introduces a bias in the hopping probability of the walker in moving between the sites while leaving unchanged the occurrence times of the attempted jumps. We name this scheme the “phenomenological approach”.

On the other hand, the experimental observation of the BQD phenomenon affords information on $\psi(t)$, which, as we shall see, depends on two parameters, μ and T. The power-law parameter μ is the complexity index and depends on the cooperative nature of the system. The parameter T determines the time scale at which the waiting time distribution $\psi(t)$ can be considered to be equivalent to an inverse power law with index μ. It is realistic to expect that the external perturbation has the effect of influencing these two parameters, and especially the parameter T, given the fact that μ has a cooperative origin, and consequently is expected to be the more robust of the two. To calculate the system's response to perturbation in this scheme is a difficult problem, and the aim of the present article is to describe a rigorous way to solve it, within the spirit of CTRW. In this letter by calculating analytically the response to a perturbation of $\psi(t)$ we obtain as main consequence the new FDT relation

$$R(t,t') = -\frac{d}{dt} C(t,t'). \tag{6}$$

Response and FDT within the Phenomenological Approach. – Let us consider the process of a BQD switching between “on” ($|+\rangle$) and “off” ($|-\rangle$) states. This process can be modeled as a random walk on a two sites lattice. At random times determined by the distribution $\psi(t)$ the walker attempts a jump to the other site, we call these attempts “events”. Whenever an event occurs the walker can either jump to the other site or remain in the same site with equal probability 1/2. Within the “phenomenological approach” the perturbation $f(t)$ simply introduces a bias in these probabilities while leaving the statistics of the events, i.e. $\psi(t)$, unchanged.

Thus, under the influence of the perturbation $f(t)$ the walker jumps to the other state or remains in the same state with probabilities respectively:

$$\begin{aligned} w_{\pm,\mp}(t) = Pr(\pm \to \mp) &= \frac{1 \pm \epsilon f(t)}{2} \\ w_{\pm,\pm}(t) = Pr(\pm \to \pm) &= \frac{1 \mp \epsilon f(t)}{2} \end{aligned} \tag{7}$$

As mentioned above, this case has already been solved in a GME approach in Ref. [15] and within a different approach in Ref. [17]. To the end of illustrating our method and showing its general validity we rederive the response in this case with a CTRW approach. Consider a random walk on a two-site lattice which starts at time $t = 0$ with equal occupation of the two sites. The occupation vector is denoted by $\mathbf{p}$ and the distribution of waiting times for the walker between the events by $\psi(t)$. We make the assumption that:

$$\psi(\tau) = \frac{\mu - 1}{T}(1 + \tau/T)^{-\mu}, \tag{8}$$

having in mind systems such as the BQDs whose blinking behavior is characterized by a residence times distribution which is a power law with diverging first moment (i.e. $1 < \mu < 2$). The occupation at time t will be determined by all the possible ways the walker can move in the interval of time $[0,t]$ between the sites. In the presence of the perturbation, turned on at time $t = 0$, a bias will occur. Let us divide all the trajectories in four groups that we indicate with g_{ij}, determined by the starting site i at time $t = 0$ and the arrival site j at time t, with $i,j = \pm$. The fraction of trajectories g_{ij} starting in site i at time $t = 0$ and switching to site j at time t is given by $A_{ij}(t)p_i(0)$, with:

$$\begin{aligned} A_{ij}(t) &= \sum_{n=0}^{\infty} \sum_{k_1,..k_n=\pm} \int_0^t dt_1 \psi(t_1) w_{i,k_1}(t_1) \cdots \\ &\cdot \int_{t_{n-1}}^t dt_n \psi(t_n - t_{n-1}) w_{k_n,j}(t_n) \Psi(t - t_n). \end{aligned} \tag{9}$$

The trajectories are then sequences of "laminar phases" between events in which the walker remains in the same site. The $\psi(t_j - t_{j-1})dt_j$ are therefore the probabilities of occurrence of an event at time $t = t_j$ without implying necessarily a change of site, while $\Psi(t)$ is just the survival probability, i.e. the probability of occurrence of no event in an interval of time of length t.

Let us focus on the group g_{++}: the trajectories belonging to this group, at time t, are again in the state $|+\rangle$, so they contribute only to the population of the first site at time t. Eqs. (7) show that, the first-order contribution to the response is obtained by keeping in the terms of the sum of Eq. (9) only one perturbed weight $w_{k_i,k_{i+1}}(t_{i+1})$ and leaving the unperturbed weight 1/2 in the remaining factors . This implies that for the trajectories in the group g_{++} characterized by the same set of n events, only the terms with the perturbed jump probabilities before the last laminar phase may give an overall nonzero contribution. In fact, consider trajectories with the same set of events (in the group g_{++}) , for every trajectory with a laminar phase weighted with the perturbed jumping probability of Eq.(7), there will be one trajectory identical in every respect apart from the following laminar phase being of the same length but of opposite sign (i.e. the walker is in the other site). These two trajectories according to Eq. (7), are weighted with a jumping probability with opposite sign and therefore their total contribution to first order in ε is null. The only surviving contribution to first order in ε will be the one with perturbed jumping probability before the last laminar phase, the latter being fixed by the group considered. The argument extends to a generic group g_{ij}, the total contribution of this group to the population of site j at time t is therefore given by $A_{ij}(t)p_i(0)$ with $A_{ij}(t) = A^0_{ij}(t) + A^\varepsilon_{ij}(t)$, that is an unperturbed plus a perturbed term. For the response we are interested only in the contribution of the perturbed part which at the first order, as explained above, simplifies to $-jA^\epsilon_L(t)p_i(0)$ with

$$\begin{aligned} A^\epsilon_L(t) &= \sum_{n=1}^{\infty}\int_0^t dt_1\psi(t_1)\cdots \\ &\cdot \int_{t_{n-1}}^t dt_n\psi(t_n - t_{n-1})\frac{\varepsilon}{2}f(t_n)\Psi(t-t_n) \end{aligned} \tag{10}$$

i.e. the contribution obtained by keeping a perturbed jumping probability only before the last laminar phase. Summarizing, the contribution of all the four groups of trajectories to the population vector at time t is:

$$\begin{aligned} g_{++} &\rightarrow -A^\epsilon_L(t)\begin{pmatrix} p_+(0) \\ 0 \end{pmatrix} \\ g_{+-} &\rightarrow A^\epsilon_L(t)\begin{pmatrix} 0 \\ p_+(0) \end{pmatrix} \\ g_{-+} &\rightarrow -A^\epsilon_L(t)\begin{pmatrix} p_-(0) \\ 0 \end{pmatrix} \\ g_{--} &\rightarrow A^\epsilon_L(t)\begin{pmatrix} 0 \\ p_-(0) \end{pmatrix} \end{aligned} \tag{11}$$

The response of the system is obtained by calculating the difference Σ of population between the two sites. From (11) one derives:

$$\Sigma(t) = p_-(t) - p_+(t) = 2A^\epsilon_L(t). \tag{12}$$

Using Eq. (10) it follows:

$$\Sigma(t) = \varepsilon\sum_{n=1}^{\infty} Re\left[\int_0^t dt_n f(t_n)\psi_n(t_n)\Psi(t-t_n)\right], \tag{13}$$

where $\psi_n(t_n)$ is just the n-times convolution of $\psi(t)$ and the sum runs from $n = 1$ since the $n = 0$ term, having no events, does not contain any term linear in ε. From Eq. (13) the response function turns out to be:

$$R(t,t') = \sum_{n=1}^{\infty}\psi_n(t')\Psi(t-t') = P(t')\Psi(t-t'). \tag{14}$$

The result of [17] and [15] is then recovered. The autocorrelation function for the unperturbed case is known to be equal to the function $\Psi(t,t')$, see [11], that is:

$$C(t,t') = \Psi(t,t') = \int_t^\infty dx\psi(x,t'), \tag{15}$$

where $\Psi(t,t')$ is the probability that, for fixed t' no events occurs until time $t > t'$, and

$$\psi(t,t') = \psi(t) + \int_0^{t'} dx\psi(t-x)P(x) \tag{16}$$

is the probability distribution that, for fixed t', the first next event occurs at time t. It follows that

$$\frac{d}{dt'}C(t,t') = \int_t^\infty dx\frac{d}{dt'}\psi(x,t'). \tag{17}$$

From Eq. (16)

$$\frac{d\psi(t,t')}{dt'} = \psi(t-t')P(t') \tag{18}$$

and then inserting this into Eq. (17) one obtains:

$$\frac{d}{dt'}C(t,t') = R(t,t'), \tag{19}$$

that is the general FDT derivable for a system perturbed from a canonical equilibrium.

Response and FDT in the dynamic approach. – Let us now consider a scheme in which the bias is due to a perturbation of the statistics of occurrence times of events. We name this approach "dynamic" because we have in mind a dynamic model generating the waiting time distribution of Eq. (8) through the equation of motion

$$\dot{y} = \alpha_0\, y^{\frac{\mu}{\mu-1}}, \tag{20}$$

with $\alpha_0 = (\mu-1)/T$, describing a particle moving on the interval $I = (0, 1]$ that, everytime it gets to 1, is re-injected back inside the interval I with a random initial condition. The arrival of the particle at the border generates the "events" that lead the random walk on the two-sites lattice. As explained in Ref. [17] the perturbation in this scheme changes the parameter T and therefore α_0 in the following way:

$$\alpha_{\pm}(t) = \alpha_0(1 \pm \varepsilon f(t)), \tag{21}$$

where the sign $\pm$ depends on the site at which the walker is residing. We remark that in this scheme, when an event occurs, the walker jumps to the other site or remains in the same site with unchanged probability equal to 1/2. The perturbation therefore modifies the unperturbed statistics of the events, i.e. $\psi(t)$ as given by Eq. (8), and introduces an "age dependence" in the statistics of the events breaking the time translation invariance of the renewal process. At first order in the perturbation (see Ref. [17]), $\psi(t)$ is changed into the following conditional probability density:

$$\begin{aligned}\psi_{\varepsilon}^{\pm}(\tau|t') &= \psi(\tau)[1 \pm \varepsilon f(t'+\tau)] \\ &\mp \varepsilon\frac{\mu-1}{T}\psi_{\mu+1}(\tau)\int_{t'}^{t'+\tau} dx f(x),\end{aligned} \tag{22}$$

i.e. $\psi_{\varepsilon}^{\pm}(\tau|t')d\tau$ is the probability that, after an event has occurred at time t', the following event occurs at $t = t'+\tau$. $\psi_{\mu+1}(t)$ is the same as Eq. (8) but with index $\mu+1$. We consider the case of harmonic perturbation, then Eq. (22) turns into:

$$\begin{aligned}\psi_{\varepsilon}^{\pm}(\tau|t') &= \psi(\tau)[1 \pm \varepsilon cos(\omega(t'+\tau))] \\ &\mp \varepsilon\frac{\mu-1}{T\omega}\psi_{\mu+1}(\tau)[sin(\omega(t'+\tau)) - sin(\omega t')].\end{aligned} \tag{23}$$

Keeping only the first term in Eq. (23) is equivalent to an unperturbed distribution of the events followed by a bias in the choice of the direction of motion similar to the phenomenological approach. The second term, besides assuring normalization breaks this equivalence entangling the perturbation with the dynamics. By adopting the same notation as in the phenomenological case, the contribution of the group g_{ij} to the population of the site j at time t, is $A_{ij}(t)p_i(0)$ with

$$\begin{aligned}A_{ij}(t) = &\sum_{n=0}^{\infty}\sum_{k_1,\ldots k_{n-1}=\pm}\int_0^t dt_1\psi_{\varepsilon}^{i}(t_1|0)\frac{1}{2}\cdots \\ &\cdot\int_{t_{n-1}}^{t} dt_n\psi_{\varepsilon}^{k_{n-1}}(t_n - t_{n-1}|t_{n-1})\frac{1}{2}\Psi_{\varepsilon}^{j}(t-t_n|t_n)\end{aligned}$$

where:

$$\Psi_{\varepsilon}^{\pm}(t-t_n|t_n) = \int_t^{\infty} dx\psi_{\varepsilon}^{\pm}(x-t_n|t_n) \tag{24}$$

is the conditional survival probability of remaining in the state $\pm$, with no events in the interval of time $t-t_n$, after an event occurred at time t_n. Let us rewrite the survival probability in the following way

$$\Psi_{\varepsilon}^{\pm}(t-t_n|t_n) = \Psi(t-t_n) \pm \varepsilon\Psi_{\varepsilon}(t-t_n|t_n) \tag{25}$$

with

$$\Psi_{\varepsilon}(t-t_n|t_n) = \Psi^1(t-t_n|t_n) - \Psi^2(t-t_n|t_n), \tag{26}$$

where, as can be deduced from Eqs. (24) and (23), $\Psi(t-t_n)$ is just the unperturbed survival probability while $\Psi^1(t-t_n|t_n)$ and $\Psi^2(t-t_n|t_n)$ are the conditional survival probabilities originating respectively from the first and the second term linear in ε in Eq. (23), that is:

$$\Psi^1(t-t_n|t_n) = Re[\int_t^{\infty} dx\psi(x-t_n)e^{i\omega x}] \tag{27}$$

and

$$\Psi^2(t-t_n|t_n) = \frac{\mu-1}{T\omega}Im[\int_t^{\infty} dx\psi_{\mu+1}(x-t_n)(e^{i\omega x}-e^{i\omega t_n})] \tag{28}$$

Adopting analogous considerations as those used for the phenomenological case, it can be inferred that the first order contribution to the population at time t is obtained considering only one laminar phase with the perturbed distribution of Eq. (23) while for the others keeping the unperturbed one. Also in this case one easily realizes that the contribution to the response comes only from those trajectories with perturbed last laminar phase, that is by replacing all the $\psi_{\varepsilon}^{k}(t_{k+1}-t_k|t_k)$ in Eq. (24) with the unperturbed probability $\psi(t_{k+1}-t_k)$ and considering only the contribution linear in ε coming from $\Psi_{\varepsilon}^{\pm}(t-t'|t')$. In this case therefore, the trajectories with no events between 0 and t will also give a contribution to first order in ε. Let us consider first only the contribution of the trajectories with at least one event, this contribution is the same for both the groups g_{ii} and g_{ij} and amounts to $jA_L^{\epsilon}(t)p_i(0)$, with:

$$A_L^{\epsilon}(t) = \frac{\varepsilon}{2}\sum_{n=1}^{\infty}\int_0^t dt_n\psi_n(t_n)\Psi_{\varepsilon}(t-t_n|t_n), \tag{29}$$

which applies to the probability vector as:

$$\begin{aligned}g_{++} &\rightarrow A_L^{\epsilon}(t)\begin{pmatrix} p_+(0) \\ 0 \end{pmatrix} \\ g_{+-} &\rightarrow -A_L^{\epsilon}(t)\begin{pmatrix} 0 \\ p_+(0) \end{pmatrix} \\ g_{-+} &\rightarrow A_L^{\epsilon}(t)\begin{pmatrix} p_-(0) \\ 0 \end{pmatrix} \\ g_{--} &\rightarrow -A_L^{\epsilon}(t)\begin{pmatrix} 0 \\ p_-(0) \end{pmatrix}.\end{aligned} \tag{30}$$

Then one has to add the contribution of the trajectories with no events which gives:

$$\varepsilon\begin{pmatrix} \Psi_{\varepsilon}(0|t)p_+(0) \\ -\Psi_{\varepsilon}(0|t)p_-(0) \end{pmatrix}. \tag{31}$$

In the end calculating $p_-(t) - p_+(t)$ one obtains:

$$\begin{aligned}\Sigma(t) &= 2A_L^{\epsilon}(t) + \Psi_{\varepsilon}(0|t) \\ &= \varepsilon\int_0^t dt'[\delta(t') + \sum_{n=1}^{\infty}\psi_n(t')]\Psi_{\varepsilon}(t-t'|t')\end{aligned} \tag{32}$$

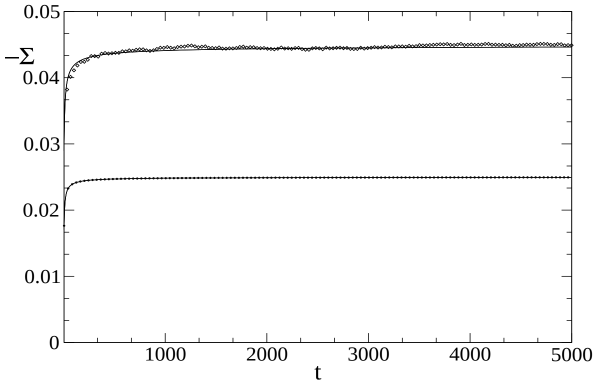

Fig. 1: Numerical simulation (dots) for $\Sigma(t)$ in response to a constant perturbation as compared to the inverse Laplace transform of the analytical expression Eq. (36) in the limit $\omega \to 0$. From top to bottom the curves refer to $\psi(t) = \frac{\mu-1}{T}(1+t/T)^{-\mu}$ with $\mu = 1.45$, $T = 1.6$ and $\mu = 1.25$, $T = 1.6$ ($\varepsilon = 0.1$).

Let us consider the contribution coming only from the first term in Eq. (26): the Laplace transform of its contribution to Eq. (32) is easily calculated:

$$\hat{\Sigma}_1(s) = \varepsilon Re[\frac{1}{s}\left(\frac{\hat{\psi}(-i\omega) - \hat{\psi}(s-i\omega)}{1-\hat{\psi}(s-i\omega)}\right)], \qquad (33)$$

which for a constant perturbation, i.e. in the limit $\omega \to 0$, gives the asymptotic value of epsilon. The contribution to Eq. (32) of the second term in Eq. (26) amounts to

$$\hat{\Sigma}_2(s) = -\varepsilon\frac{\mu-1}{T\omega} Im\left[\frac{1}{s(1-\hat{\psi}(s-i\omega))}\Big(\hat{\psi}_{\mu+1}(-i\omega) - \hat{\psi}_{\mu+1}(s-i\omega) - (1-\hat{\psi}_{\mu+1}(s))\Big)\right]. \qquad (34)$$

Considering that $\hat{\psi}_{\mu+1}(s) = 1 + Ts\hat{\psi}(s)/(1-\mu)$ and making a convenient simplification we get:

$$\hat{\Sigma}_2(s) = \varepsilon Im\left[\frac{(i\omega)\hat{\psi}(-i\omega) + (s-i\omega)\hat{\psi}(s-i\omega) - s\hat{\psi}(s)}{-s\omega(1-\hat{\psi}(s-i\omega))}\right]$$
$$= -\varepsilon Re\left[\frac{\hat{\psi}(-i\omega)-\hat{\psi}(s-i\omega)}{s(1-\hat{\psi}(s-i\omega))} + \frac{\hat{\psi}(s)-\hat{\psi}(s-i\omega)}{i\omega(1-\hat{\psi}(s-i\omega))}\right]. \qquad (35)$$

Taking the limit for $\omega \to 0$ carefully so as to obtain the case of a constant perturbation, and then taking the limit $s \to 0$ (i.e. $t \to \infty$), in Eq. (35), the final contribution amounts to $\varepsilon(-\mu)$. This contribution added to that of the first term given by (33) gives the value $\varepsilon(1-\mu)$. We confirm this result with a numerical simulation for the case of constant perturbation, the good agreement is shown in Fig. 1.

Remarkably, summing (35) and (33) and simplifying

$$\hat{\Sigma}(s) = \hat{\Sigma}_1(s) + \hat{\Sigma}_2(s) = \varepsilon Re[\frac{(\hat{\psi}(s-i\omega) - \hat{\psi}(s))}{i\omega(1-\hat{\psi}(s-i\omega))}], \qquad (36)$$

which is just the real part of the Laplace transform of

$$\varepsilon\int_0^t \psi(t,t')e^{i\omega t'}dt', \qquad (37)$$

where $\psi(t,t')$ is given by Eq. (16). This expression uniquely identifies $\psi(t,t')$ as the response function and is straightforwardly extended to a generic perturbation $f(t)$, simply by using its Fourier representation and following the same steps used to prove Eq. (37).

FDT in the dynamic case. We conclude that in the dynamic case the response to an external perturbation $f(t)$ can be written in the form

$$\Sigma(t) = \varepsilon\int_0^t dt' R(t,t')f(t') \qquad (38)$$

with $R(t,t') = \psi(t,t')$. It can be seen, by differentiating Eq. (15) respect to t, that in this case a different Fluctuation Dissipation Relation (FDR) is obtained, namely:

$$\frac{d}{dt}C(t,t') = -R(t,t'). \qquad (39)$$

Of course, if a stationary condition is reached, time translation invariance is fulfilled and the two FDRs are equivalent. In order to address the discussion of how general the new FDT may be, we point out that the same result has been recently obtained by Allegrini at al. [18] using a dynamical model different from that of Eqs. (20) and (21), fitting only the constraint of yielding the distribution of Eq. (8). On the basis of this we argue that Eq. (39) might be indeed a universal result, requiring only that the perturbation changes slightly the occurrence times of the events of the system, thereby producing a small change of the parameter T of Eq. (8).

Summarizing, in this letter we analyzed the problem of the response to perturbation of systems with slow relaxation properties described by non-Poissonian renewal processes with diverging characteristic time. We considered two different mechanism for perturbing the system. For the first one, which we termed phenomenological, we rederived the results obtained in Refs. [16,17] and a Fluctuation Dissipation Relation equal to the one that can be demonstrated to be valid for Hamiltonian systems perturbed out of canonical equilibrium [14]. For the second approach, which we termed dynamical and consider more realistic because it involved a perturbation of the statistics of the "events" generating the random walk process, we derived an exact analytical expression for the response, and showed that a different FDR, Eq. (6) is fulfilled.

Concluding Remarks. – We devote these concluding remarks to discussing the possible consequence of the main result of this article: Eq. (6). The first important result is that the equivalence between GME and CTRW not only is questionable, but is incorrect. This is expected to have significant consequences on the foundation of a theory for BQD phenomenon. Altough the response to external perturbation of single quantum dots within a Hamiltonian model has been addressed in several works (e.g. [19]), this has not been achieved in the BQD ''regime", which occurs for quantum dots embedded in a disordered environment. Our result indicates that the derivation of the BQD phenomenon from a Hamiltonian picture is not only difficult but impossible, thereby locating the intermittent fluorescence at the level of those emergent cooperative processes that determine reductionism failure.

We speculate here also a second possible consequence of the main result of this article. This is connected with the concept of an effective temperature such as was introduced into the physics of glassy systems. In 1997 the authors of Refs. [20,21], on the basis of earlier work [22] (see also [23]) argued that it is convenient to replace Eq. (3) with

$$R_{AB}(t,t') = \beta K(t,t') \frac{d}{dt'} \langle A(t)B(t') \rangle \quad t > t', \tag{40}$$

where the function $K(t,t')$ has the role of defining an effective temperature depending on the age of the system t'. The effective temperature has then the role of linking the out-of-equilibrium configurations occupied by the system during its relaxation at a given temperature, to equilibrium configurations relative to a different temperature. There has been an intense research activity in this direction, and we limit ourselves to quoting some papers representative of the experimental and theoretical work done in this direction [24–27], with controversial results including the report of no deviation from the FDT over several decade in frequency [26]. Is the generalization of FDT derived in this Letter with exact dynamic arguments compatible with the concept of an effective temperature as introduced in Eq. (40)? Consider Eqs. (15) and (16) from which the following relations with the survival probability:

$$\begin{aligned} -\frac{d\Psi(t,t')}{dt} &= \psi(t,t') \qquad (41) \\ \frac{d\Psi(t,t')}{dt'} &= P(t')\Psi(t-t'), \end{aligned}$$

are determined. Thus, the result of this letter can be expressed in the form of Eq. (40) (β=1, $A = B = \xi$) with

$$K(t,t') = \psi(t,t')[P(t')\Psi(t-t')]^{-1}. \tag{42}$$

The introduction of an effective temperature in this case has the role of linking our description to one compatible with the ordinary FDT obtained in Refs. [15,16], which in turn is directly linked to a Hamiltonian derivation.

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