Non-Poisson processes: regression to equilibrium versus equilibrium correlation functions

Paolo Allegrini[1], Paolo Grigolini[2,3,4], Luigi Palatella[5], Angelo Rosa[6], Bruce J. West[7]
[1] *INFM, unità di Como, via Valleggio 11, 22100 Como, Italy*
[2] *Center for Nonlinear Science, University of North Texas, P.O. Box 311427, Denton, Texas 76203-1427*
[3] *Dipartimento di Fisica dell'Università di Pisa and INFM, Via Buonarroti 2, 56127 Pisa, Italy*
[4] *Istituto dei Processi Chimico Fisici del CNR, Area della Ricerca di Pisa, Via G. Moruzzi 1, 56124 Pisa, Italy*
[5] *Dipartimento di Fisica and INFM, Center for Statistical Mechanics and Complexity, Università di Roma "La Sapienza", P.le A. Moro 2, 00185 Rome, Italy*
[6] *Institut de Mathématiques Bernoulli, Faculté des Sciences de Base, École Polytechique Fédérale de Lausanne, 1015 Lausanne, Switzerland and*
[7] *Mathematics Division, Army Research Office, Research Triangle Park, NC 27709, USA*

We study the response to perturbation of non-Poisson dichotomous fluctuations that generate super-diffusion. We adopt the Liouville perspective and with it a quantum-like approach based on splitting the density distribution into a symmetric and an anti-symmetric component. To fit the equilibrium condition behind the stationary correlation function, we study the time evolution of the anti-symmetric component, while keeping the symmetric component at equilibrium. For any realistic form of perturbed distribution density we expect a breakdown of the Onsager principle, namely, of the property that the subsequent regression of the perturbation to equilibrium is identical to the corresponding equilibrium correlation function. We find the directions to follow for the calculation of higher-order correlation functions, an unsettled problem, which has been addressed in the past by means of approximations yielding quite different physical effects.

PACS numbers:

I. INTRODUCTION

Complex physical systems typically have both nonlinear dynamical and stochastic components, with neither one dominating. The response of such systems to external perturbations determines the measurable characteristics of phenomena from the beating of the human heart to the relaxation of stressed polymers. What distinguishes such complex phenomena from processes successfully studied using equilibrium statistical mechanics is how these systems internalize and respond to environmental changes. Consequently, it is of broad interest to determine which of the prescriptions from equilibrium statistical physics is still applicable to complex dynamical phenomena and which are not. Herein we address the breakdown of one of these fundamental relations, that being, the Onsager Principle. In the case of ordinary statistical mechanics an exhaustive treatment of the relaxation of perturbations to equilibrium can be found in Ref. [1]. Let us consider as a prototype of ordinary statistical mechanics the case when the stochastic variable under study $\xi(t)$ is described by the linear Langevin equation

$$\frac{d\xi}{dt} = -\gamma\xi(t) + \eta(t), \tag{1}$$

where the random driving force $\eta(t)$ is white noise. Let us imagine that $\xi(t)$ is the velocity of a particle with unit mass and a given electrical charge. Furthermore, we assume that this system reaches the condition of equilibrium, and at a given time $t = 0$, we apply an electrical field $E(t)$. The external field $E(t)$ is an arbitrary function of time, fitting the condition that $E(t) = 0$, for $t < 0$. The adoption of linear response theory yields the prescription for the mean response of the system to the external field

$$\langle\xi(t)\rangle = \int_0^t \Phi_\xi(t')E(t-t')dt', \tag{2}$$

where $\Phi_\xi(t)$ is the equilibrium correlation function of ξ. In this familiar case, of dissipative Brownian motion, the autocorrelation function of the particle velocity is the exponential $\exp(-\gamma t)$.

There are two limiting cases of time dependence of the perturbation: (a) the electric field is proportional to the Heaviside step function, $E(t) = K\Theta(t)$, and is therefore a constant field after it is turned on at $t = 0$; (b) the electric field is proportional to the Dirac delta function, $E(t) = K\delta(t)$, and is consequently an initial pulse that perturbs the particle velocity. In these two limiting cases we obtain for the velocity of the Brownian particle

$$\langle\xi(t)\rangle = K\int_0^t \Phi_\xi(t')dt' \tag{3}$$

and

$$\langle\xi(t)\rangle = K\Phi_\xi(t), \tag{4}$$

respectively. These two limiting cases show that in the case of ordinary statistical mechanics the system's response to an external perturbation is expressed in terms of the unperturbed autocorrelation function. We shall refer to the conditions of Eq. (3) and Eq. (4) as the Green-Kubo relation and the Onsager relation, respectively.

The search for a dynamical derivation of anomalous diffusion, that being where the mean square value of the dynamic variable is not linear in time, has been a subject of great interest in recent years. There are two main theoretical perspectives on how to explain the origin of anomalous diffusion. The first perspective is based on the assumption that there are unpredictable events, that the occurrence of these events obey non-Poisson statistics, and is related to the pioneering paper by Montroll and Weiss [2]. The other perspective rests on the assumption that the single diffusion trajectories have an infinite memory. The prototype of the latter perspective is the concept of fractional Brownian motion introduced by Mandelbrot [3]. A problem worthy of investigation is as to whether or not, in the case of anomalous diffusion, the response to external perturbation departs from the predictions of Eqs. (3) and (4). In the last few years, this problem has been addressed by some investigators [4, 5, 6, 7, 8, 9]. These authors have discussed the Green-Kubo relation of Eq. (3). Notice that in the special case of ordinary statistical mechanics this relation can also be written in the following form

$$\langle x(t)\rangle = \frac{K}{2\langle\xi^2\rangle}\langle x^2(t)\rangle_0. \tag{5}$$

To understand how to derive this equation, originally proposed by Bouchaud and George [10], we have to refer ourselves to the following equation of motion

$$\frac{dx}{dt} = \xi(t). \tag{6}$$

Since $\xi(t)$ is a fluctuating velocity, it generates spatial diffusion and we denote by $x(t)$ the position of the corresponding diffusing particle. The external field affects the velocity fluctuation and, consequently, the diffusion process generated by these fluctuations. In the absence of perturbation, the second moment of the diffusing particle, $\langle x(t)^2\rangle_0$, obeys the prescription

$$\langle x(t)^2\rangle_0 = 2\langle\xi^2\rangle\int_0^t dt'\int_0^{t'} dt''\Phi_\xi(t''). \tag{7}$$

It is straightforward to prove that Eq. (3) yields Eq. (5). This is done by considering the mean value of Eq.(6)

$$\frac{d\langle x\rangle}{dt} = \langle\xi(t)\rangle. \tag{8}$$

The time integration of the left hand term of this equation yields the left hand term of Eq. (5) and the time integration of the right hand term of it, using Eq.(3) and Eq.(7), yields the right hand term of Eq. (5).

The relation of Eq. (5) is denoted as *generalized Einstein relation*, because it might hold true also when the equilibrium correlation function does not exist [7]. However, in the case of ordinary statistical mechanics Eq. (5) becomes equivalent to the Green-Kubo property. In this generalized sense we can state that the authors of Refs. [4, 5, 6, 7, 8, 9] studied the Green-Kubo relation of Eq. (3), all of them but the authors of Ref. [4], devoting their attention to the subdiffusional case.

Herein we focus our attention on the Onsager relation of Eq. (4). The only earlier work on the issue of the breakdown of the Onsager principle, caused by anomalous statistics, known to us, is that of Ref. [11]. However, here we plan to address the problem with the adoption of a Liouville-like approach, a fact that will allow us to establish some general conclusions concerning the breakdown of the Onsager prescription. We shall illustrate the rules for the calculation of the four-time correlation, with a prescription that can be easily extended to correlation functions of any order. We expect that these prescriptions might lead to a successful evaluation of the fourth-order correlation function, which has been studied so far by means of a factorization assumption which is violated by the non-Poisson statistics.

II. AN IDEALIZED MODEL OF INTERMITTENT RANDOMNESS AND THE CORRESPONDING DENSITY EQUATION

As done in Ref. [12], let us focus on the following dynamical system. Let us consider a variable y moving within the interval $I = [0, 2]$. The interval is defined over an overdamped potential V, with a cusp-like minimum located at $y = 1$. If the initial condition of the particle is $y(0) > 1$, the particle moves from the right to the left towards the potential minimum. If the initial condition is $y(0) < 1$, then the motion of the particle towards the potential minimum takes place from the left to the right. When the particle reaches the potential bottom it is injected to an initial condition, different from $y = 1$, chosen in a random manner. We thus realize a mixture of randomness and slow deterministic dynamics. The left and right portions of the potential $V(y)$ correspond to the laminar regions of turbulent dynamics, while randomness is concentrated at $y = 1$. In other words, this is an idealization of the map used by Zumofen and Klafter [13], which does not affect the long-time dynamics of the process, yielding only the benefit of a clear distinction between random and deterministic dynamics. Note that the waiting time distribution in the two laminar phases of the reduced form has the same time asymptotic form as

$$\psi(t) = (\mu - 1)\frac{T^{\mu-1}}{(t+T)^\mu}. \tag{9}$$

We select this form as the simplest possible way to ensure the normalization condition

$$\int_0^\infty dt\psi(t) = 1. \tag{10}$$

We note that Eq. (10) implies $\mu > 1$. The condition $\mu > 2$ corresponds to the existence of a finite mean sojourn time, and, thus, to the possibility itself of defining the stationary correlation function of the fluctuation ξ, which, with the choice of Eq. (9) reads [14]

$$\Phi_\xi(t) \equiv \frac{\langle \xi(t)\xi(0)\rangle}{\langle \xi^2\rangle} = \left[\frac{T}{t+T}\right]^{\mu-2}. \tag{11}$$

From within the perspective of a single trajectory this dynamical model reads

$$\dot{y} = \lambda[\Theta(1-y)y^z - \Theta(y-1)(2-y)^z] + \frac{\Delta_y(t)}{\tau_{random}}\delta(y-1). \tag{12}$$

The function $\Theta(x)$ is the ordinary Heaviside step function, $\Delta_y(t)$ is a random function of time that can achieve any value on the interval $[-1,+1]$, and τ_{random} is the injection time that must fulfill the condition of being infinitely smaller than the time of sojourn in one of the laminar phases. Note that z is a real number fitting the condition $z > 1$. In fact, the equality

$$z = \frac{\mu}{(\mu-1)} \tag{13}$$

relates the dynamics of Eq. (12) to the distribution Eq. (9). The Poisson condition is recovered in the limit $z \to 1$, namely, in the limit $\mu \to \infty$. Thus, in a sense, the whole region $z > 1$ ($\mu < \infty$) corresponds to anomalous statistical mechanics. However, the deviation from normal statistical mechanics is especially evident when $z > 1.5$, a condition implying that the second moment diverges. In the case $z > 2$ the departure from ordinary statistical mechanics becomes even more dramatic, due to the fact that the first moment also diverges and, as we shall see in this Section, the process becomes non-ergodic.

Let us move now to the density picture, namely, to a formulation of Eq. (12) from within the Gibbs perspective. The form of this equation is:

$$\frac{\partial}{\partial t}p(y,t) = -\lambda\frac{\partial}{\partial y}[\Theta(1-y)y^z - \Theta(y-1)(2-y)^z]p(y,t) + C(t), \tag{14}$$

where

$$C(t) = 2\lambda p(1,t). \tag{15}$$

It is important to stress that we are forced to set the equality of Eq. (15) to fulfill the following physical conditions

$$\frac{d}{dt}\int_{I=[0,2]} p(y,t)dy = \int_{I=[0,2]} \frac{\partial}{\partial t}p(y,t)dy = 0, \tag{16}$$

which, in turn, ensures the conservation of probability. We assume the ordinary normalization condition

$$\int_{I=[0,2]} p(y,t)dy = 1, \tag{17}$$

which is kept constant in time, as a consequence of Eq. (16). It is evident that the inhomogeneous term $C(t)$ corresponds to the action of the stochastic term, namely, the second term on the right hand side of Eq. (12).

It is important to point out that our dynamic perspective allowed us to describe the intermittent process through the Liouville-like equation

$$\frac{\partial p(\xi,y,t)}{\partial t} = \mathcal{R}p(\xi,y,t), \tag{18}$$

where y denotes a continuous variable moving either in the right or in the left laminar region, with ξ getting the values W or $-W$, correspondingly, and the operator $\mathcal{R}$ reading

$$\mathcal{R} = -\lambda\frac{\partial}{\partial y}[\Theta(1-y)y^z - \Theta(y-1)(2-y)^z] + 2\lambda\int_0^2 dy\delta(y-1). \tag{19}$$

This operator departs from the conventional form of a differential operator, since the last term corresponds to the unusual role of an injection process, which is random rather than being deterministic. In the ordinary Fokker-Planck approach the role of the stochastic force is played by a second-order derivative, which is not as unusual as the integral operator of Eq. (19). Here the role of randomness is played by the back-injection process, which, from within the density perspective is described by an operator that selects from all possible values $p(y,t)$, the specific value of $p(y,t)$ at $y = 1$. The idealization that we have adopted, of reducing the size of the chaotic region to zero, with the choice of the process of back injection located at $y = 1$, makes it possible for us to use the continuous time representation and the equation of motion Eq. (18) rather than the conventional Frobenius-Perron representation. This representation will allow us to obtain analytical results. However, the same physical conclusions would be reached, albeit with more extensive algebra, using the conventional maps and the Frobenius-Perron procedure described in the recent book by Driebe [15].

We note that the equilibrium probability density solving Eq. (14) is given by

$$p_0(y) = \frac{2-z}{2}\left[\frac{\Theta(1-y)}{y^{z-1}} + \frac{\Theta(y-1)}{(2-y)^{z-1}}\right]. \tag{20}$$

This equilibrium density becomes negative for $z > 2$ and signals the important fact that for $z > 2$ there no longer exists an invariant distribution. The lack of an invariant distribution accounts for the nonergodicity in the fluorescence of single nanocrystals, recently pointed out by Brokmann *et al.* [16].

For the purposes of calculation in the next few Sections, it is convenient to split the density $p(y,t)$ into a symmetric and an anti-symmetric part with respect to $y = 1$,

$$p(y,t) = p_S(y,t) + p_A(y,t). \tag{21}$$

This separation based on symmetry yields the following two equations from Eq. (14)

$$\begin{aligned}\frac{\partial}{\partial t}p_S(y,t) &= -\lambda\Theta(1-y)\frac{\partial}{\partial y}[y^z p_S(y,t)] \\ &\quad +\lambda\Theta(y-1)\frac{\partial}{\partial y}[(2-y)^z p_S(y,t)] + C(t)\end{aligned} \tag{22}$$

and

$$\begin{aligned}\frac{\partial}{\partial t}p_A(y,t) &= -\lambda\Theta(1-y)\frac{\partial}{\partial y}[y^z p_A(y,t)] \\ &\quad +\lambda\Theta(y-1)\frac{\partial}{\partial y}[(2-y)^z p_A(y,t)].\end{aligned} \tag{23}$$

We note that the anti-symmetric part of the density is driven by a conventional differential operator, which we denote by $\hat{\Gamma}$. Thus, we rewrite Eq. (23) as follows

$$\frac{\partial}{\partial t}p_A(y,t) = \hat{\Gamma}p_A(y,t), \tag{24}$$

where

$$\hat{\Gamma} \equiv -\lambda\Theta(1-y)\frac{\partial}{\partial y}y^z + \lambda\Theta(y-1)\frac{\partial}{\partial y}(2-y)^z. \tag{25}$$

The operator with the unusual form, containing $C(t)$, is only responsible for the time evolution of the symmetric part of the probability density. We notice, on the other hand, that any physical effect producing a departure of $C(t)$ from its equilibrium value, if this exists, namely, if $z < 2$, implies a departure from equilibrium. A stationary correlation function can be evaluated, as we shall see in the next few Sections, using only Eq. (23), without forcing Eq. (22) to depart from the equilibrium condition.

As we shall see in Section VI, the evaluation of correlation functions of order higher than the second cannot be done without producing a departure of $C(t)$ from its equilibrium value. This might generate the impression that the correlation functions of order higher than the second cannot be evaluated without internal inconsistencies, if we use only the density picture. The evaluation of these higher-order correlation functions was done in Ref. [17], by using a procedure based on the time evolution of single trajectories. Actually, as we shall see in Section VI, the density approach should yield the same result. However, we think that deriving this result using only the Liouville-like equation of this section is a hard task, which was bypassed in the past by means of the factorization approximation [12], violated by the non-Poisson case.

III. THE CORRELATION FUNCTION OF THE DICHOTOMOUS FLUCTUATION FROM THE TRAJECTORY PICTURE

Let us focus our attention on Eq. (12) and consider the initial condition $y_0 \in [0,1]$. Then, it is straightforward to prove that the solution, for $y < 1$ is,

$$y(t) = y_0\left(1-\lambda(z-1)y_0^{z-1}t\right)^{-1/(z-1)}. \tag{26}$$

From (26) and imposing the condition $y(T) = 1$, we can find the time at which the trajectory reaches the point $y = 1$, which is

$$T = T(y_0) = \frac{1-y_0^{z-1}}{\lambda(z-1)y_0^{z-1}}. \tag{27}$$

Since we have to find $\xi(t)$ and $\xi(t) = \xi(y(t))$, from the general form of Eq. (27) we obtain:

$$\begin{aligned}\frac{\xi(t)}{W} &= [\Theta(1-y_0)\Theta(T(y_0)-t) \\ &\quad -\Theta(y_0-1)\Theta(T(2-y_0)-t)] \\ &\quad -\sum_{i=0}^{+\infty}\text{sign}\left[\Delta_y\left(\sum_{k=0}^{i}\tau_k\right)\right]\times \\ &\quad \times\left[\Theta\left(\sum_{k=0}^{i+1}\tau_k - t\right) - \Theta\left(\sum_{k=0}^{i}\tau_k - t\right)\right],\end{aligned} \tag{28}$$

where the time increments are given by

$$\begin{aligned}\tau_0 &= T(y_0) = \frac{1-y_0^{z-1}}{\lambda(z-1)y_0^{z-1}}\Theta(1-y_0) \\ &\quad +\frac{1-(2-y_0)^{z-1}}{\lambda(z-1)(2-y_0)^{z-1}}\Theta(y_0-1) \\ \tau_{i\geq 1} &= \frac{1-[1+\Delta_y(\tau_i)]^{z-1}}{\lambda(z-1)[1+\Delta_y(\tau_i)]^{z-1}}\Theta(-\Delta_y(\tau_i)) \\ &\quad +\frac{1-[1-\Delta_y(\tau_i)]^{z-1}}{\lambda(z-1)[1-\Delta_y(\tau_i)]^{z-1}}\Theta(\Delta_y(\tau_i)).\end{aligned} \tag{29}$$

Then, for the autocorrelation function we obtain the following expression:

$$\begin{aligned}\frac{\langle\xi(t)\xi(0)\rangle}{W^2} &= \langle[\Theta(1-y_0)\Theta(T(y_0)-t) \\ &\quad +\Theta(y_0-1)\Theta(T(2-y_0)-t)]\rangle + \\ &\quad \sum_{i=0}^{+\infty}\left\langle\text{sign}(y_0-1)\text{sign}\left[\Delta_y\left(\sum_{k=0}^{i}\tau_k\right)\right]\times\right. \\ &\quad \left.\times\left[\Theta\left(\sum_{k=0}^{i+1}\tau_k - t\right) - \Theta\left(\sum_{k=0}^{i}\tau_k - t\right)\right]\right\rangle.\end{aligned} \tag{30}$$

As pointed out in Section II, the calculation of the correlation function rests on averaging on the invariant distribution given by Eq. (20). As a consequence of this averaging, the second term in (30) vanishes. In fact, the quantity to average is anti-symmetric, whereas the statistical weight is symmetric.

It is possible to write the surviving term in the autocorrelation as

$$\begin{aligned}\frac{\langle\xi(t)\xi(0)\rangle}{W^2} &= (2-z)\int_0^1 \Theta\left(\frac{1-y^{z-1}}{\lambda(z-1)y^{z-1}}-t\right)\frac{1}{y^{z-1}}dy \\ &= (2-z)\int_0^{(1+\lambda(z-1)t)^{-1/(z-1)}} y^{-z+1}dy \\ &= (1+\lambda(z-1)t)^{-(2-z)/(z-1)} \\ &\equiv (1+\lambda(z-1)t)^{-\beta}, \end{aligned} \tag{31}$$

with

$$\beta = \frac{2-z}{z-1}. \tag{32}$$

Since we focus our attention on $0<\beta<1$, we have to consider $3/2<z<2$. Note that the region $1<z<3/2$ does not produce evident signs of deviation from ordinary statistics. However, as we shall see in Section V, an exact agreement between density and trajectory is recovered only at $z=1$, when the correlation function becomes identical to the exponential function $\exp(-\lambda t)$. Note also that Eq. (31) becomes identical to Eq. (11) after setting the condition

$$\lambda(z-1) = \frac{1}{T}. \tag{33}$$

IV. THE CORRELATION FUNCTION OF THE DICHOTOMOUS FLUCTUATION FROM THE DENSITY PICTURE

The result of the preceding Section is reassuring, since it establishes that the intermittent model we are using generates the wanted inverse power law form for the correlation function of the dichotomous variable $\xi(t)$. In this Section we show that exactly the same result can be derived from the adoption of the Frobenius-Perron form of Eq. (14).

To fix ideas, let us consider the following system: a particle in the interval $[0,1]$ moves towards $y=1$ following the prescription $\dot{y}=\lambda y^z$ and when it reaches $y=1$ it is injected backwards at a random position in the interval. The evolution equation obeyed by the densities defined on this interval is the same as Eq. (14), with $C(t)=\lambda p(1,t)$. This dynamic problem was already addressed in Refs. [18, 19], and solved using the method of characteristics as detailed in Ref. [20]. It is important to stress that this approach is the requisite price for adopting the idealized version of intermittency. The adoption of the more conventional reduced map of Ref. [13] would have made it possible for us to adopt the elegant prescriptions of Driebe [15], as done in Ref. [18].

Let us remind the reader that the solution afforded by the method of characteristics, in the case of this simple nonlinear equation with stochastic boundary conditions is

$$\begin{aligned} p(y,t) &= \int_0^t \frac{\lambda p(1,\xi)}{g_z((t-\xi)y^{z-1})}d\xi \\ &\quad + p\left(\left[\frac{y}{[g_z(y^{z-1}t)]^{1/z}}\right],0\right)\times \\ &\quad \times\frac{1}{g_z(y^{z-1}t)} \end{aligned} \tag{34}$$

where

$$g_z(x) \equiv (1+\lambda(z-1)x)^{z/(z-1)}. \tag{35}$$

To find the autocorrelation function $\langle\xi(t)\xi(0)\rangle$ using only densities, we have to solve Eqs. (22) and (23), which are equations of the same form as that yielding Eq. (34). For this reason we adopt the method of characteristics again. To do the calculation in this case, it is convenient to adopt a frame symmetric with respect to $y=1$. Then, let us define $Y=y-1$. Using the new variable and Eq. (34), we find for Eqs. (22) and (23) the following solution

$$\begin{aligned} p_S(Y,t) &= \int_0^t \frac{\lambda p_S(0,\tau)}{g_z((t-\tau)(1-|Y|)^{z-1})}d\tau + \\ &\quad p_S\left(\left[1-\frac{1-|Y|}{[g_z((1-|Y|)^{z-1}t)]^{1/z}}\right],0\right)\times \\ &\quad \times\frac{1}{g_z((1-|Y|)^{z-1}t)} \end{aligned} \tag{36}$$

and

$$\begin{aligned} p_A(Y,t) &= p_A\left(\mathrm{sign}(Y)\left[1-\frac{1-|Y|}{[g_z((1-|Y|)^{z-1}t)]^{1/z}}\right],0\right)\times \\ &\quad \times\frac{1}{g((1-|Y|)^{z-1}t)}. \end{aligned} \tag{37}$$

Then, the solution consists of two terms: (1) the former is an *even* term and is responsible for the long time limit of the distribution evolution and (2) the latter is an *odd* term which disappears in the long-time limit. We note that (2) is a desirable property because Eq. (23) does not contain the injection term $C(t)$ and the equilibrium density (20) is an *even* function, independently of the symmetry of the initial distribution.

As pointed out in Section II, the two-time correlation function is determined by the anti-symmetric part of the probability density alone. Thus, the definition of the autocorrelation function does not conflict with the equilibrium assumption. In Section V, we shall see that this conflict reemerges when we attempt to evaluate higher-order correlation functions. Let us calculate the autocorrelation function explicitly:

$$\begin{aligned} \langle\xi(t)\xi(0)\rangle &= (2-z)\int_0^1 \frac{1}{(1-\tau)^{z-1}}\bigg|_{\tau=1-\frac{1-Y}{[g_z((1-Y)^{z-1}t)]^{1/z}}} \times \\ &\quad \times\frac{1}{g_z((1-Y)^{z-1}t)}dY \end{aligned} \tag{38}$$

The integral (38) is exactly solvable and leads to the expression

$$\langle \xi(t)\xi(0)\rangle = (1+\lambda(z-1)t)^{-(2-z)/(z-1)}, \qquad (39)$$

which is the same result as that found in Section III, using trajectories rather than the probability densities.

In a similar way, it is possible to calculate the autocorrelation $\langle Y(t)Y(0)\rangle$ and determine that its temporal behavior is an inverse power law with the same exponent as that in Eq. (39).

V. ONSAGER REGRESSION TO EQUILIBRIUM

In conclusion, in the two preceding Sections we have established that the Liouville-like representation of Eq. (18) yields, as expected, the equilibrium correlation function of Eq. (11) with

$$T \equiv \frac{\mu-1}{\lambda}. \qquad (40)$$

What about Onsager's regression to equilibrium? Let us first of all discuss a physical condition where the prescription of Eq. (4) is fulfilled. Let us consider the initial distribution $p(y,0)$ defined as follows

$$p(y,0) = 0,\ y < 1 \qquad (41)$$

and

$$p(y,0) = 2p_0\left(y\right),\ y > 1, \qquad (42)$$

where $p_0(y)$ denotes the equilibrium distribution of Eq. (20). It is convenient to point that this equilibrium distribution is symmetric and that the factor of 2 serves the purpose of normalizing the out-of equilibrium condition that we are studying. Let us denote by $\Delta P(t)$ the population difference between the left and the right state. The choice we made sets the initial condition $\Delta P(0) = 1$. It is straightforward to show that Onsager's regression implies that $\Delta P(t) = \Phi_\xi(t)$. Let us split the initial distribution in the symmetric and anti-symmetric part,

$$p(y,0) = p_0(y) + p_A^{(eq)}(y,0). \qquad (43)$$

We note that

$$p_A^{(eq)}(y,0) = -p_0(y), y < 1 \qquad (44)$$

and

$$p_A^{(eq)}(y,0) = p_0(y), y > 1. \qquad (45)$$

The symmetric part does not contribute to $\Delta P(t)$, only the anti-symmetric part does. Thus, we obtain

$$\Delta P(t) = 2\int_0^1 \exp(\hat{\Gamma}t)p_A^{(eq)}(y,0), \qquad (46)$$

where $\hat{\Gamma}$ is the operator driving the anti-symmetric part, defined by Eq. (25). With the choice of initial condition made it is straightforward to prove that the right end side of Eq. (46) is the equilibrium correlation function evaluated in Sections III and IV. Thus, the prescription of ordinary statistical physics is fulfilled.

What about the regression to equilibrium in general? We note that we can adapt the earlier arguments to any initial condition thereby yielding

$$\Delta P(t) = 2\int_0^1 \exp(\hat{\Gamma}t)p_A^{(noneq)}(y,0). \qquad (47)$$

Note that $p_A^{(noneq)}(y,t)$ obeys the Liouville-like prescription corresponding to $\frac{dy}{dt} = \lambda y^z$, namely, the equation of motion of the left laminar region, without any back-injection process. Since, as we have seen in Section II, the back-injection term serves the purpose of keeping constant the population of the system, we have that $\Delta P(t) \to 0$ for $t \to \infty$. More precisely, we obtain

$$\frac{d\Delta P(t)}{dt} = 2\hat{\Gamma}\int_0^1 \exp(\hat{\Gamma}t)p_A^{(noneq)}(y,0) = -2p_A^{(noneq)}(1,t). \qquad (48)$$

The superscript *noneq* serves the purpose of pointing out that in general the perturbation process creating the necessary initial asymmetry will not realize, for the left portion of the asymmetric component, a form exactly identical to the left portion of the equilibrium distribution. This observation makes it possible to estimate the time asymptotic behavior of $\Delta P(t)$ in general. The exact time evolution of $\Delta P(t)$ depends on the detailed effects of the perturbation. However, for any realistic perturbation, we can prove that the time asymptotic behavior obeys an universal prescription. If the perturbation does not affect the equilibrium distribution in the regions close to the borders, namely for $y \leq \epsilon$ and $y \geq 2-\epsilon$, where $\epsilon \ll 1$, we have that $p_A^{(noneq)}(y,0)$ vanishes for $y \leq \epsilon$. To evaluate the asymptotic behavior of $\Delta P(t)$ we select the infinitesimal portion $dy(0)$ of the interval $[0,1]$, closest to $y = 0$, where $p_A^{(noneq)}(y,0)$ does not vanish. We call M the number of trajectories located in this interval at $t = 0$. The asymptotic behavior of $\Delta P(t)$ is determined by the time necessary for these trajectories to reach the border $y = 1$. The first trajectory will reach the border after a given time $t = T_{first}$, after which the number M will begin decreasing, thereby determining the decay of $\Delta P(t)$ in this time asymptotic region. It is straightforward to prove that the time of arrival at $y = 1$ of the trajectory with initial condition $y(0)$, called t, is related to $y(0)$ by

$$y(0) = \frac{1}{[1+(z-1)\lambda t]^{\frac{1}{z-1}}}. \qquad (49)$$

Thus we obtain

$$\frac{d\Delta P(t)}{dt} \approx \frac{dM}{dt} = \frac{1}{[1+(z-1)\lambda t]^{\frac{z}{z-1}}}. \qquad (50)$$

By integrating this equation and taking into account that $z = \mu/(\mu - 1)$, we finally obtain

$$\lim_{t\to\infty} \Delta P(t) \propto \frac{1}{t^{\mu-1}}, \tag{51}$$

which sanctions the breakdown of Eq. (4).

VI. HIGHER-ORDER CORRELATION FUNCTIONS

We now show that the calculation of higher-order correlation functions, though difficult, can be done by establishing an even deeper connection with the quantum mechanical perspective. Let us address the problem of evaluating the fourth-order correlation function $\langle\xi(t_4)\xi(t_3)\xi(t_2)\xi(t_1)\rangle$. According to the prescription that we adopted in Section IV we must proceed as follows. We move from the equilibrium distribution and let it evolve for a time t_1. The distribution selected is in equilibrium. Therefore it will remain unchanged. At time $t = t_1$ we apply the operator ξ to the distribution. Since the equilibrium distribution is symmetric, the application of the sign operator changes it into the anti-symmetric distribution. We let this distribution evolve for the time $t_2 - t_1$. This has the effect of yielding

$$\Phi_\xi(t_2 - t_1)p_A^{(eq)}(y, 0) + p_A^{(noneq)}(y, t_2 - t_1).$$

At this stage there are two possibilities:

(a) $p_A^{(noneq)}(y, t_2 - t_1) = 0$;

(b) $p_A^{(noneq)}(y, t_2 - t_1) \neq 0$.

Let us consider the case (a) first. In this case, we proceed as follows. At time t_2 we apply to the distribution the operator ξ, and we change it into the original equilibrium distribution. This means that the time evolution from t_2 to t_3 leaves it unchanged. At time t_3 we apply to it the operator ξ and we turn it into $p_A^{(eq)}$ again. We let this distribution evolve till to time t_4. At this time we apply to it the operator ξ again and we make the final average. The result of the condition (a) yields

$$\langle\xi(t_4)\xi(t_3)\xi(t_2)\xi(t_1)\rangle = \langle\xi(t_4)\xi(t_3)\rangle\langle\xi(t_2)\xi(t_1)\rangle. \tag{52}$$

In a recent work [17] it has been shown that this factorization condition is violated by non-Poisson statistics. The demonstration was made by applying the method of conditional probabilities to the study of single trajectories. Thus we are forced to consider case (b).

The problem with condition (b) is that it yields a distribution with the symmetric component departing from equilibrium. This departure is in an apparent conflict with the assumption that the autocorrelation function is calculated using the equilibrium condition. In fact it seems to be equivalent to stating that the calculation of an equilibrium correlation function generates an out of equilibrium condition. Let us see why.

In the case of the two-time correlation function we apply the operator ξ to the density distribution twice. The first application allows us to observe the time evolution of the anti-symmetric component distribution density, with no conflict with equilibrium, given the fact that the symmetric component remains at equilibrium. The second application of ξ turns

$$\Phi_\xi(t_2 - t_1)p_A^{(eq)}(y, 0) + p_A^{(noneq)}(y, t_2 - t_1)$$

into

$$\Phi_\xi(t_2 - t_1)p_S^{(eq)}(y, 0) + p_S^{(noneq)}(y, t_2 - t_1).$$

The calculation done in Section IV proves that $\mathrm{Tr}[p_S^{(noneq)}(y, t_2 - t_1)] = 0$, with the symbol Tr denoting, for simplicity, the integration over y from 0 to 2. To evaluate the fourth-order correlation function, after applying the operator ξ for the second time, we must study the time evolution of $p_S^{(noneq)}(y, t_2-t_1)$ from t_2 to t_3, yielding to $p_S^{(noneq)}(y, t_3, t_2, t_2 - t_1)$. This is a contribution generating some concern, since it activates again the back injection process, which we have seen to be intimately related to the deviation from equilibrium. However, the compatibility with equilibrium condition is ensured by the property $\mathrm{Tr}[p_S^{(noneq)}(y, t_3, t_2, t_2 - t_1)] = 0$.

At time t_3 we have to apply the operator ξ again, and this allows us to make an excursion in the anti-symmetric representation, with the time evolution given by the operator $\exp(\hat{\Gamma}(t_4 - t_3))$. At time t_4 we apply the operator ξ again, we go back to the symmetric representation and we conclude the calculation by means of the trace operation.

In conclusion, the compatibility with equilibrium is guaranteed by the fact that at the intermediate steps of the calculation $\mathrm{Tr}[p] = 0$ (the final step, of course, generates the density-generated correlation function, thereby implying $\mathrm{Tr}[p] \neq 0$). If the intermediate p is anti-symmetric, this condition is obvious. If the intermediate p is symmetric, the vanishing trace condition generates the apparently unphysical property that the symmetric contribution gets negative values over some portions of the interval I. We have to stress, however, that the Liouville-like approach illustrated in this paper keeps the distribution density $p(y, t)$ definite positive, as it must. The generation of a negative distribution density refers to the calculation of the equilibrium correlation functions, of any order, and it is a quantum-like property that must be adopted to guarantee that the genuine distribution density remains in the equilibrium condition.

VII. CONCLUDING REMARKS

In conclusion, this paper shows how to derive the equilibrium correlation function using only information afforded by the Liouville-like approach. The intriguing problem to solve was how to use the Liouville equation, without conflicting with the equilibrium condition. The solution of this intriguing problem is obtained by splitting the Liouville equation into two independent components, the symmetric component corresponding to Eq. (22) and the anti-symmetric component corresponding to Eq. (23). This splitting allows us to study the regression to equilibrium of the correlation function $\Phi_\xi(t)$, through Eq. (23), without ever departing from equilibrium, a physical condition that is controlled by the independent equation of motion for the symmetric component, Eq. (22). This is very formal, and Section V makes it possible for us to reconcile it with physical intuition. We imagine that the Liouville equation is used to evaluate the difference between the population of the left and right state. This makes it possible to establish a direct connection between the experiment of regression to equilibrium and the formalism of Sections II and IV. We have to create an asymmetrical initial condition, with more population on the left than on the right. The time evolution of $\Delta P(t)$ depends only on the time evolution of the anti-symmetric component of the distribution density, and consequently only on the operator $\hat{\Gamma}$. This is the reason why it is possible in principle to connect regression from an out of equilibrium initial condition to the equilibrium correlation function, a condition that implies no deviation from equilibrium distribution. However, creating in a finite time an out of equilibrium condition such that the left part of the anti-symmetric component is identical to the left part of the equilibrium distribution, is impossible. This is the reason why we predict the breakdown of the Onsager principle in general.

Section VI explains why in the literature on dichotomic fluctuations the factorization assumption of Eq. (52) is often made regardless of the Poisson or non-Poisson nature of the underlying process. See, for instance, the work of Fulinski [21] as well as Ref. [12]. In fact, if condition (a) applies, the higher-order correlation functions are factorized, thereby making their calculation easy. However, this assumption conflicts with the trajectory arguments of Ref. [17], which prove that the factorization condition is violated by the non-Poisson condition. A rigorous use of the Liouville equation shows that condition (a) does not apply, and that we have to use condition (b) instead. The calculation of the fourth-order correlation function is not straightforward, and this is the reason why, to the best of our knowledge, it was never done using the Liouville approach.

This is a fact of some relevance for the creation of master equations with memory. There are two major classes of generalized master equations. The first class is discussed, for instance, in Ref. [11]. The master equations of this class are equivalent to the Continuous Time Random Walk (CTRW) of Montroll and Weiss [2] and are based on the waiting time distribution $\psi(t)$. The second class of master equations is based on the correlation function $\Phi_\xi(t)$. Recent examples of this second class can be found in Ref. [12] and in Ref. [22]. Due to the direct dependence on the correlation function $\Phi_\xi(t)$, the derivation of the master equations of this second class is made easy by the factorization assumption. It must be pointed out, on the other hand, that the factorization property, which is not legitimate with renewal non-Poisson processes, is a correct property if the deviation from the exponential relaxation is obtained by time modulation of a Poisson process [22]. Beck [23] is the advocate of the modulation process as generator of complexity. Thus, we find that the master equations of the first class are generated by the renewal perspective of Montroll and Weiss [2] and those of the second class are the appropriate tool to study complexity along the lines advocated by Beck [23]. We think that the results of the present paper might help the investigators in the field of complexity to make the proper choice, either modulation or renewal[24], or a mixture of the two conditions.

Acknowledgements. PG thankfully aknowledges the Army Research Office for financial support through Grant DAAD19-02-0037

[1] R. Kubo, M. Toda, N. Hashitsume, *Statistical Physics II, Nonequilibrium Statistical Mechanics*, Springer-Verlag, Berlin (1991).
[2] E.W. Montroll and G.H. Weiss, J. Math. Phys. **6**, 167 (1965).
[3] B.B. Mandelbrot, *The Fractal Geometry of Nature*, W.H. Freeman and Company, New York (1983).
[4] G. Trefán, E. Floriani, B. J. West, and P. Grigolini, Phys. Rev. E **50**,2564 (1994).
[5] E. Barkai and J. Klafter, Phys. Rev. Lett. **79**, 2245 (1997).
[6] E. Barkai and J. Klafter, Phys. Rev. E **57**, 5237 (1998).
[7] E. Barkai and V.N. Fleurov, Phys. Rev. E **58**, 1296 (1998).
[8] R. Metzler, J. Klafter, I. M. Sokolov, Phys. Rev. E **58**, 1621 (1998).
[9] I. M. Sokolov, A. Blumen and J. Klafter, Europhys. Lett.**56**, 175 (2001).
[10] J.P. Bouchaud and A. Georges, Phys. Rep. **195**, 127 (1990).
[11] P. Allegrini, G. Aquino, P. Grigolini, L. Palatella, and A. Rosa, Phys. Rev. E **68**, 056123 (2003).
[12] M. Bologna, P. Grigolini and B.J. West, Chem. Phys. **284**, 115-128 (2002).
[13] G. Zumofen and J. Klafter, Phys. Rev. E **47**, 851 (1993).
[14] T. Geisel, J. Nierwetberg, and A. Zacherl, Phys. Rev. Lett. **54**, 616 (1985).
[15] D. J. Driebe, *Fully Chaotic Maps and Broken Time Symmetry*, Kluwer Academic Publishers, Dordrecht (1999).
[16] X. Brokman, J. -P.Hermier, G. Messin, P. Desbiolles, J.-P. Bouchaud, and M. Dahan, Phys. Rev. Lett. **90**, 120601 (2003).
[17] P. Allegrini, P. Grigolini, L. Palatella, B.J. West, submitted to Phys. Rev. E.

[18] M. Bologna, P. Grigolini, M. Karagiorgis, and A. Rosa, Phys. Rev. E, **64**, 016223 (2001).

[19] M. Ignaccolo, P. Grigolini, and A. Rosa, Phys. Rev. E, **64**, 026210 (2001).

[20] N. Goldenfeld, *Lectures on Phase Transitions and the Renormalization Group*, Addison-Wesley Publisher, Cambridge (1992).

[21] A. Fulinski, Phys. Rev. E **52**. 4523 (1995).

[22] M. Bologna, P. Grigolini, M.G. Pala, L. Palatella, Chaos, Solitons and Fractals, **17**, 601 (2003).

[23] C. Beck, Phys. Rev. Lett. **87**, 180601 (2001).

[24] In the case where the renewal condition applies, we want to point out that, as shown in [11], the generalized master equation can be derived from the CTRW [2]. It is still unknown how to do that using the Liouville-like approach of Section II, probably due to the complexity of the problem illustrated in Section VI. Furthermore, the authors of Ref. [9] made the following impressive discovery. In the non-Poisson case even if a correct generalized master equation is available [25], its response to external perturbations departs from the correct prediction that one can obtain by perturbing the CTRW instead. Thus the trajectory-density conflict [12] seems to be the manifestation of technical problems, emerging from the density perspective, which can be settled using the trajectory (CTRW) perspective.

[25] Within the theoretical framework of this paper, the fractional Fokker-Planck equation used by Sokolov, Blumen and Klafter [9] belong to the class of generalized master equations that can be derived from the CTRW,