PHYSICAL REVIEW E **76**, 062601 (2007)

Dynamic correlations of macroscopic quantities

Olivier Pierre-Louis
CNRS/Rudolf Peierls Centre for Theoretical Physics, 1 Keble Road, Oxford OX1 3NP, United Kingdom

(Received 3 September 2007; published 4 December 2007)

The dynamics of fluctuations of a family of macroscopic quantities in one-dimensional systems (which includes the roughness and the total length of an interface, and the end-to-end distance of a filament) are analyzed. At thermodynamic equilibrium, the time-correlation function of these quantities can be expressed in terms of time-correlation functions of local quantities. In some cases, macroscopic quantities exhibit a universal diffusive behavior, which is not reached by local quantities.

DOI: 10.1103/PhysRevE.76.062601 PACS number(s): 68.03.Kn, 05.40.−a

Large fluctuations are ubiquitous in one-dimensional systems. Some examples of fluctuating one-dimensional systems are biopolymers [1] in Hele-Shaw cells, atomic steps on crystal surfaces [2], or magnetic domain walls in two dimensions [3]. Fluctuation dynamics are usually analyzed by means of time-correlation functions (TCF) of local quantities. But many important features or applications are related to global quantities. As an example, the surface contribution to conductance fluctuations in nanometric electric lines is proportional to the fluctuations of the total number of surface defects. This number is proportional to the length of atomic steps on the surface [4]. Therefore, there is a contribution to conductance fluctuation which is proportional to total step length fluctuations [4]. Other macroscopic quantities may be of interest, such as the roughness of an atomic step, or the end-to-end length of a fluctuating biopolymer [1,5,6].

In the present paper, we focus on a family of quantities defined as quadratic functionals. All quantities mentioned above, such as the total length and the roughness of an interface, or the end-to-end length of a filament, may be written in terms of these functionals. The short-time behavior of their TCFs is calculated. Two main results are obtained: (i) macroscopic TCFs are explicitly related to the local TCFs at thermodynamic equilibrium, and (ii) the equilibrium TCF of some macroscopic quantities exhibits a universal diffusive behavior, which cannot be reached by means of local quantities.

Let us consider a one-dimensional system along x, described by the function $h(x,t)$, where t is the time. The evolution of h is governed by a linear Langevin equation as follows:

$$\partial_t h(x,t) = \Omega[h(x,t)] + \eta(x,t), \tag{1}$$

where Ω is a linear function of h and its derivatives with respect to x, and η is a fluctuating Langevin force. The function h accounts for the position of an interface, or for the local orientation of a filament. In the following the derivation of the main results is first presented without reference to a specific system. Thereafter, several precise cases will be discussed.

Assuming that the system is periodic along x, of period L, the spatial Fourier transform of $h(x,t)$ is defined as $h_k(t) = \int dx e^{-ikx} h(x,t)$, with $k = 2\pi\ell/L$, where ℓ is an integer. Here, and in the following, the bounds of the integrals will be $-L/2$, and $L/2$ when they are not specified. The amplitude of the Fourier modes obeys a linear Langevin equation as follows:

$$\partial_t h_k(t) = i\omega_k h_k(t) + \eta_k(t). \tag{2}$$

The noise has zero average, i.e., $\langle \eta_k(t) \rangle = 0$, and

$$\langle \eta_k(t) \eta_{k'}(t') \rangle = B_k \delta_{k+k'} \delta(t-t')(L/2\pi), \tag{3}$$

where δ_k is the Kronecker delta symbol, and $\langle\ \rangle$ denotes an average over the realizations of the noise η, which is chosen to be Gaussian. It is assumed that the systems enjoy the $x \to -x$ symmetry and is stable, so that $i\omega_k = i\omega_{-k}$ and $\mathrm{Re}[i\omega_k] < 0$. For the sake of simplicity, it is also assumed that the initial system is flat, i.e., $h(x,0) = 0$ [7].

In order to focus on shape fluctuations rather than on the global motion of the system, one may define

$$\zeta(x,t) = h(x,t) - \frac{1}{L}\int dx h(x,t). \tag{4}$$

From this definition, $\zeta_k(t) = h_k(t)$ when $k \neq 0$ and $\zeta_{k=0}(t) = 0$.

Local quantities are quantities which can be measured at a given abscissa x without the knowledge of what happens in the rest of the system, and are therefore combinations of $\zeta(x)$ and its spatial derivatives. Local TCFs are thus defined for $m \geq 0$ and $t' > t$ as

$$G^{\partial_x^m \zeta}(t,t') = \langle \partial_x^m \zeta(x,t) \partial_x^m \zeta(x,t') \rangle, \tag{5}$$

where $\partial_x^m \zeta(x,t)$ is the mth spatial derivative of ζ, and the notation $\partial_x^0 \zeta(x,t) = \zeta(x,t)$ is used. Upon substitution of the solution of Eq. (2) into Eq. (5), one finds

$$G^{\partial_x^m \zeta}(t,t') = \frac{1}{2\pi L} \sum_{k \neq 0} k^{2m} \beta_k(t,t'), \tag{6}$$

where

$$\beta_k(t,t') = \frac{B_k}{2i\omega_k} [e^{i\omega_k(t'+t)} - e^{i\omega_k(t'-t)}]. \tag{7}$$

Let us consider the family of quadratic functionals

$$Q_p(t) = \int dx[\partial_x^p \zeta(x,t)]^2. \tag{8}$$

We shall provide examples of macroscopic quantities related to Q_p later, when we discuss some specific systems.

The temporal correlations of Q are defined as

$$G^{\delta Q_p}(t,t') = \langle \delta Q_p(t) \delta Q_p(t') \rangle, \tag{9}$$

where δQ_p is the deviation of Q_p from its average value as follows:

$$\delta Q_p(t) = Q_p(t) - \langle Q_p(t) \rangle. \tag{10}$$

The calculation of $G^{\delta Q_p}$ involves the evaluation of four-point correlation functions, which are decomposed into two-point correlation functions for Gaussian processes [8], leading to

$$G^{\delta Q_p}(t,t') = 2\sum_{k\neq 0} k^{4p}\beta_k(t,t')^2. \tag{11}$$

During the initial roughening from the flat state, when $t,t' \ll \mathrm{Re}[i\omega]^{-1}$, one finds $\beta_k(t,t') \sim t$, and one may thus infer from Eqs. (6) and (11) that $G^{\partial_x^m \zeta}(t,t') \sim t$, while $G^{\delta Q_p}(t,t') \sim t^2$ [9]. These early time scalings do not depend on the precise form of $i\omega_k$.

Since $\mathrm{Re}[i\omega_k] < 0$, the system reaches a stable steady state at long times, which is identified with thermodynamic equilibrium. The energy, defined as

$$\mathcal{E}(t) = \frac{\sigma}{2}\int dx[\partial_x \zeta(x,t)]^2, \tag{12}$$

then imposes the static spectrum as follows:

$$\langle \zeta_k(t)\zeta_{k'}(t)\rangle = L\delta_{k+k'}\frac{k_B T}{\sigma k^2}. \tag{13}$$

The compatibility of this spectrum with that of the solution $\zeta(x,t)$ of Eq. (2) when $t\to\infty$ leads to

$$B_k = 4\pi i\omega_k \frac{k_B T}{\sigma k^2}. \tag{14}$$

At equilibrium (i.e., when $t,t' \to \infty$), correlation functions $G(t,t')$ only depend on $t-t'$, and we shall define $G_{eq}(\tau) = G(t,t+\tau)$, with $\tau > 0$. An inspection of Eqs. (6), (7), and (11) in the limit $t,t' \to \infty$ allows one to write

$$G_{eq}^{\delta Q_p}(\tau) = 8\pi^2 L\frac{k_B T}{\sigma} G_{eq}^{\partial_x^{2p-1}\zeta}(2\tau). \tag{15}$$

Note that this relation is not valid during the initial transient relaxation to equilibrium.

Using Eq. (15), the derivation of TCFs of the macroscopic and nonlinear quantities Q_p, reduces to the derivation of local TCFs for arbitrary m (including $m<0$, which is consistent if Eq. (6) is taken as a definition of $G_{eq}^{\partial_x^m \zeta}(\tau)$). The following paragraphs are therefore devoted to the derivation of the local TCFs.

We start with the static correlation functions. We shall first notice that, when m increases, the correlation functions probe the fluctuations in an increasingly local fashion. As a consequence, the local static correlation functions diverge at small scales for $m > 1/2$, and at large scales for $m < 1/2$ [10] as follows:

$$G_{eq}^{\partial_x^m \zeta}(0) = \frac{k_B T}{\pi\sigma} \times \begin{cases} \dfrac{\left(\frac{2\pi}{a}\right)^{2m-1}}{2m-1}, & m > \frac{1}{2} \\ Z[2-2m]\left(\frac{L}{2\pi}\right)^{1-2m}, & m < \frac{1}{2} \end{cases}, \tag{16}$$

where a is a microscopic cutoff (with $a/L \to 0$), and Z is the Riemann zeta function.

In order to proceed with the calculation of dynamic TCF, one needs to specify the relaxation dynamics. For the sake of simplicity, a power law is assumed as follows:

$$i\omega_k = -A|k|^{n+2}, \tag{17}$$

where $A > 0$ is a system-dependent constant.

Equilibrium will be reached when the slowest long wavelength modes have relaxed, i.e., for $\tau \gg \tau_{sat}$, where

$$\tau_{sat} = (L/2\pi)^{2+n}/A. \tag{18}$$

The time scale τ_{sat} is, in general, the time above which correlations vanish. In the following, the TCFs are derived at equilibrium in the short-time limit $\tau \ll \tau_{sat}$. In such a limit, the dynamics at a given point x of the system is essentially independent of the boundary conditions. Indeed, their influence does not have enough time to propagate over the whole system up to the point x. Hence, we expect our results to be qualitatively valid not only for periodic boundary conditions, but also for other types of boundary conditions.

When $m > 1/2$, the TCF reads [11]

$$G_{eq}^{\partial_x^m \zeta}(\tau) = \frac{k_B T}{2\pi\sigma}\frac{\Gamma(\mu)}{2+n}(A\tau)^{-\mu}, \tag{19}$$

where Γ denotes the gamma function, and

$$\mu = \frac{2m-1}{2+n}. \tag{20}$$

Since $G_{eq}^{\partial_x^m \zeta}(\tau)$ diverges for $m < 1/2$, a more suitable correlation function is designed as

$$\begin{aligned} F^{\partial_x^m \zeta}(t,t') &= \langle [\partial_x^m \zeta(t) - \partial_x^m \zeta(t')]^2 \rangle \\ &= G^{\partial_x^m \zeta}(t,t) + G^{\partial_x^m \zeta}(t',t') - 2G_{eq}^{\partial_x^m \zeta}(t,t'), \end{aligned} \tag{21}$$

so that

$$F_{eq}^{\partial_x^m \zeta}(\tau) = 2[G_{eq}^{\partial_x^m \zeta}(0) - G_{eq}^{\partial_x^m \zeta}(\tau)]. \tag{22}$$

The correlation function $F^{\delta Q_p}$ is defined in a similar way. With these definitions, the equilibrium relation (15) also holds when G is replaced by F. We may now discuss the case $m < 1/2$, which leads to

$$F_{eq}^{\partial_x^m \zeta}(\tau) = \frac{k_B T}{2\pi\sigma} \times \begin{cases} \frac{-\Gamma[\mu]}{2+n}(A\tau)^{-\mu}, & m > -\frac{1+n}{2} \\ \left[\ln\left(\frac{(L/2\pi)^{2+n}}{2A\tau}\right) + 1 - \gamma\right](A\tau), & m = -\frac{1+n}{2} \\ \frac{1}{1-2m-n}\left(\frac{L}{2\pi}\right)^{1-2m-n}(A\tau), & m < -\frac{1+n}{2} \end{cases}, \tag{23}$$

where $\gamma \approx 0.577$ is the Euler constant.

Since it corresponds to the correlations of the directly measurable quantity ζ, the case $m=0$, with $F_{eq}^{\zeta}(\tau) \sim \tau^{-\mu}$, has been widely studied in the literature. But beyond the usual $\sim \tau^{-\mu}$ regime, the TCFs exhibit, for m negative enough, a regime $\sim \tau$, with a nontrivial L scaling. We call this regime the diffusive regime, from the analogy with simple diffusion, where the correlation function is linear in time.

While the diffusive regime cannot be observed from local TCFs (which correspond to $m \geq 0$), it appears to be observable via macroscopic TCFs. Indeed, from Eqs. (15) and (23), $F_{eq}^{\delta Q_p} \sim \tau$ when

$$p \leq p^* = \frac{1-n}{4}. \tag{24}$$

Thus, the TCF of Q_1, which corresponds to $p=1>p^*$, is found to obey Eq. (19). But Q_0 may exhibit a diffusive behavior as follows:

$$F_{eq}^{\delta Q_0}(\tau) \sim \begin{cases} \tau^{3/4} L, & n=2 \\ \tau[\ln(\tau_{sat}/\tau) + 1 - \gamma]L, & n=1 \\ \tau L^2, & n=0 \end{cases} . \tag{25}$$

The diffusive scaling of the TCFs roots in their long wavelength divergence. Indeed, when $p<p^*$, the TCFs are dominated by the behavior at $k \to 0$ [12].

The diffusive scaling is universal in the sense that any spatially integrated quantity may exhibit such a behavior. As an example, consider the integral of ζ,

$$I_1(x,t) = \int_{-x/2}^{x/2} dx' \, \zeta(x',t). \tag{26}$$

From the definition of ζ, one has $I_1(L,t)=0$. Its equilibrium TCF reads

$$G_{eq}^{\delta I_1}(\tau) = \frac{1}{\pi L} \lim_{t\to\infty} \sum_k k^{-2} \beta_k(t,t+\tau)[1-\cos(kx)]. \tag{27}$$

In the limit $x^{n+2}/A \ll \tau \ll L^{n+2}/A$, the process is fully relaxed at the scale x, and the system segment of size x behaves as a local quantity, so that $F_{eq}^{\delta I_1}(\tau) \approx x^2 F_{eq}^{\zeta}(\tau)$. At short times, when $\tau \ll x^{n+2}/A$, with $x \ll L$, the dynamics is not fully relaxed at the scale x, and the TCF of I_1 exhibits the same temporal scaling as that of Q_0. Such a result can be traced back to the fact that both quantities I_1 and Q_0 are obtained from ζ by means of one spatial integration. Note that the fact that I_1 and Q_0 are, respectively, linear and nonlinear in ζ does not play a role here.

The above results are relevant to a variety of systems. In the case where $h(x,t)$ is the position of an interface along the Cartesian coordinate x, the root mean square roughness $\mathcal{W}$ and the total interface length $\mathcal{L}$ may be written as a function of Q_p as follows:

$$\mathcal{W}(t)^2 = \frac{Q_0(t)}{L}, \tag{28}$$

$$\mathcal{L}(t) \approx L + \frac{Q_1(t)}{2}, \tag{29}$$

where the second relation is an approximation for small $\partial_x \zeta$. Such an expression applies to atomic steps on crystal surfaces [2], to a liquid-liquid interface in a Hele-Shaw cell, or to a domain wall between two-dimensional (2D) magnetic domains [3]. The term 2 in the exponent of Eq. (17) then comes from the assumption of linear relaxation of the energy, which should be proportional to the driving force $\delta\mathcal{E}/\delta\zeta \sim \partial_{xx}\zeta$, where δ denotes the functional derivative.

In the case of an isolated atomic step, n depends on the mass transport mechanism [13]: $n=0$ for attachment-detachment limited dynamics, $n=1$ for diffusion limited dynamics, and $n=2$ for edge diffusion limited dynamics (more general, non-power-law relaxations have also been derived [13–16]). The exponent n depends on the kinetics. For the relaxation of the interface between two incompressible fluids in a Hele-Shaw cell, hydrodynamics is described by Darcy's law, and $n=1$. In the case of a domain wall between 2D magnetic domains with Glauber dynamics, one expects $n=0$ [3].

The interface roughness $\mathcal{W}^2$ is related to Q_0 via Eq. (28), so that $F_{eq}^{\delta\mathcal{W}^2}(\tau) = F_{eq}^{\delta Q_0}(\tau)/L^2$. Therefore, the diffusive regime can be probed for $n=0$, which corresponds, for example, to attachment-detachment dynamics of an atomic step. Liquid-liquid interfaces, with $n=1$, do not exhibit a diffusive behavior for Q_0.

For interface length fluctuations $p=1>p^*$, and one finds from Eq. (19),

$$G_{eq}^{\delta\mathcal{L}}(\tau) = \pi\Gamma\left(\frac{3+n}{2+n}\right)\left(\frac{k_B T}{\sigma}\right)^2 L(2A\tau)^{-1/(2+n)}. \tag{30}$$

Considering that h is the local orientation along the arclength x, a description of a filament (such as an actin filament or a microtubule) confined in a Hele-Shaw cell [1] is obtained. In such a case, $\kappa = \partial_x \zeta$ is the curvature, $\mathcal{E}$ is the bending energy, and σ is the bending modulus.

The normal velocity v_n of the filament in the cell plane is related to the dynamics of the local orientation ζ via the geometric relation $\partial_t \zeta = v_t \kappa - \partial_x v_n$, where v_t is the tangential velocity. The driving force is now $\delta\mathcal{E}/\delta u \sim \partial_{xx}\kappa$, where u is the filament position. Neglecting the tangential velocity and assuming simple overdamped dynamics $v_n \sim \partial_{xx}\kappa$, which is linearized as $\partial_t \zeta \sim -\partial_{xxxx}\zeta$, leading to $n=2$, as confirmed by experiments [1].

Using Eq. (16) with $p=0$, it is found that the distortions ζ will be small when $L<L_\sigma$, where $L = \int dx$ is now the fixed total arclength of the polymer, and $L_\sigma = \sigma/k_B T$ is the persistence length. The end-to-end length $\mathcal{L}_{ee}$ is then related to Q_0 via

$$\mathcal{L}_{ee}(t) = L - \frac{Q_0}{2}. \tag{31}$$

Thus, $G_{eq}^{\delta\mathcal{L}_{ee}}(\tau) = G_{eq}^{\delta Q_0}(\tau)/4$, and using Eq. (25) with $n=2$, one finds

$$F_{eq}^{\delta\mathcal{L}_{ee}}(\tau) \sim \tau^{3/4}L. \tag{32}$$

We here recover a result which was already derived in the literature [6]. Our approach allows one to place this result within a systematic framework. For example, other quantities, such as the time correlations of the total curvature energy $\mathcal{E}=\sigma Q_1/2$, may be derived as $G_{eq}^{\delta\mathcal{E}} \sim L\tau^{-1/4}$.

As a summary, the time-correlation function of some macroscopic quantities in a 1D system can be simply expressed as a function of local-correlation functions in thermodynamic equilibrium. The correlations of macroscopic quantities allows one to explore a universal diffusive regime, which could not be observed by means of local-correlation functions. We have mentioned several systems where our results could be relevant.

The author would like to thank the hospitality of Professor J. Yeomans at the Rudolf Peierls Centre for Theoretical Physics, where this work was performed.

---

[1] C. Brangwynne, G. H. Koenderink, E. Barry, Z. Dogic, F. C. MacKintosh, and D. A. Weitz, Biophys. J. **93**, 346 (2007).

[2] H.-C. Jeong and E. D. Williams, Surf. Sci. Rep. **34**, 171 (1999).

[3] A. K. Schmid, K. L. Man, N. C. Bartelt, H. Poppa, and M. S. Altman, Microsc. Microanal. **9**, 134 (2003).

[4] E. D. Williams, A. Bondarchuk, C. G. Tao, W. Yan, W. G. Cullen, P. J. Rous, and T. Bole, New J. Phys. **9**, 387 (2007).

[5] J. Wilhelm and E. Frey, Phys. Rev. Lett. **77**, 2581 (1996).

[6] R. Granek, J. Phys. II **7**, 1761 (1997).

[7] Since dynamics is linear and stable, the evolution from an arbitrary initial condition will be the sum of the decay of this initial condition and the evolution from a flat initial condition.

[8] M. C. Wang and G. E. Uhlenbeck, Rev. Mod. Phys. **17**, 323 (1945).

[9] For arbitrary times, using the Schwarz inequality, for any t, and t': $G^{\delta Q_p}(t,t') \geq 8\pi^2\,\mathrm{L}a[G^{\partial_x^p \zeta}(t,t')]^2$, where a is a microscopic cutoff.

[10] Formally, $m=1/2$ corresponds to the case where $G_{eq}^{\partial_x^m \zeta}(0)$ diverges logarithmically, in an extended definition of $G_{eq}^{\partial_x^m \zeta}(\tau)$ based on Eq. (6).

[11] In order to obtain explicit analytic formulas, we approximate the sum over k with a continuous integration, and the size L is here simply introduced by considering a cutoff at $k=2\pi/L$. While this approach gives the correct scaling behavior, the numerical prefactors may not be accurate.

[12] Nevertheless, the diffusive behavior of the TCFs must be distinguished from the diffusion of the center of mass of the system [16]. The position of the center of mass is $h_{k=0}(t)/L$. Its diffusion results from the fact that $i\omega_{k=0}=0$, so that $\partial_t h_{k=0} = \eta_{k=0}$. Solving this equation, one finds $\langle[h_{k=0}(t+\tau) - h_{k=0}(t)]^2\rangle = B_{k=0}\tau$, so that $B_{k=0}$ is the diffusion constant of the center of mass. Using Eq. (14), $B_{k=0}=(4\pi A k_B T/\sigma)$ for $n=0$, and $B_{k=0}=0$ for $n>1$. Taking the example of atomic steps, we recover the fact that the center of mass of a straight step should diffuse for $n=0$ (nonconserved, or Glauber dynamics), and cannot move due to mass conservation for $n=2$ (conserved, or Kawasaki dynamics). Since ζ—defined in Eq. (4)—does not contain the mode $k=0$, the quantities Q_p, which are functionals of ζ, also do not account for this mode. (In order to be more explicit, we may define $R_0=\int dx h^2$ by analogy with Q_0. We then find $F^{\delta R_0}(\tau)=F^{\delta Q_0}(\tau)+B_{k=0}^2(2t+\tau)\tau$. Hence, we see explicitly the consequences of the diffusion of the center of mass in the term $B_{k=0}^2(2t+\tau)\tau$.)

[13] S. V. Khare and T. L. Einstein, Phys. Rev. B **57**, 4782 (1998).

[14] O. Pierre-Louis, Phys. Rev. Lett. **87**, 106104 (2001).

[15] T. Ihle, C. Misbah, and O. Pierre-Louis, Phys. Rev. B **58**, 2289 (1998).

[16] S. V. Khare and T. L. Einstein, Phys. Rev. B **54**, 11752 (1996); S. V. Khare, N. C. Bartelt, and T. L. Einstein, Phys. Rev. Lett. **75**, 2148 (1995).