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LETTER

On anomalous diffusion and the out of equilibrium response function in one-dimensional models

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Abstract. We study how the Einstein relation between spontaneous fluctuations and the response to an external perturbation holds in the absence of currents, for the comb model and the elastic single-file, which are examples of systems with subdiffusive transport properties. The relevance of non-equilibrium conditions is investigated: when a stationary current (in the form of a drift or an energy flux) is present, the Einstein relation breaks down, as is known to happen in systems with standard diffusion. In the case of the comb model, a general relation, which has appeared in the recent literature, between the response function and an unperturbed suitable correlation function, allows us to explain the observed results. This suggests that a relevant ingredient in breaking the Einstein formula, for stationary regimes, is not the anomalous diffusion but the presence of currents driving the system out of equilibrium.

Keywords: stochastic particle dynamics (theory), fluctuations (theory), transport properties (theory), diffusion

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Contents

1. Introduction

In his seminal paper on Brownian motion, Einstein, beyond the celebrated relation between the diffusion coefficient D and the Avogadro number, found the first example of the fluctuation-dissipation relation (FDR). In the absence of external forcing one has, for large times $t \to \infty$,

$$\langle x(t)\rangle = 0, \qquad \langle x^2(t)\rangle \simeq 2Dt, \tag{1}$$

where x is the position of the Brownian particle and the average is taken over the unperturbed dynamic. Once a small constant external force F is applied one has a linear drift

$$\overline{\delta x}(t) = \langle x(t)\rangle_F - \langle x(t)\rangle \simeq \mu F t, \tag{2}$$

where $\langle \cdots \rangle_F$ indicates the average on the perturbed system, and μ is the mobility of the colloidal particle. It is remarkable that $\langle x^2(t)\rangle$ is proportional to $\overline{\delta x}(t)$ at any time:

$$\frac{\langle x^2(t)\rangle}{\overline{\delta x}(t)} = \frac{2}{\beta F}, \tag{3}$$

and the Einstein relation (a special case of the fluctuation-dissipation theorem [1]) holds: $\mu = \beta D$, with $\beta = 1/k_B T$ the inverse temperature and k_B the Boltzmann constant.

On the other hand it is now well established that beyond the standard diffusion, as in (1), one can have systems with anomalous diffusion (see for instance [2]–[6]), i.e.

$$\langle x^2(t)\rangle \sim t^{2\nu} \qquad \text{with } \nu \neq 1/2. \tag{4}$$

Formally this corresponds to having $D = \infty$ if $\nu > 1/2$ (superdiffusion) and $D = 0$ if $\nu < 1/2$ (subdiffusion). In this letter we will limit the study to the case $\nu < 1/2$. It is quite natural to wonder if (and how) the FDR changes in the presence of anomalous diffusion, i.e. if instead of (1), equation (4) holds. In some systems it has been shown that (3) holds even in the subdiffusive case. This has been explicitly proved in systems described by a fractional Fokker–Planck equation [7], see also [8, 9]. In addition there is clear analytical [10] and numerical [11] evidence that (3) is valid for the elastic single-file, i.e. a gas of hard rods on a ring with elastic collisions, driven by an external thermostat, which exhibits subdiffusive behaviour, $\langle x^2\rangle \sim t^{1/2}$ [12].

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The aim of this paper is to discuss the validity of the fluctuation-dissipation relation in the form (3) for systems with anomalous diffusion which are not fully described by a fractional Fokker–Planck equation. In particular we will investigate the relevance of the anomalous diffusion, the presence of non-equilibrium conditions and the (possible) role of finite size. Since we are also interested in the study of transient regimes, we will consider models with microscopic dynamics described in terms of transition rates or microscopic interactions.

First, we focus on the study of a particle moving on a 'finite comb' lattice with teeth of size L [13]. In the limit $L = \infty$ an anomalous subdiffusive behaviour, $\langle x^2 \rangle \sim t^{1/2}$, holds and the system can be mapped, for large times, onto a continuous time random walk [13]. For finite L the subdiffusion is only transient and at very large times $t > t^*(L) \sim L^2$ one has a standard diffusion: $\langle x^2 \rangle \sim t$. We will see that equation (3), where in this case the perturbed average is obtained with unbalanced transition rates driving the particle along the backbone of the comb, holds both for $t > t^*(L)$ and $t < t^*(L)$ with the same constant. This in spite of the fact that the probability densities $P(x,t)$ in the two regimes are very different. The scenario changes in the presence of 'non-equilibrium' conditions, i.e. with a drift, which induces a current in the unperturbed state: the relation (3) does not hold any more. On the other hand, in this case it is possible to use a generalized fluctuation-dissipation relation, derived by Lippiello *et al* in [14], which gives the response function in terms of unperturbed correlation functions and is an example of non-equilibrium FDR valid under rather general conditions [14]–[21]. A generalization of the Einstein formula was also proved in the framework of continuous time random walks in [22]. So we can say that the Einstein relation (3) also holds in cases with anomalous diffusion when no current is present, but it is necessary to introduce suitable corrections when a perturbation is applied to a system with non-zero drift.

In addition we compare the results found in comb models with those obtained for single-file diffusion with a finite number of particles. We will also consider a non-equilibrium case, with the introduction of inelastic collisions which induce an energy flux crossing the system. Our results suggest that the presence of non-equilibrium currents plays a relevant role in modifying equation (3) in stationary states.

2. Comb: diffusion and response function

The comb lattice is a discrete structure consisting of an infinite linear chain (backbone), the sites of which are connected with other linear chains (teeth) of length L [13]. We denote by $x \in (-\infty, \infty)$ the position of the particle performing the random walk along the backbone and by $y \in [-L, L]$ that along a tooth. The transition probabilities from (x, y) to (x', y') are

$$\begin{aligned} &W^d[(x,0) \to (x \pm 1, 0)] = 1/4 \pm d \\ &W^d[(x,0) \to (x, \pm 1)] = 1/4 \\ &W^d[(x,y) \to (x, y \pm 1)] = 1/2 \qquad \text{for } y \neq 0, \pm L. \end{aligned} \tag{5}$$

On the boundaries of each tooth, $y = \pm L$, the particle is reflected with probability 1. The case $L = \infty$ is obtained in numerical simulations by letting the y coordinate increase without boundaries. Here we consider a discrete time process and, of course, the

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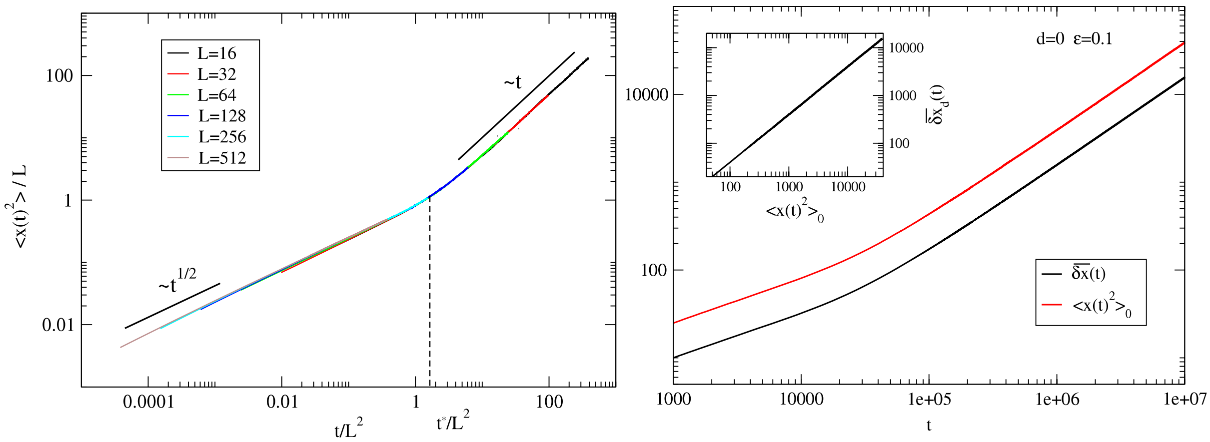

Figure 1. Left panel: $\langle x^2(t)\rangle_0/L$ versus t/L^2 is plotted for several values of L in the comb model. Right panel: $\langle x^2(t)\rangle_0$ and the response function $\overline{\delta x}(t)$ for $L = 512$. In the inset the parametric plot of $\overline{\delta x}(t)$ versus $\langle x^2(t)\rangle_0$ is shown.

normalization $\sum_{(x',y')} W^d[(x,y) \to (x',y')] = 1$ holds. The parameter $d \in [0, 1/4]$ allows us to consider also the case where a constant external field is applied along the x axis, producing a non-zero drift of the particle. A state with a non-zero drift can be considered as a perturbed state (in which case we denote the perturbing field by ε), or it can be itself the starting state where a further perturbation can be added changing $d \to d + \varepsilon$.

Let us start by considering the case $d = 0$. For finite tooth length $L < \infty$, we have numerical evidence of a dynamical crossover from a subdiffusive to a simple diffusive asymptotic behaviour (see figure 1):

$$\langle x^2(t)\rangle_0 \simeq \begin{cases} Ct^{1/2} & t < t^*(L) \\ 2D(L)t & t > t^*(L), \end{cases} \tag{6}$$

where C is a constant and $D(L)$ is an effective diffusion coefficient depending on L. The symbol $\langle \cdots \rangle_0$ denotes an average over different realizations of the dynamics (5) with $d = 0$ and initial condition $x(0) = y(0) = 0$. We find $t^*(L) \sim L^2$ and $D(L) \sim 1/L$, and in the left panel of figure 1 we plot $\langle x^2(t)\rangle_0/L$ as a function of t/L^2 for several values of L, showing an excellent data collapse.

In the limit of infinite teeth, $L \to \infty$, $D \to 0$ and $t^* \to \infty$ and the system shows a pure subdiffusive behaviour [23]:

$$\langle x^2(t)\rangle_0 \sim t^{1/2}. \tag{7}$$

In this case, the probability distribution function behaves as

$$P_0(x,t) \sim t^{-1/4} \mathrm{e}^{-c\left(|x|/t^{1/4}\right)^{4/3}}, \tag{8}$$

where c is a constant, in agreement with an argument *à la* Flory [2]. The behaviour (8) also holds in the case of finite L, provided that $t < t^*$. For larger times a simple Gaussian distribution is observed. Note that, in general, the scaling exponent ν, in this case $\nu = 1/4$, does not determine univocally the shape of the pdf. Indeed, for the single-file model, discussed below, we have the same ν but the pdf is Gaussian [24].

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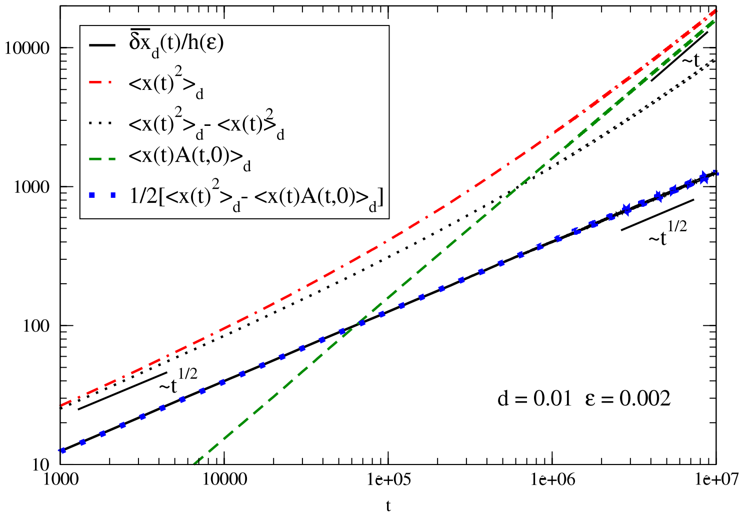

Figure 2. Response function (black line), msd (red dotted line) and second cumulant (black dotted line) measured in the comb model with $L = \infty$, field $d = 0.01$ and perturbation $\varepsilon = 0.002$. The correlation with activity (green dotted line) yields the right correction to recover the full response function (blue dotted line), in agreement with the FDR (15).

In the comb model with infinite teeth, the FDR in its standard form is fulfilled, namely if we apply a constant perturbation ε pulling the particles along the 1d lattice one has numerical evidence that

$$\langle x^2(t)\rangle_0 \simeq C\overline{\delta x}(t) \sim t^{1/2}. \tag{9}$$

In the following section we derive this result from a generalized FDR. Moreover, the proportionality between $\langle x^2(t)\rangle_0$ and $\overline{\delta x}(t)$ is fulfilled also with $L < \infty$, where both the mean square displacement (msd) and the drift with an applied force exhibit the same crossover from subdiffusive, $\sim t^{1/2}$, to diffusive, $\sim t$ (see figure 1, right panel). Therefore what we can say is that the FDR is somehow 'blind' to the dynamical crossover experienced by the system. When the perturbation is applied to a state without any current, the proportionality between response and correlation holds despite anomalous transport phenomena.

Our aim here is to show that, differently from what was depicted above for the zero current situation, within a state with a non-zero drift [25] the emergence of a dynamical crossover is connected to the breaking of the FDR. Indeed, the msd in the presence of a non-zero current, even with $L = \infty$, shows a dynamical crossover

$$\langle x^2(t)\rangle_d \sim a\, t^{1/2} + b\, t, \tag{10}$$

where a and b are two constants, whereas

$$\overline{\delta x}_d(t) \sim t^{1/2}, \tag{11}$$

with $\overline{\delta x}_d(t) = \langle x(t)\rangle_{d+\varepsilon} - \langle x(t)\rangle_d$: at large times the Einstein relation breaks down (see figure 2). The proportionality between response and fluctuations cannot be recovered by simply replacing $\langle x^2(t)\rangle_d$ with $\langle x^2(t)\rangle_d - \langle x(t)\rangle_d^2$, as happens for Gaussian processes (see

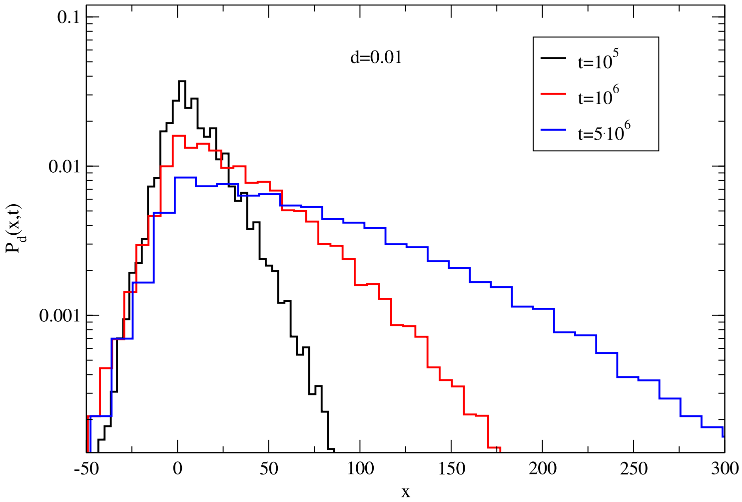

Figure 3. $P_d(x,t)$ in the comb model with $L=\infty$ and $d=0.01$ at different times. Notice that the mean value increases with time mostly due to the spreading, while the most probable value remains always close to zero.

discussion below), namely we find numerically

$$\langle [x(t) - \langle x(t)\rangle_d]^2\rangle_d \sim a'\ t^{1/2} + b'\ t, \tag{12}$$

where a' and b' are two constants, as reported in figure 2.

3. Comb: application of a generalized FDR

The discussion of section 2 shows that the first moment of the probability distribution function with drift $P_d(x,t)$ and the second moment of $P_0(x,t)$ are always proportional. Note that in the presence of a drift the pdf is strongly asymmetric with respect to the mean value, as shown in figure 3 for a system with $L=\infty$. In contrast, the first moment of $P_{d+\varepsilon}(x,t)$ is not proportional to the second moment of $P_d(x,t)$, namely $\langle x(t)\rangle_{d+\varepsilon} \nsim \langle x^2(t)\rangle_d - \langle x(t)\rangle_d^2$. In order to find out a relation between such quantities, we need a generalized fluctuation-dissipation relation.

According to the definition (5), one has for the backbone

$$W^{d+\varepsilon}[(x,y)\to(x',y')] = W^d[(x,y)\to(x',y')]\left(1+\frac{\varepsilon(x'-x)}{W^0+d(x'-x)}\right) \simeq W^d \mathrm{e}^{(\varepsilon/W^0)(x'-x)}, \tag{13}$$

where $W^0=1/4$, and the last expression holds under the condition $d/W^0 \ll 1$. Regarding the above expression as a *local detailed balance* condition for our Markov process we can rewrite it, for $(x,y)\neq(x',y')$, as

$$W^{d+\varepsilon}[(x,y)\to(x',y')] = W^d[(x,y)\to(x',y')]\mathrm{e}^{(h(\varepsilon)/2)(x'-x)}, \tag{14}$$

where $h(\varepsilon)=2\varepsilon/W^0$. For general models where the perturbation enters the transition probabilities according to equation (14), the following formula for the integrated linear response function has been derived [14, 19, 21]:

$$\frac{\overline{\delta\mathcal{O}}_d}{h(\varepsilon)} = \frac{\langle\mathcal{O}(t)\rangle_{d+\varepsilon} - \langle\mathcal{O}(t)\rangle_d}{h(\varepsilon)} = \frac{1}{2}\left[\langle\mathcal{O}(t)x(t)\rangle_d - \langle\mathcal{O}(t)x(0)\rangle_d - \langle\mathcal{O}(t)A(t,0)\rangle_d\right], \tag{15}$$

where $\mathcal{O}$ is a generic observable, and $A(t,0) = \sum_{t'=0}^{t} B(t')$, with

$$B[(x,y)] = \sum_{(x',y')} (x'-x)W^d[(x,y)\to(x',y')]. \tag{16}$$

The above observable yields an effective measure of the propensity of the system to leave a certain state (x,y) and, in some contexts, it is referred to as *activity* [26]. Recalling the definitions (5), from the above equation we have $B[(x,y)] = 2d\delta_{y,0}$ and therefore the sum on B has an intuitive meaning: it counts the time spent by the particle on the x axis. The results described in section 2 can be then read in the light of the fluctuation-dissipation relation (15).

(i) Putting $\mathcal{O}(t) = x(t)$, in the case without drift, i.e. $d = 0$, one has $B = 0$ and, recalling the choice of the initial condition $x(0) = 0$,

$$\frac{\overline{\delta x}}{h(\varepsilon)} = \frac{\langle x(t)\rangle_\varepsilon - \langle x(t)\rangle_0}{h(\varepsilon)} = \frac{1}{2}\langle x^2(t)\rangle_0. \tag{17}$$

This explains the observed behaviour (9) even in the anomalous regime and predicts the correct proportionality factor, $\overline{\delta x}(t) = \varepsilon/W^0\langle x^2(t)\rangle_0$.

(ii) Putting $\mathcal{O}(t) = x(t)$, in the case with $d \neq 0$, one has

$$\frac{\overline{\delta x}_d}{h(\varepsilon)} = \frac{1}{2}\left[\langle x^2(t)\rangle_d - \langle x(t)A(t,0)\rangle_d\right]. \tag{18}$$

This explains the observed behaviours (10) and (11): the leading behaviour at large times of $\langle x^2(t)\rangle_d \sim t$ turns out to be exactly cancelled by the term $\langle x(t)A(t,0)\rangle_d$, so that the relation between response and unperturbed correlation functions is recovered (see figure 2).

(iii) As discussed above, it is not enough to substitute $\langle x^2(t)\rangle_d$ with $\langle x^2(t)\rangle_d - \langle x(t)\rangle_d^2$ to recover the proportionality with $\overline{\delta x}_d(t)$ when the process is not Gaussian. This can be explained in the following manner. By making use of the second order out of equilibrium FDR derived by Lippiello *et al* in [27]–[29], which is needed due to the vanishing of the first order term for symmetry, we can explicitly evaluate

$$\langle x^2(t)\rangle_d = \langle x^2(t)\rangle_0 + h^2(d)\tfrac{1}{2}\left[\tfrac{1}{4}\langle x^4(t)\rangle_0 + \tfrac{1}{4}\langle x^2(t)A^{(2)}(t,0)\rangle_0\right], \tag{19}$$

where $A^{(2)}(t,0) = \sum_{t'=0}^{t} B^{(2)}(t')$ with $B^{(2)} = -\sum_{x'}(x'-x)^2 W[(x,y)\to(x',y')] = -1/2\delta_{y,0}$. Then, recalling equation (17), we obtain

$$\langle x^2(t)\rangle_d - \langle x(t)\rangle_d^2 = \langle x^2(t)\rangle_0 + h^2(d)\left[\tfrac{1}{8}\langle x^4(t)\rangle_0 + \tfrac{1}{8}\langle x^2(t)A^{(2)}(t,0)\rangle_0 - \tfrac{1}{4}\langle x^2(t)\rangle_0^2\right]. \tag{20}$$

Numerical simulations show that the term in the square brackets grows like t yielding a scaling behaviour with time consistent with equation (12). On the other hand, in the case of the simple random walk, one has $B^{(2)} = -1$ and $A^{(2)}(t,0) = -t$ and then

$$\langle x^2(t)\rangle_d - \langle x(t)\rangle_d^2 = \langle x^2(t)\rangle_0 + h^2(d)\left[\tfrac{1}{8}\langle x^4(t)\rangle_0 - \tfrac{1}{8}t\langle x^2(t)\rangle_0 - \tfrac{1}{4}\langle x^2(t)\rangle_0^2\right]. \tag{21}$$

Since in the Gaussian case $\langle x^4(t)\rangle_0 = 3\langle x^2(t)\rangle_0^2$ and $\langle x^2(t)\rangle_0 = t$, the term in the square brackets vanishes identically and that explains why, in the presence of a drift, the second cumulant grows exactly as the second moment with no drift.

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4. Conclusions and perspectives

In order to evaluate the generality of the above results, let us conclude by discussing another system. Indeed, subdiffusion is present in many different problems where geometrical constraints play a central role. In this framework, a well studied phenomenon is the so-called single-file diffusion. Namely, we have N Brownian rods on a ring of length L interacting with elastic collisions and coupled with a thermal bath. The equation of motion for the single particle velocity between collisions is

$$m\dot{v}(t) = -\gamma v(t) + \eta(t), \tag{22}$$

where m is the mass, γ is the friction coefficient, and η is a white noise with variance $\langle \eta(t)\eta(t')\rangle = 2T\gamma\delta(t-t')$. The combined effect of collisions, noise and geometry (since the system is one-dimensional the particles cannot overcome each other) produces a non-trivial behaviour. In the thermodynamic limit, i.e. L, $N \to \infty$ with $N/L \to \rho$, a subdiffusive behaviour occurs [12].

Analogously to the comb model, the case of N and L finite presents some interesting aspects. In order to avoid trivial results due to the periodic boundary conditions on the ring, it is suitable to define the position of a tagged particle as $s(t) = \int_0^t v(t')\,\mathrm{d}t'$, where $v(t)$ is its velocity. For the msd $\langle s^2(t)\rangle$, averaged over the thermalized initial conditions and over the noise, we find, after a transient ballistic behaviour for short times, a dynamical crossover between two different regimes:

$$\langle s^2(t)\rangle \simeq \begin{cases} \dfrac{2(1-\sigma\rho)}{\rho}\sqrt{\dfrac{D}{\pi}}\,t^{1/2} & t < \tau^*(N) \\ \dfrac{2D}{N}t & t > \tau^*(N), \end{cases} \tag{23}$$

where σ is the length of the rods and D is the diffusion coefficient of the single Brownian particle [12]. Note that the asymptotic behaviour is completely determined by the motion of the centre of mass, which is not affected by the collisions and simply diffuses. Moreover, as evident from numerical simulations, $\tau^* \sim N^2$, and in the limit of infinite number of particles the behaviour becomes subdiffusive, in perfect analogy with what observed for the comb model, where the role of L is here played by N. The main difference is that, in this case, the probability distribution is Gaussian in both regimes. As a consequence of the Gaussian nature of the problem, applying a perturbation as a small force F in equation (22), one finds that the Einstein relation is always fulfilled [10, 11, 18, 30], also for finite N and L (see figure 4). Strong violations of the Einstein relation can be obtained in dense cases, when the collisions between the rods are inelastic so that a homogeneous energy current crosses the system [11].

In this letter we have considered systems with subdiffusive behaviour, showing that the proportionality between response function and correlation breaks down when 'non-equilibrium' conditions are introduced. In the comb model, non-equilibrium effects are induced by unbalanced transition probabilities driving the particle along the backbone, while the single-file model is driven away from equilibrium by inelastic collisions. In the first case, the generalized FDR of equation (15), developed in the framework of ageing systems [14], can be explicitly written, providing the off equilibrium corrections to the Einstein relation. In the second case, the transition rates are not known and

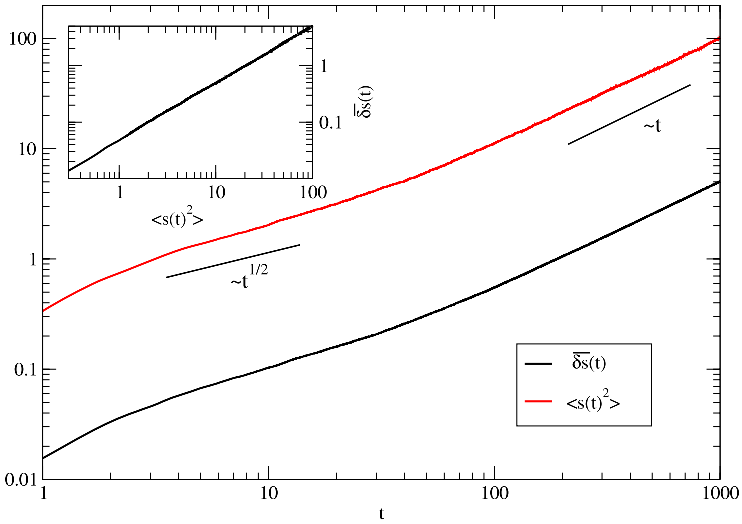

Figure 4. $\langle s^2(t)\rangle$ and the response function $\overline{\delta s}(t)$ for the single-file model with the parameters $N = 10$, $L = 10$, $\sigma = 0.1$, $m = 1$, $\gamma = 2$, $T = 1$ and perturbation $F = 0.1$. In the inset the parametric plot of $\overline{\delta s}(t)$ versus $\langle s^2(t)\rangle$ is shown.

another formalism must be exploited [18], which requires the knowledge of the probability distribution for the relevant dynamical variables of the model. For instance, following the ideas of [11], a distribution which couples the velocities of neighbouring particles could be a reasonable guess. Still, the identification of the relevant variables and their coupling in the single-file and other granular systems is a central issue, requiring further investigations.

Acknowledgments

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