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LETTER

Einstein relation in superdiffusive systems

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Abstract. We study the Einstein relation between diffusion and response to an external field in systems showing superdiffusion. In particular, we investigate a continuous time Lévy walk where the velocity remains constant for a time τ with distribution $P_\tau(\tau) \sim \tau^{-g}$. At varying g the diffusion can be standard or anomalous; in spite of this, if in the unperturbed system a current is absent, the Einstein relation holds. In the case where a current is present the scenario is more complicated and the usual Einstein relation fails. This suggests that the main ingredient for the breaking of the Einstein relation is not the anomalous diffusion but the presence of a mean drift (current).

Keywords: driven diffusive systems (theory), stochastic particle dynamics (theory), transport processes/heat transfer (theory), current fluctuations

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1. Introduction

The fluctuation-dissipation relation (FDR) is one of the fundamental results of statistical physics [1]. In his celebrated work on Brownian motion, Einstein gave the first example of FDR. In the absence of external forcing, at large times, one has

$$\langle x(t) \rangle = 0 \quad \text{and} \quad \langle x^2(t) \rangle \simeq 2Dt, \quad (1)$$

where x is the position of the colloidal particle, D is the diffusion coefficient and the average refers to the unperturbed dynamics. If a small constant external force \mathcal{E} is applied, one obtains a linear drift

$$\langle x(t) \rangle_{\mathcal{E}} \simeq \mu \mathcal{E} t, \quad (2)$$

where $\langle \cdot \cdot \cdot \rangle_{\mathcal{E}}$ is the average on the perturbed system, and μ is the mobility. The remarkable result obtained by Einstein is the proportionality between $\langle x(t) \rangle_{\mathcal{E}}$ and the mean square displacement (MSD) $\langle x^2(t) \rangle$:

$$\frac{\langle x^2(t) \rangle}{\langle x(t) \rangle_{\mathcal{E}}^2} = \frac{2}{\beta \mathcal{E}}, \quad (3)$$

namely $\mu = \beta D$, where β is the inverse temperature and we have taken the Boltzmann constant $k_{\text{B}} = 1$.

In the last few decades much research has been devoted to the study of anomalous diffusion, where, instead of (1), one has

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

see for instance [2]–[7]. The case $\nu < 1/2$ is called subdiffusion while if $\nu > 1/2$ we are in the presence of superdiffusion. It is quite natural to wonder about the validity of the FDR in these anomalous situations. Important results showing the proportionality between $\langle x(t) \rangle_{\mathcal{E}}$ and $\langle x^2(t) \rangle$, which we refer to as the Einstein relation ‘at equilibrium’, have already been obtained for both superdiffusive [8, 9, 7] and subdiffusive [2, 10, 7] anomalous dynamics. Differently, the situation where the drift is compared to the mean square

displacement in a state which is already ‘out of equilibrium’, either due to a current [11] or due to dissipation [12], has been studied only in the subdiffusive cases.

The aim of this letter is to discuss the validity of the Einstein relation in equilibrium and out-of-equilibrium situations in the presence of superdiffusive dynamics. In particular, we consider a Lévy walk collision process [13] and we show that the Einstein relation is violated when the perturbation is applied on a reference state where a current is already present.

2. The model

We consider an ensemble of probe particles of mass m endowed with scalar velocity v and position x . Each probe particle only interacts with particles of mass M and velocity V extracted from an equilibrium bath at temperature T . We assume that the scattering probability does not depend on the relative velocity between the probe particle and the colliders, as, for instance, in the case of Maxwell-molecule models [14]. The velocity of the probe particle changes from v to v' at each collision, according to the rule:

$$v' = \gamma v + (1 - \gamma)V, \quad (5)$$

where $\gamma = (\zeta - \alpha)/(1 + \zeta)$, with $\zeta = m/M$, and α is the coefficient of restitution ($\alpha = 1$ for an elastic collision). The velocity V of the bath particles is a random variable generated from a Gaussian distribution with zero mean and variance T/M :

$$P_S(V) = \sqrt{\frac{M}{2\pi T}} \exp\left\{-\frac{M}{2T}V^2\right\}. \quad (6)$$

The elementary steps of the dynamics are made by: (i) a flight, $x(t + \tau) = x(t) + v'\tau + 1/2\mathcal{E}\tau^2$, where $x(t)$ is the position of the probe particle at time t , with τ taken from a distribution $P_\tau(\tau)$ and \mathcal{E} a constant acceleration, followed by (ii) a collision $v' = \gamma v + (1 - \gamma)V$, with V taken from the Gaussian distribution (6). In the specific case $\alpha = 1$ and $M = m$, one has $\gamma = 0$ and the collision rule (5) results in a random update of the velocity according to the distribution (6). The duration of each flight, τ , is an independent identically distributed random variable with probability

$$P_\tau(\tau) \sim \tau^{-g}, \quad (7)$$

with $g > 1$. This kind of process is called a Lévy walk collision process [13], and *may* be interpreted as due to scattering centers randomly distributed on a fractal spatial structure, such as for instance in the case of molecular diffusion in porous media [15]. If $\alpha \neq 1$ or $m \neq M$, with $\alpha \neq \zeta$, a dependence on the last velocity before the collision remains. In this case velocity correlations can be measured in the system, as discussed in section 3.1.

According to the dynamic rules of the process described above, the displacement of the probe particle is always finite in a finite time. The anomalous dynamics of such a model has been studied in [16], showing that the process is an example of ‘strong’ anomalous diffusion, namely that it is not possible to find a scaling for the probability density function (PDF) of displacements. Such a collision process becomes a standard diffusive system when P_τ decays fast enough: in this regime the dynamics is qualitatively equivalent to that of a system with exponential P_τ , studied for instance in [17]–[19]. Lévy

walk collision processes have been thoroughly studied, see for instance [7], where the behavior of the mean square displacement has been obtained analytically.

We recall here a simple argument already presented in [16] to study the asymptotic behavior of higher order moments of displacement and that can be easily applied also to the case with an external perturbing field that we are going to discuss here in section 3. In order to obtain in a simple way the dominant asymptotic behavior of $\langle x^2(t) \rangle$, we introduce a cut-off t_c for $P_\tau(\tau)$:

$$P_\tau(\tau) \sim \begin{cases} \tau^{-g} & \text{if } \tau < t_c \\ 0 & \text{if } \tau > t_c. \end{cases} \quad (8)$$

Assuming $x = 0$ as the initial condition for each trajectory, the mean square displacement after time t , where $N(t)$ collisions occurred, can be written in full generality as:

$$\langle x^2(t) \rangle = \left\langle \left[\sum_{i=1}^{N(t)} v_i \tau_i \right]^2 \right\rangle \simeq \sum_{i=1}^{\bar{N}(t)} \langle v_i^2 \tau_i^2 \rangle + 2\bar{N}(t) \sum_{i=1}^{\bar{N}(t)} \langle v_i v_0 \tau_i \tau_0 \rangle. \quad (9)$$

Here v_i denotes the velocity of the probe particle after the i th collision, τ_i is the time elapsed between the collisions i and $i + 1$ and $\bar{N}(t)$ is the average number of collisions occurring up to time t . The average $\langle \dots \rangle$ is taken over the distributions (6) and (7). From equation (8) we have, for $n + 1 - g > 0$,

$$\langle \tau^n \rangle_c \sim t_c^{n+1-g}, \quad (10)$$

where $\langle \dots \rangle_c$ denotes an average over the distribution (8) with the cut-off t_c .

We start by considering the case with independent velocities v_i , corresponding to a choice of parameters such that $\gamma = 0$. Then, estimating $\langle x^2(t) \rangle$ at a time $t \gg t_c$, so that the average number of collisions along the trajectory can be approximated to $\bar{N}(t) \simeq t/\langle \tau \rangle_c$, and considering that the cross terms in equation (9) are zero, we can write

$$\langle x^2(t) \rangle \simeq \frac{t}{\langle \tau \rangle_c} \langle v^2 \rangle \langle \tau^2 \rangle_c. \quad (11)$$

In the case $g > 3$, both $\langle \tau \rangle_c$ and $\langle \tau^2 \rangle_c$ are finite, even in the limit $t_c \rightarrow \infty$, so that we find the simple diffusive behavior $\langle x^2(t) \rangle \sim t$. For $t \lesssim t_c$ and $1 < g < 3$, instead of (11) we expect

$$\langle x^2(t) \rangle \sim t^{2\nu}. \quad (12)$$

One can easily find the exponent ν with a matching argument. Comparing (11) and (12) at $t \sim t_c$ and using (10), we obtain $\nu = 1/2$ for $g > 3$, $\nu = 2 - g/2$ for $2 < g < 3$, and $\nu = 1$ for $1 < g < 2$ (logarithmic corrections appear at the values $g = 2$ and 3 [7]).

3. Einstein relation

The main concern of our study is the question about the validity of the Einstein relation for superdiffusive anomalous dynamics. In particular such a relation can be checked in two different situations.

- (A) The drift due to the external force is compared with the MSD of the probe particle in the absence of any pulling force. This case corresponds to a fluctuation-dissipation experiment realized by switching on from zero the external perturbation.
- (B) The drift is compared to the MSD in a state where a current is already present. This procedure corresponds to increasing the intensity of the perturbation in a state already perturbed and comparing the average current in this state with the fluctuations in the initial reference state.

In the following, we will refer to situation (A) as a test of the fluctuation-dissipation relation at *equilibrium* while to case (B) as a test *out of equilibrium*. We will show that these two cases are very different.

3.1. Perturbation of a state without current

The argument used in section 2 to study the MSD can be applied to the drift, yielding

$$\langle x(t) \rangle_{\mathcal{E}} = \left\langle \sum_{i=1}^{N(t)} \left(v_i \tau_i + \frac{\mathcal{E}}{2} \tau_i^2 \right) \right\rangle = \frac{t}{\langle \tau \rangle_c} \left[\langle \tau \rangle_c \langle v \rangle + \frac{\mathcal{E}}{2} \langle \tau^2 \rangle_c \right] = \frac{\mathcal{E}}{2} \frac{t}{\langle \tau \rangle_c} \langle \tau^2 \rangle_c, \quad (13)$$

which perfectly matches the result for the MSD found in equation (11). Therefore, when we perturb an equilibrium state, namely a state without currents, we have for any value of $g > 1$

$$\frac{\langle x^2(t) \rangle}{\langle x(t) \rangle_{\mathcal{E}}^2} = \text{const.} \quad (14)$$

Let us note that the Einstein relation holds quite generally, namely it persists even if we make our process non-Markovian by allowing some memory across collisions, that is, we put $\gamma > 0$ in the collision rule equation (5), preventing a complete reshuffling of velocities. In this case the MSD reads as:

$$\langle x^2(t) \rangle = \sum_{i,j}^{\bar{N}(t)} \langle v_i \tau_i v_j \tau_j \rangle \sim \frac{t}{\langle \tau \rangle_c} \left[\langle v^2 \rangle \langle \tau^2 \rangle_c + 2 \langle \tau \rangle_c^2 \sum_i^{\bar{N}(t)} \langle v_i v_0 \rangle \right], \quad (15)$$

because the collision times are still not dependent on the velocity but correlations between the velocities must be taken into account. Note that, even in the presence of non-independent $\{v_i\}$, if $\sum_i^n \langle v_i v_0 \rangle$ yields a finite contribution at large times the second term on the right of equation (15) grows as $\langle \tau \rangle_c^2$, namely it is subdominant compared to $\langle \tau^2 \rangle_c$ in all situations. If the exponent of the flight time distribution is $1 < g < 2$, then $\langle \tau^2 \rangle_c \sim t^{3-g} > t^{4-2g} \sim \langle \tau \rangle_c^2$; if $2 < g < 3$ we have that $\langle \tau \rangle_c^2$ is a finite number, while for $g > 3$ we recover a simple diffusive behavior for both the MSD and the drift. In particular, in the case $g > 3$ the presence of correlations of velocities amounts to a renormalization of the diffusion coefficient.

We conclude that the asymptotic behavior of the MSD is the same as in equation (11). From equation (13) one can see that no cross correlations between velocities at different times appear, so that the drift is not influenced by velocity correlations across collisions. Therefore, we can conclude that the Einstein relation is preserved for all values of the exponent g of the flight times' distribution, see for instance figure 1 for the case $g = 3/2$. This example shows how the Einstein relation is a stable result, valid under quite general conditions also in the presence of anomalous dynamics [7, 20, 11].

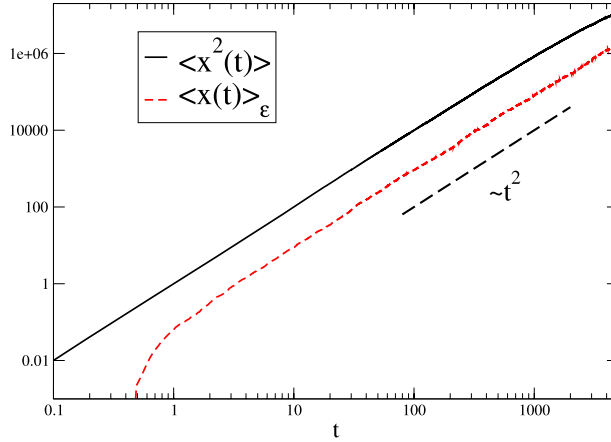


Figure 1. Log-log plot of the MSD in the case of superdiffusive dynamics with $g = 3/2$, $\langle x^2(t) \rangle$ (black line) and of the drift $\langle x(t) \rangle_{\mathcal{E}}$ (red dashed line) in the case of uniform external field $\mathcal{E} = 1.5$. Both quantities behave as $\sim t^2$ and the Einstein relation is verified.

3.2. Perturbation of a state with a current

In order to study the Einstein relation out of equilibrium, let us first consider a simple Gaussian process, namely the Brownian motion of a colloidal particle when we add a constant force pulling the particle. In this case it is sufficient to replace the MSD around the average position $\langle [\delta x(t)]^2 \rangle_{\mathcal{E}} = \langle x^2(t) \rangle_{\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}}^2$ for the simple MSD, in order to recover the Einstein relation with the drift $\langle x(t) \rangle_{\mathcal{E}+\delta\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}}$ (here $\langle \dots \rangle_{\mathcal{E}+\delta\mathcal{E}}$ denotes the average over a state where the further perturbation $\delta\mathcal{E}$ is applied). In what follows we consider that a non-trivial violation of the Einstein relation happens when also the MSD around the drift lacks proportionality with the drift itself. This is indeed the case of superdiffusive dynamics.

For simplicity we will refer to the case $\gamma = 0$ but the physical picture remains the same also when the case with memory is considered. In our model, by applying the constant field $\mathcal{E} > 0$, we have:

$$\begin{aligned} \langle x^2(t) \rangle_{\mathcal{E}} &= \left\langle \left[\sum_{i=1}^{N(t)} \left(v_i \tau_i + \frac{\mathcal{E}}{2} \tau_i^2 \right) \right]^2 \right\rangle_{t_c} \\ &\simeq \frac{\mathcal{E}^2}{4} t^2 \frac{\langle \tau^2 \rangle_c^2}{\langle \tau \rangle_c^2} + t \left(\frac{\mathcal{E}^2 \langle \tau^4 \rangle_c - \langle \tau^2 \rangle_c^2}{4 \langle \tau \rangle_c} + \frac{\langle v^2 \rangle \langle \tau^2 \rangle_c}{\langle \tau \rangle_c} \right) \end{aligned} \quad (16)$$

$$\langle [\delta x(t)]^2 \rangle_{\mathcal{E}} = \langle x^2(t) \rangle_{\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}}^2 \simeq t \left(\frac{\mathcal{E}^2 \langle \tau^4 \rangle_c - \langle \tau^2 \rangle_c^2}{4 \langle \tau \rangle_c} + \frac{\langle v^2 \rangle \langle \tau^2 \rangle_c}{\langle \tau \rangle_c} \right). \quad (17)$$

In the case $2 < g < 3$, namely when the distribution $P_{\tau}(\tau)$ has finite mean and infinite variance, the diffusion around the average position behaves asymptotically as

$$\langle [\delta x(t)]^2 \rangle_{\mathcal{E}} \simeq t \left(\frac{\mathcal{E}^2 t_c^{5-g} - t_c^{6-2g}}{4 \langle \tau \rangle_c} + \frac{\langle v^2 \rangle t_c^{3-g}}{\langle \tau \rangle_c} \right). \quad (18)$$

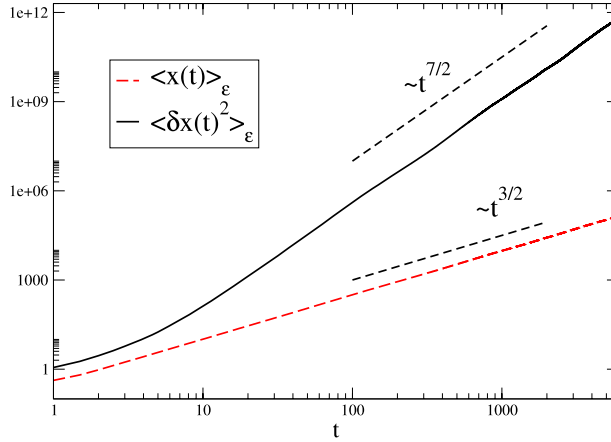


Figure 2. Log–log plot of the MSD (black line) in the presence of a constant external field $\mathcal{E} = 1.5$ and drift (red dashed line) with the same value of the field, in the case of superdiffusive dynamics with $g = 5/2$. We observe a breaking of the Einstein relation with leading asymptotic behaviors, $\langle [\delta x(t)]^2 \rangle_{\mathcal{E}} \sim t^{7/2}$, see equation (18), and $\langle x(t) \rangle_{\mathcal{E}} \sim t^{3/2}$, see equation (13).

Table 1. Columns from left to right: MSD for unperturbed dynamics; drift in the case of constant field $\mathcal{E} > 0$; second cumulant, i.e. $\langle [\delta x(t)]^2 \rangle_{\mathcal{E}} = \langle x^2(t) \rangle_{\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}}^2$, in the presence of a constant field \mathcal{E} .

	$\langle x^2(t) \rangle$	$\langle x(t) \rangle_{\mathcal{E}}$	$\langle [\delta x(t)]^2 \rangle_{\mathcal{E}}$
$g > 5$	t	t	t
$3 < g < 5$	t	t	t^{6-g}
$2 < g < 3$	t^{4-g}	t^{4-g}	t^{6-g}
$1 < g < 2$	t^2	t^2	t^4

Considering for instance the case $g = 5/2$, by applying the matching argument to equation (18), we find that the leading behaviors are

$$\langle x^2(t) \rangle_{\mathcal{E}} \sim t^3 \quad \langle [\delta x(t)]^2 \rangle_{\mathcal{E}} \sim t^{7/2}, \quad (19)$$

whereas, from equation (13), we have that

$$\langle x(t) \rangle_{\mathcal{E}+\delta\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}} \propto \langle x(t) \rangle_{\mathcal{E}} \sim t^{3/2}, \quad (20)$$

as shown in figure 2. The Einstein relation is therefore violated in the out-of-equilibrium regime for both the MSD and MSD around the average current for all the values of the flight time distribution exponent $g < 5$, as summarized in table 1.

As already noticed in [11] in the context of subdiffusive dynamics, such a violation of the Einstein relation is accompanied by an asymmetric spreading of the PDF $P(x, t)$ of observing the particle in x at time t . In the case of the standard random walk, when a perturbation \mathcal{E} is applied, the $P(x, t)$ remains Gaussian and the mean value coincides with the most probable one. On the contrary, in the present case, the average value of $x(t)$, due to the strongly asymmetric shape, grows much faster than the most probable value. The tail of $P(x, t)$ is reported in figure 3 (main frame). The stationary distribution

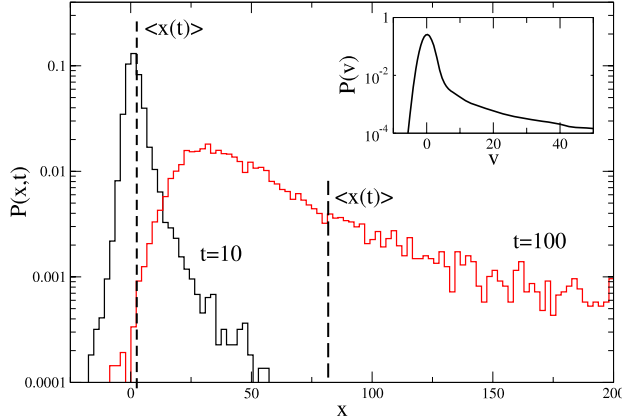


Figure 3. Main frame: semi-log plot of $P(x,t)$ at $t = 10$ (black histogram) and $t = 100$ (red histogram) for superdiffusive dynamics with $g = 5/2$ and constant external field $\mathcal{E} = 1.5$. Notice that for increasing time the typical (most frequent) value and the mean (indicated with a vertical dashed line) of the distribution become increasingly different for growing times. The increasing average value of the distribution is due to the arising of a power-law tail for $P(x,t)$. Inset: semi-log plot of the velocity PDF $P(v)$.

of the velocities is also asymmetric, and with a power-law tail, as shown in the inset of figure 3. The study of the velocity distribution in the diffusive case can be found in [19], while a careful study of the behavior of higher moments and of the scaling properties of $P(x,t)$ in similar models in the absence of external field can be found in [16] and [21], respectively.

The study of the Einstein relation on a state with non-zero current induced by a constant field \mathcal{E} allows us to show that there is some ‘anomaly’ in the dynamics also when the exponent of the power-law distribution of times is $g > 3$. More precisely, when $3 < g < 5$, at equilibrium, i.e. in the absence of current, a fluctuation-dissipation experiment would not show any anomaly in the dynamics, because $\langle [\delta x(t)]^2 \rangle \sim \langle x(t) \rangle_{\mathcal{E}}$. On the other hand, the same experiment performed out of equilibrium, i.e. comparing the MSD around the drift with the drift itself, shows an evident violation

$$\frac{\langle [\delta x(t)]^2 \rangle_{\mathcal{E}}}{\langle x(t) \rangle_{\mathcal{E}}} \sim t^{5-g}. \quad (21)$$

4. Conclusions

The present study concerns the comparison of the drift of a probe particle in the presence of an external field with its MSD, measured either in the presence or in the absence of an external perturbation. The outcome of our study is that, while for a Lévy walk process the *equilibrium* fluctuation-dissipation relation is always valid [8], the situation is very different in the *out-of-equilibrium* case, where we have a breaking of the Einstein relation. In particular, we find that the standard fluctuation-dissipation relation can be recovered by replacing the MSD with the diffusion around the average current $\langle x^2(t) \rangle_{\mathcal{E}} - \langle x(t) \rangle_{\mathcal{E}}^2$, as happens for standard random walks, *only* for values of the exponent $g > 5$. This

is a new and unexpected result, since already for $g > 3$ we observe a simple diffusive dynamics at equilibrium, even if the $P(x, t)$ is non-Gaussian. The non-Gaussian nature of the distribution of displacements emerges for $3 < g < 5$ only through a ‘response experiment’ in the presence of some currents.

As shown in figure 3, the violation of the fluctuation-dissipation relation for $g < 5$ comes together with a strongly asymmetric shape of the $P(x, t)$ for large times. Let us note that such a mechanism, namely a transport mechanism which does not correspond to a uniform shift of the $P(x, t)$, is peculiar not only to this superdiffusive model, but has already been observed in the context of subdiffusive dynamics [11].

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