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Time-averaged Einstein relation and fluctuating diffusivities for the Lévy walk

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The Lévy walk model is a stochastic framework of enhanced diffusion with many applications in physics and biology. Here we investigate the time-averaged mean squared displacement $\overline{\delta^2}$ often used to analyze single particle tracking experiments. The ballistic phase of the motion is nonergodic and we obtain analytical expressions for the fluctuations of $\overline{\delta^2}$. For enhanced subballistic diffusion we observe numerically apparent ergodicity breaking on long time scales. As observed by Akimoto [Phys. Rev. Lett. **108**, 164101 (2012)], deviations of temporal averages $\overline{\delta^2}$ from the ensemble average $\langle x^2 \rangle$ depend on the initial preparation of the system, and here we quantify this discrepancy from normal diffusive behavior. Time-averaged response to a bias is considered and the resultant generalized Einstein relations are discussed.

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In recent years there has been growing interest in the physics of weak ergodicity breaking [1–3]. In statistical mechanics the ergodic hypothesis states that ensemble averages and time averages are equal in the limit of long measurement times. Weak ergodicity breaking is found in systems whose temporal dynamics is governed by broad power law distributed waiting times with a diverging mean [1,2]. Weak ergodicity breaking allows for the exploration of the whole phase space (unlike strong ergodicity breaking) yet ergodicity is not attained since the diverging time scale of the dynamics always exceeds the measurement time [4]. The lack of a time scale for the dynamics leads to distributional limit theorems for time-averaged observables which are not trivial [3,5–8], while in the ergodic phase the distribution of the time averages are delta functions centered on the ensemble averages and in that sense are trivial. Weak ergodicity breaking is observed in many systems ranging from blinking quantum dots [9,10] (where sojourn times in on and off states are power law distributed) to models of glassy dynamics [1], and diffusion of molecules in the cell environment [11–13].

An observable that was extensively studied is the mean squared displacement (MSD) of a diffusing particle [13]. Let us first define the time averages. Experimentalists routinely track individual trajectories of particles and use the information for precise measurements of diffusion constants. The time-averaged MSD is defined through the path $x(t)$ in terms of

$$\overline{\delta^2} = \frac{1}{T-\Delta}\int_0^{T-\Delta}[x(t+\Delta)-x(t)]^2 dt, \qquad (1)$$

with the lag time Δ much smaller than the measurement time T. In the case of Brownian motion, due to the stationary increments, $\lim_{T\to\infty}\overline{\delta^2} = 2D\Delta$ is precisely the same as the MSD averaged over a large ensemble of particles $\langle x^2\rangle = 2D\Delta$, indicating ergodicity in the MSD sense. Here D is the diffusion constant. Now consider a weak force F acting on the particle, which will induce a net drift $\langle x\rangle_F \propto \Delta$. Throughout this Rapid Communication $\langle\cdots\rangle$ denotes ensemble averages while $\overline{\cdots}$ stands for time averaging. For Brownian particles the response to the field is related to the fluctuations via the celebrated Einstein relation $\langle x\rangle_F = F\langle x^2\rangle/(2\mathcal{T})$ [2,14], where $\mathcal{T}$ is the temperature and throughout this Rapid Communication $k_B = 1$. Since the Brownian process is ergodic, the Einstein relation will hold also for the time-averaged response, defined according to

$$\overline{\delta}_F = \frac{1}{T-\Delta}\int_0^{T-\Delta}[x(t+\Delta)-x(t)]dt, \qquad (2)$$

thus $\overline{\delta}_F = F\overline{\delta^2}/(2\mathcal{T})$.

Very recently Akimoto investigated temporal averages of anomalous diffusion and response to bias within the framework of deterministically generated Lévy walks [15]. In this well investigated and widely applicable process the diffusion is anomalous. Here two interesting issues arise. The first is the question of ergodicity of these processes, and the second the applicability of the Einstein relation to the time averages. Though time-averaged response to a bias was found to be intrinsically random, surprisingly the "temporal averaged MSDs are not random" [15]. This is diametrically opposed to results in previous examples of weak ergodicity breaking where temporal averages are intrinsically random [5–7,16]. For that reason we analytically investigate the previously ignored nontrivial fluctuations of the time-averaged MSD showing that the fluctuations are universal. We then formulate an Einstein relation for the time averages. We show that the Einstein relation for time averages differs considerably from the corresponding Einstein relation for the ensemble averages.

The Lévy walk model is a generalization of the classical Drude model describing a particle moving with constant velocity and changing its direction randomly. While in the Drude model exponential waiting times between turning events due to strong collisions result in a Markov process, the Lévy walk model postulates power law distributed waiting times between randomization events resulting in long flights [17–19]. The Lévy walk [18] describes enhanced transport phenomena in many systems, ranging from chaotic diffusion to animal foraging patterns [16,20–26]. For some very recent applications, see also Refs. [27–30]. The ubiquity of Lévy walks makes it particularly interesting to characterize and quantify their ergodic properties, leading to a better understanding of the physics at the core of such processes.

Lévy walk: Model and ensemble-averaged MSD. Superdiffusion based on power law waiting times is naturally described by the Lévy walk model [17]. We consider a particle alternating its velocity between $+v_0$ and $-v_0$ at random times. The times $0 < \tau < \infty$ between turning events are independent, identically distributed random variables with a common probability density function (PDF) $\psi(\tau)$. The position of the particle is $x = \int_0^t v(t')dt'$, so that the particle starts at $t = 0$ with velocity $+v_0$, travels a distance $v_0\tau_1$ with τ_1 drawn from $\psi(\tau)$, and after that is displaced $-v_0\tau_2$. The process is then renewed. The PDF of flight times τ is power law distributed, $\psi(\tau) \sim A\tau^{-(1+\alpha)}/|\Gamma(-\alpha)|$. When $0 < \alpha < 1$, the mean $\langle\tau\rangle$ diverges, while for $1 < \alpha < 2$ it is finite though $\langle\tau^2\rangle = \infty$. Our working example in simulations will be $\psi(\tau) = \alpha\tau^{-(1+\alpha)}$ for $\tau > 1$. Importantly, the displacements $\pm v_0\tau$ are broadly distributed, though they never become larger than $\pm v_0 t$.

The ensemble-averaged MSD is [17,31,32]

$$\langle x^2\rangle \sim \begin{cases} v_0^2(1-\alpha)t^2, & 0 < \alpha < 1, \\ 2K_\alpha t^{3-\alpha}, & 1 < \alpha < 2. \end{cases} \tag{3}$$

The case $0 < \alpha < 1$ is called the ballistic phase, while we refer to the parameter range $1 < \alpha < 2$ as enhanced diffusion which is subballistic. Here the anomalous diffusion coefficient is given by

$$K_\alpha = v_0^2 \frac{A(\alpha-1)}{\langle\tau\rangle\Gamma(4-\alpha)}. \tag{4}$$

Note that this transport coefficient was derived for a process which started at time $t = 0$. For both ballistic and enhanced regimes simulations reveal fluctuations of $\overline{\delta^2}$ at finite T, which surprisingly are more pronounced in the latter case (Fig. 1). Before we turn to these fluctuations, we will study the ensemble averages $\langle\overline{\delta^2}\rangle$, i.e., the mean of the distributions of the $\overline{\delta^2}$, denoted by the dashed lines in Fig. 1.

Averaging Eq. (1) we notice a relation between $\langle\overline{\delta^2}\rangle$ and the ensemble-averaged position correlation function,

$$\langle\overline{\delta^2}\rangle = \int_0^{T-\Delta} \frac{\langle x^2(t+\Delta)\rangle + \langle x^2(t)\rangle - 2\langle x(t)x(t+\Delta)\rangle}{T-\Delta} dt. \tag{5}$$

The correlation function $\langle x(t_1)x(t_2)\rangle$ is related to the velocity correlation function as

$$\langle x(t_1)x(t_2)\rangle = \int_0^{t_1} dt_1' \int_0^{t_2} dt_2' \langle v(t_1')v(t_2')\rangle. \tag{6}$$

Since $v(t_1) = v(t_2)$ [or $v(t_1) = -v(t_2)$], when the number of transitions n in the time interval (t_1,t_2) is even (or odd), we have

$$\langle v(t_1)v(t_2)\rangle = \sum_{n=0}^{\infty}(-1)^n v_0^2 p_n(t_1,t_2), \tag{7}$$

where $p_n(t_1,t_2)$ is the probability for n transitions of direction in the time interval (t_1,t_2). The behavior of the velocity correlation function Eq. (7) was studied by Godrèche and Luck [33] and it is described by two limits depending on the value of α.

The ballistic phase. For $\alpha < 1$ the dynamics is free of a time scale since $\langle\tau\rangle = \infty$ so that the particle will get stuck in a velocity state (either $+v_0$ or $-v_0$) for a duration of the order of the measurement time. Hence the dominating term in Eq. (7) is $n = 0$ and only the persistence probability $p_0(t_1,t_2)$ is important in the scaling limit of the problem [33],

$$\langle v(t_1)v(t_2)\rangle \simeq v_0^2 p_0(t_1,t_2) = v_0^2\frac{\sin\pi\alpha}{\pi} B\left(\frac{t_1}{t_2};\alpha,1-\alpha\right), \tag{8}$$

where $B(z;a,b)$ is the incomplete Beta function and $t_2 \geqslant t_1$. This velocity correlation function cannot be expressed as a function of the time difference $|t_2 - t_1|$, reflecting the nonstationarity of the process. Inserting Eq. (8) in Eq. (6) and integrating we get

$$\begin{aligned}\langle x(t_1)x(t_2)\rangle = v_0^2\frac{\sin\pi\alpha}{\pi}\Bigg[& t_1t_2B\left(\frac{t_1}{t_2};\alpha,1-\alpha\right) \\ & - \frac{1}{2}(t_2)^2B\left(\frac{t_1}{t_2};1+\alpha,1-\alpha\right) \\ & - \frac{1}{2}(t_1)^2B\left(\frac{t_1}{t_2},-1+\alpha,1-\alpha\right)\Bigg] - \alpha\frac{(v_0t_1)^2}{2},\end{aligned} \tag{9}$$

which reduces to the first line of Eq. (3) when $t_1 = t_2$. In the limit $\alpha \to 0$ the particle remains in state $+v_0$ or $-v_0$ for the whole duration of measurement time, hence we expect and indeed get $\langle x(t_2)x(t_1)\rangle = v_0^2 t_2 t_1$, which describes a deterministic motion. In contrast, for Brownian motion we have $\langle x(t_1)x(t_2)\rangle = 2D\min(t_1,t_2)$ reflecting independent increments of the process. Compared with the diffusive case, the Lévy walk exhibits strong correlations due to the long

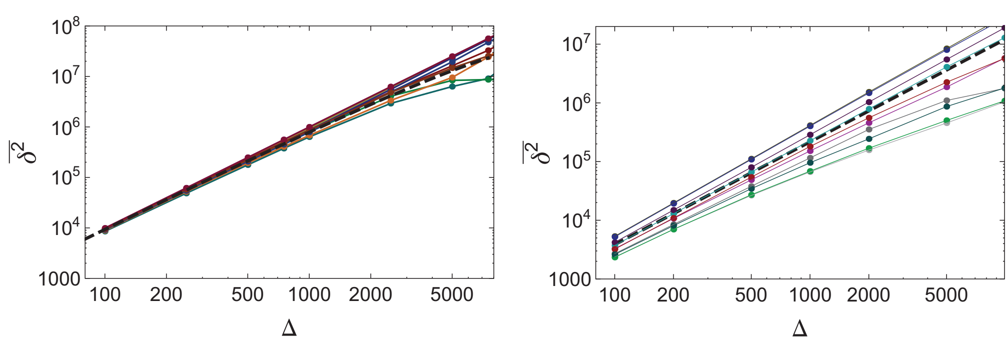

FIG. 1. (Color online) $\overline{\delta^2}$ of ten sample trajectories vs lag time Δ, $T = 10^4$, $\alpha = 1/2$ (left); $T = 10^5$, $\alpha = 5/4$, dashed lines indicate ensemble averages $\langle\overline{\delta^2}\rangle$, Eqs. (10) and (15). Larger times T and smaller lag times Δ result in smaller fluctuations. For comparable times T, the fluctuations in the enhanced case are larger than in the ballistic case.

sticking times in the positive or negative velocity states. For $\Delta \ll t_1$ we find $\langle x(t_1)x(t_1+\Delta)\rangle \sim \langle x^2(t_1)\rangle[1+(\Delta/t_1)]$, thus the correlations are strong in the sense that they are increasing with Δ.

Inserting the correlation function Eq. (9) in Eq. (5) and integrating, we find, in the limit $\Delta/T \ll 1$,

$$\langle\overline{\delta^2}\rangle \sim v_0^2\left[\Delta^2 - \frac{\sin\pi\alpha}{\pi\alpha}\frac{2\Delta^2\left(\frac{\Delta}{T}\right)^{1-\alpha}}{6-11\alpha+6\alpha^2-\alpha^3}\right]. \quad (10)$$

The leading term $\langle\overline{\delta^2}\rangle \sim (v_0\Delta)^2$ was found in Ref. [15] and corresponds to a deterministic, ballistic motion with velocity v_0. To see this, insert $[x(t+\Delta)-x(t)]^2 = (v_0\Delta)^2$ in Eq. (1), which yields $\overline{\delta^2} = (v_0\Delta)^2$.

More important are the fluctuations of the time-averaged MSD which quantify the ergodicity breaking. To explore this issue note that a particle not changing its direction at all $\overline{\delta^2} = (v_0\Delta)^2$ corresponds to a ballistic path. If the particle changes its velocity only once in the interval $(0,T)$, it is easy to show [34] that $\overline{\delta^2} = (v_0\Delta)^2 - (2/3)v_0^2\Delta^3/T$ for $T \gg \Delta$. Thus a single switching event reduces $\overline{\delta^2}$ by a term χ^2 proportional to $v_0^2\Delta^3/T$. If we have two transitions between $+v_0$ and $-v_0$ states, the correction term is twice as large [35]. Altogether we deduce that for a random amount n_T of switching events within the observation time T,

$$\overline{\delta^2} = (v_0\Delta)^2 - \chi^2 n_T. \quad (11)$$

Notice that this result is valid for a single trajectory, and both $\overline{\delta^2}$ and n_T are random. Once we find χ^2, this equation gives the sought after fluctuations of the time-averaged MSD, as will become clear soon. Further, we see that the natural random variable is the shifted MSD $(v_0\Delta)^2 - \overline{\delta^2}$, which is plotted in Fig. 2 versus the lag time Δ. Now the fluctuations are clearly visible, unlike the presentation in Fig. 1.

We now determine χ^2. From renewal theory [2,36] the average number of switchings (renewals) is $\langle n_T\rangle \sim T^\alpha/A\Gamma(1+\alpha)$. Comparison of the average of Eq. (11) with Eq. (10) thus yields

$$\chi^2 = \frac{2\sin\pi\alpha A\Gamma(1+\alpha)}{\pi\alpha(6-11\alpha+6\alpha^2-\alpha^3)}\frac{v_0^2\Delta^{3-\alpha}}{T}, \quad (12)$$

a result valid for $\Delta \ll T$ [37].

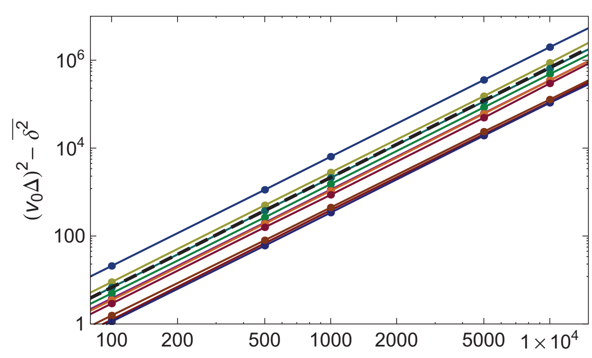

FIG. 2. (Color online) Deviations from ballistic motion of the time-averaged MSDs $(v_0\Delta)^2 - \overline{\delta^2}$ vs the lag time Δ for ten trajectories; $\alpha = 1/2$, $v_0 = 1$, $T = 10^8$. The dashed line denotes the theoretical ensemble average Eq. (10).

To quantify the fluctuations we introduce the dimensionless random parameter

$$\xi = \frac{\overline{\delta^2} - (v_0\Delta)^2}{\langle\overline{\delta^2}\rangle - (v_0\Delta)^2} = \frac{n_T}{\langle n_T\rangle}, \quad (13)$$

which has mean equal one. The fluctuations of the number of switchings n_T are well known from renewal theory [2,3,36] as they are determined by the waiting time distribution $\psi(\tau)$ only. The case $\alpha < 1$ implies that Lévy's central limit theorem holds, which gives the PDF of ξ,

$$g(\xi) = \frac{\Gamma^{1/\alpha}(1+\alpha)}{\alpha\xi^{1+1/\alpha}} l_{\alpha,1}\left[\frac{\Gamma^{1/\alpha}(1+\alpha)}{\xi^{1/\alpha}}\right]. \quad (14)$$

Here $l_{\alpha,1}(t)$ denotes the one sided Lévy stable PDF whose Laplace transform is given by $\exp(-u^\alpha)$ [2,36,38]. Figure 3 shows excellent agreement between the PDF Eq. (14) and the respective simulation results. As mentioned above, Ref. [15] showed that transport, i.e., time-averaged response to external bias, is random. Equation (14) shows that also the time-averaged diffusivity of the process is random, though one must consider the shifted MSD defined in Eq. (13) to observe the fluctuations typical for weak ergodicity breaking.

The enhanced diffusion phase. For $1 < \alpha < 2$ the dynamics has a finite time scale $\langle\tau\rangle$ and therefore one may naively expect the normal behavior $\langle\overline{\delta^2}\rangle = \langle x^2\rangle$. Similarly to the ballistic phase, we find the correlation function [39]

$$\langle x(t_1)x(t_1+\Delta)\rangle = \frac{K_\alpha}{(\alpha-1)}t_1^{3-\alpha}h(\theta),$$

with

$$h(\theta) = \alpha + (1+\theta)^{3-\alpha} + (\alpha-3)(1+\theta)^{2-\alpha} - \theta^{3-\alpha},$$

where $\theta = \Delta/t_1$. Inserting this expression in Eq. (5) we find

$$\langle\overline{\delta^2}\rangle = \frac{\langle x^2\rangle}{\alpha-1}. \quad (15)$$

Thus, except for the normal diffusion limit of $\alpha \to 2$, the ensemble-averaged MSD differs from the time-averaged MSD by a factor. Numerical evidence for a difference between $\langle x^2\rangle$ and $\langle\overline{\delta^2}\rangle$ was presented earlier [15] in the context of diffusion generated by deterministic maps. Equation (15) quantifies this deviation with $\alpha = 1/(z-1)$ and $3/2 < z < 2$ being the nonlinearity parameter of the deterministic map in Ref. [15].

To explain this effect note that $\langle x^2\rangle$ is calculated for a process which starts at time $t = 0$. Physically this corresponds to a particle immersed in a system at time $t = 0$ when the process begins. Alternatively we may measure or calculate the *stationary* MSD $\langle x^2\rangle_{\rm st}$ of a process which started long before the measurement begins at $t = 0$. In this case, since $\langle\tau\rangle$ is finite, the system is in the stationary state throughout the measurement time, i.e., from $t = 0$ on. Hence one may use the Green-Kubo formalism to obtain the stationary MSD $\langle x^2\rangle_{\rm st} = 2K_\alpha t^{3-\alpha}/(\alpha-1)$, as was done earlier in Refs. [40,41]. Thus we find that $\langle\overline{\delta^2}\rangle = \langle x^2\rangle_{\rm st} \neq \langle x^2\rangle$. The assessment of the ergodic properties of the process in the sense of equal time- and ensemble-averaged MSD is therefore a subtle issue which depends on the initial preparation of the system. Such a behavior is not found for normal diffusion processes.

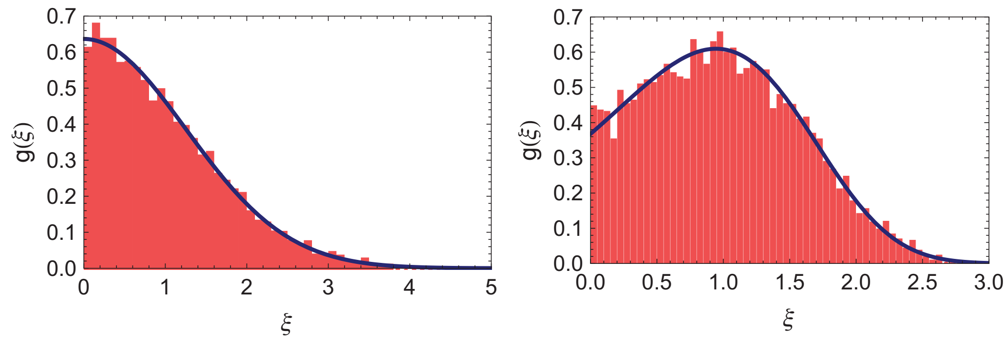

FIG. 3. (Color online) PDF of $\xi = [(v_0\Delta)^2 - \overline{\delta^2}]/[(v_0\Delta)^2 - \langle\overline{\delta^2}\rangle]$ for $\alpha = 0.5$ (left) and $\alpha = 0.7$ (right). Simulations (histograms) comply with theory (solid lines); $T = 10^6$, $\Delta = 1$, sample size 10^4. When $\alpha = 0.5$, the peak of the PDF is on $\xi = 0$; for $\alpha \to 1$ the peak tends to $\xi = 1$ and fluctuations of ξ vanish.

In biophysical experiments the time-averaged MSDs of trajectories measured up to a certain observation time are often distributed [42,43]. In single particle tracking experiments in the living cell the origins of these fluctuations and of the anomalous transport are still an object of controversy [13,44,45]. Recent careful statistical analyses revealed multiscaling of the moments of active motion of small polymeric particles in living cells [26]. Multiscaling is a non-Gaussian feature which is consistent with Lévy walks rather than with the Gaussian and ergodic Langevin picture. Reference [26] suggests Lévy walks as a first simplified approach to the active transport in this specific setting. Time averages are made over finite times which are limited by biological function, e.g., the measurement time cannot be larger than the lifetime of the cell. Hence the usual infinite long time limit and ideas on stationarity are not relevant in many single biomolecule experiments. In our simulations we find large fluctuations intrinsic to the Lévy walk model for $\alpha = 5/4$ (see the left panel of Fig. 1) among the $\overline{\delta^2}$ even for $\Delta/T = 10^{-3}$. To check whether $\overline{\delta^2}$ remains random, we investigated the fluctuations which vanish in the long time limit, though slowly (in contrast to Lévy flights, see Ref. [46]). Thus we find that

$$\lim_{T\to\infty} \frac{\overline{\delta^2}}{\langle x^2\rangle} = \frac{1}{|1-\alpha|}, \tag{16}$$

both for the ballistic and enhanced diffusion phase.

Response to bias and generalized Einstein relation. Now we assume a small constant force F acting on the particle, a case that leads to an anomalous drift. Thereby the force accelerates the particle according to Newton's law of motion, similarly to the Drude model, while the waiting times for the collisions are still drawn from $\psi(\tau)$. We consider the time average Eq. (2) and find, using $\langle x\rangle_F$,

$$\langle\overline{\delta}\rangle_F = \begin{cases} (1-\alpha)FT\Delta/(2M), & 0<\alpha<1, \\ K_\alpha F T^{2-\alpha}\Delta/\left(Mv_0^2\right), & 1<\alpha<2. \end{cases} \tag{17}$$

These results differ from their corresponding ensemble average $\langle x\rangle_F$ in that they depend on both the lag time Δ and the measurement time T. In contrast, $\langle x\rangle_F$ clearly depends only on the measurement time, namely, $\langle x\rangle_F = F(1-\alpha)/(2M)t^2$ for $0<\alpha<1$ and $\langle x\rangle_F = FK_\alpha/(Mv_0^2)t^{3-\alpha}$ for $1<\alpha<2$ [14]. The equivalence of time and ensemble averaging is thus broken. This is a consequence of the nonlinear dependence of $\langle x\rangle_F$ on time and thus in fact a very general behavior valid for any system whose response to the driving force is anomalous. The limit of normal diffusion $\alpha \to 2$ renders $\langle\overline{\delta}\rangle_F$ a function of the lag time only so that ergodicity is retained.

Using Eqs. (10), (15), and (17), we find the generalized Einstein relation for the time averages,

$$\frac{\langle\overline{\delta}\rangle_F}{\langle\overline{\delta^2}\rangle} = \frac{|1-\alpha|F}{2\mathcal{T}_{\rm eff}}\left(\frac{T}{\Delta}\right)^{\gamma}. \tag{18}$$

Here $\gamma = 1$ in the ballistic phase, while in the enhanced phase $\gamma = 2-\alpha$. The effective temperature is defined with the averaged kinetic energy of the particle $\mathcal{T}_{\rm eff}/2 = Mv_0^2/2$ $(k_B = 1)$. Our relation Eq. (18) is very different from the standard Einstein relation for the ensemble averages $\langle x\rangle_F/\langle x^2\rangle = F/(2\mathcal{T}_{\rm eff})$ [2], the normal diffusion limit $\alpha \to 2$ being the exception.

Conclusion. Since the days of Einstein huge attention has been given to ensemble-averaged response functions (e.g., mobility) and its relation to fluctuations via fluctuation dissipation relations. In this Rapid Communication we have shown that for a widely applicable class of anomalous processes the time averages do not obey simple Einstein relations, contrary to the ensemble averages. Due to the lack of a time scale in the dynamics, we find a different type of Einstein relation, Eq. (18), which depends on the measurement time and thus exhibits aging. This type of Einstein relation entails a mobility effectively increasing with the measurement time, reflecting the large excursions in the Lévy walk. Further, we have unraveled the nature of the fluctuations of the time-averaged MSDs of the Lévy walk which exhibit Mittag-Leffler universality in the ballistic phase, Eqs. (13) and (14). In the enhanced phase the fluctuations were comparably large though slowly decaying, and revealed a delicate sensitivity to the initial preparation of the system, characteristics that cannot be found for normal processes.

Note added: After this work was completed, related work on the enhanced phase was published [47].

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[35] Provided the two transitions do not fall within a time interval Δ.
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[37] Here we assume that $1\ll\Delta$ where the time scale 1 refers to the cutoff of $\psi(\tau)=\alpha\tau^{-(1+\alpha)}$ for $\tau>1$. In the opposite limit $\Delta\ll 1\ll T$ we have $\chi^2=2v_0^2\Delta^3/(3T)$.
[38] $g(\xi)$ is the density of the so-called Mittag-Leffler distribution.
[39] We obtain the velocity correlation function Eq. (7) in the scaling limit and used Eq. (6) to find $\langle x(t_1)x(t_1+\Delta)\rangle$ (details will be published elsewhere). To get Eq. (15) we used the $\langle x(t_1)$ $x(t_1+\Delta)\rangle$, Eqs. (3) and (5).
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