

Fluctuations of Time Averages for Langevin Dynamics in a Binding Force Field

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(Received 3 June 2011; revised manuscript received 22 September 2011; published 9 December 2011)

We derive a simple formula for the fluctuations of the time average $\bar{x}(t)$ around the thermal mean $\langle x \rangle_{\text{eq}}$ for overdamped Brownian motion in a binding potential $U(x)$. Using a backward Fokker-Planck equation, introduced by Szabo, Schulten, and Schulten in the context of reaction kinetics, we show that for ergodic processes these finite measurement time fluctuations are determined by the Boltzmann measure. For the widely applicable logarithmic potential, ergodicity is broken. We quantify the large nonergodic fluctuations and show how they are related to a superaging correlation function.

DOI: 10.1103/PhysRevLett.107.240603

PACS numbers: 05.10.Gg, 05.20.Gg, 05.40.-a

Current technology permits tracking of trajectories of individual molecules with exquisite precision. The motion of a Brownian particle in a binding potential field $U(x)$ is used to model many such physical, biological, and chemical processes. From statistical mechanics, we know that if the process is ergodic, and if the measurement time $t \rightarrow \infty$, then the time average $\bar{x}(t) = \int_0^t x(t') dt' / t$ is equal to the corresponding ensemble average $\langle x \rangle_{\text{eq}}$. In experiment the measurement time might be long, but it is always finite. Hence it is natural to ask what the fluctuations of \bar{x} are. Such an analysis sheds light on deviations from the thermal equilibrium average due to finite time measurement, a general theme which has attracted much interest in the context of fluctuation theorems [1]. The Boltzmann measure, due to ergodicity, yields equilibrium properties of thermal systems. Surprisingly, we find that for Langevin dynamics, the Boltzmann measure also determines the deviations from ergodicity.

As we will show, for binding fields $U(x)$ where the Fokker-Planck (FP) operator exhibits a discrete eigenspectrum, the fluctuations of the time average \bar{x} become small as time increases, as expected from ordinary ergodic statistical mechanics. For this type of field, ergodicity is related to the work of Szabo, Schulten, and Schulten [2] on the seemingly unrelated problem of reaction kinetics (see details below). A more interesting case is that of a logarithmic binding field [3] $U(x) \sim U_0 \ln(|x|)$ when $|x| \rightarrow \infty$, since for such a potential the fluctuations of \bar{x} are not small even in the long time limit. Here the Boltzmann measure exhibits power-law tails, $P^{\text{eq}}(x) \propto |x|^{-U_0/(k_B T)}$. Starting at the origin, the particle during its evolution tends to sample larger and larger values of $|x|$ as illustrated in Fig. 1. Large fluctuations in the amplitude of $x(t)$ cause the time average of this special process to remain random even in the long time limit. In what follows, we calculate the magnitude of these fluctuations and show how they are related to a superaging correlation function. Importantly,

such logarithmic potentials model many physical systems, ranging from optical lattices [4], charges in vicinity of a long charged polymer [5], DNA dynamics [6], membrane induced forces [7], a nanoparticle in a trap [8], to long ranged interacting models [9]. At the end of this Letter we discuss the connection between our theory and a recent experiment [10].

Model and observable.—Brownian dynamics in a force field $f(x) = -dU(x)/dx$ obeys the equation [11]

$$\frac{dx}{dt} = -\frac{f(x)}{\gamma} + \eta(t). \quad (1)$$

Here γ is the friction constant, $\eta(t)$ is Gaussian white noise obeying the fluctuation dissipation relation $\langle \eta(t)\eta(t') \rangle = 2D\delta(t-t')$, and $D = k_B T / \gamma$ according to the Einstein relation. From the trajectory $x(t)$ we construct the time average $\bar{x}(t) = \int_0^t x(t') dt' / t$. For a binding potential, in the

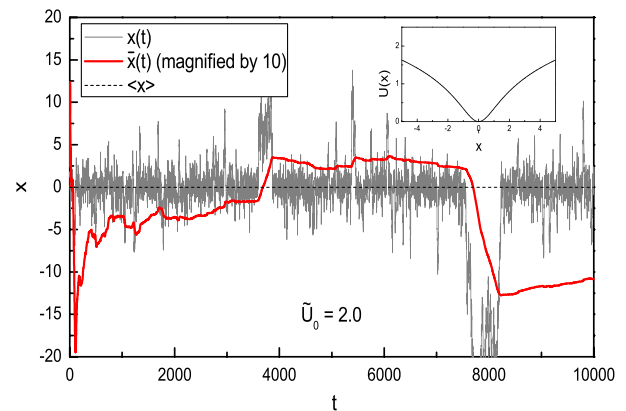


FIG. 1 (color online). The trajectory of a Brownian particle in a logarithmic potential exhibits large amplitude fluctuations. As a consequence, the time average of the process \bar{x} (red curve) does not converge to a fixed value even though the Boltzmann equilibrium ensemble average $\langle x \rangle_{\text{eq}}$ is zero. Here $U(x) = \ln(1 + x^2)/2$, $k_B T = 1/2$ and the diffusion constant $D = 1$.

long time limit, x obeys the equilibrium Boltzmann distribution:

$$P^{\text{eq}}(x) = \frac{\exp[-\frac{U(x)}{k_B T}]}{Z}; \quad Z = \int_{-\infty}^{\infty} e^{-U(x)/(k_B T)} dx \quad (2)$$

where Z is the normalizing partition function which is assumed to be finite. We consider symmetric potentials $U(x) = U(-x)$ and then the ensemble average in equilibrium $\langle x \rangle_{\text{eq}} = \int_{-\infty}^{\infty} x P^{\text{eq}}(x) dx = 0$. If the process is ergodic then in the long time limit $\bar{x} \rightarrow \langle x \rangle_{\text{eq}} = 0$. If $\lim_{t \rightarrow \infty} \langle \bar{x}^2(t) \rangle \neq 0$ the process is nonergodic, where $\langle \cdot \cdot \cdot \rangle$ stands for an ensemble mean. In the second part of our

work we show that not all binding potentials satisfy the ergodic hypothesis.

Szabo-Schulten-Schulten equation yields the fluctuations of the time average.—The variance of the time average is given by

$$\langle \bar{x}^2(t) \rangle = \frac{1}{t^2} \int_0^t dt_2 \int_0^{t_2} dt_1 \langle x(t_2)x(t_1) \rangle \quad (3)$$

where $\langle x(t_2)x(t_1) \rangle$ is the correlation function. For the Markovian process under investigation, and for a particle starting at the origin at time $t = 0$ we have [11]

$$\langle \bar{x}^2(t) \rangle = \frac{2}{t^2} \int_0^t dt_2 \int_0^{t_2} dt_1 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} x_2 x_1 P(x_2, t_2 | x_1, t_1) P(x_1, t_1 | 0, 0) dx_1 dx_2 \quad (4)$$

where $P(x_2, t_2 | x_1, t_1)$ is the conditional probability density to find the particle on x_2 at time t_2 once it is located at x_1 at time t_1 . In the limit of long times, the major contribution to the integration over t_1 comes from long times; hence one replaces $P(x_1, t_1 | 0, 0)$ with $P^{\text{eq}}(x_1)$. To proceed, it is useful to define

$$\xi(x_1) = \int_0^{\infty} E(x_1, \tau) d\tau \quad (5)$$

where $E(x_1, \tau)$ is the averaged position of a particle at a time τ after it starts at x_1 . Two cases are of interest: the first is when $\xi(x_1)$ is finite, the other when it diverges. We shall start with the former case which is clearly relevant to potential fields where the FP eigenspectrum [11] has a finite energy gap to the ground state, since then the relaxation of $E(x_1, \tau)$ is exponential. From Eq. (4) it follows that in the long time limit

$$\langle \bar{x}^2(t) \rangle \sim \frac{2}{t} \int_{-\infty}^{\infty} x_1 \xi(x_1) P^{\text{eq}}(x_1) dx_1. \quad (6)$$

As is well known the backward FP equation [11]

$$L_{\text{FP}}^{\dagger} P(x_2, \tau | x_1, 0) = \frac{\partial}{\partial \tau} P(x_2, \tau | x_1, 0), \quad (7)$$

$$L_{\text{FP}}^{\dagger} = D \left[\frac{\partial^2}{\partial (x_1)^2} + \frac{f(x_1)}{k_B T} \frac{\partial}{\partial x_1} \right]$$

governs the dynamics where L_{FP}^{\dagger} is the adjoint FP operator and $P(x_2, 0 | x_1, 0) = \delta(x_2 - x_1)$. By definition $E(x_1, \tau) = \int_{-\infty}^{\infty} x_2 P(x_2, \tau | x_1, 0) dx_2$ which implies

$$L_{\text{FP}}^{\dagger} E(x_1, \tau) = \frac{\partial}{\partial \tau} E(x_1, \tau) \quad (8)$$

with $E(x_1, 0) = x_1$. Using Eq. (5), we find

$$L_{\text{FP}}^{\dagger} \xi(x_1) = -x_1 \quad (9)$$

with $\xi(0) = 0$. Equation (9) was obtained previously in [2] in the context of reaction kinetics. Equations (4)–(9) are so

general that they could be extended to arbitrary Markovian processes. The latter equations thus serve as a starting point for the investigation of fluctuations of time averages for a wide class of systems.

Fluctuations of time averages determined from Boltzmann statistics.—Equation (9) is easy to solve, and upon using Eq. (6) we find the general formula

$$\langle \bar{x}^2 \rangle \sim \frac{2}{Dt} \int_{-\infty}^{\infty} \frac{e^{U(x)/(k_B T)}}{Z} dx \left[\int_x^{\infty} x' e^{-U(x')/(k_B T)} dx' \right]^2. \quad (10)$$

As is well known, Boltzmann statistics can be used to determine the time average of ergodic processes: $\bar{x} \rightarrow \langle x \rangle$ in the long time limit. Equation (10) shows that the finite time fluctuations of \bar{x} are also determined by the Boltzmann distribution. Surprisingly, Eq. (10) shows that the difficult task of finding the entire eigenspectrum of the FP operator is not required. Equation (10) is easily generalized to dimensions greater than one, and to nonthermal processes whose equilibrium density is non-Boltzmannian. As expected from ergodicity, the magnitude of the fluctuations decays to zero with time, provided that the integrals in Eq. (10) converge. For example, for the harmonic potential $U(x) = m\omega^2 x^2/2$ we get $\langle \bar{x}^2 \rangle \sim 2(k_B T)^2/[D(m\omega^2)^2 t]$. An interesting case where the integrals diverge is the logarithmic potential $U(x) \sim U_0 \ln(|x|)$ for $|x| \rightarrow \infty$ and $U_0/(k_B T) < 5$. This leads to a nonergodic behavior which we now investigate.

Logarithmic potential.—We will first find the two-point correlation function $\langle x(t_2)x(t_1) \rangle$ for a general logarithmic potential which satisfies $U(x) \sim U_0 \ln(|x/a|)$, e.g. $U(x) = 0.5U_0 \ln[1 + (x/a)^2]$. We will then use (3) to obtain the fluctuations of the time average showing that for high enough temperature the fluctuations increase with time. For this potential, for $1 < U_0/(k_B T) < 5$, due to the slow convergence of the tail of the distribution to P^{eq} , and the slow power-law decay of $E(x_1, \tau)$ (which we shall shortly

demonstrate), rendering $\xi(x_1)$ infinite for $U_0/(k_B T) < 2$, one must consider the full time dependent problem instead of the time independent Eq. (9) and $P^{\text{eq}}(x)$. Generally the correlation function is given by

$$\langle x(t_2)x(t_1) \rangle = \int_{-\infty}^{\infty} x_1 E(x_1, t_2 - t_1) P(x_1, t_1 | 0, 0) dx_1. \quad (11)$$

To solve this problem we used two approaches; the first is based on an eigenfunction expansion of the solution of the FP equation [12]. Such a calculation is lengthy and hence we adopt here a scaling approach. As seen from Eq. (11) the key quantity to calculate is the ensemble mean $E(x_1, \tau)$ using Eq. (8). Because of the homogenous character of the large x Fokker-Planck operator, it is natural to adopt a scaling ansatz:

$$E(x_1, \tau) \sim \tau^\alpha g\left(\frac{x_1}{\tau^\beta}\right) \quad (12)$$

where α and β are scaling exponents. Since for short time $E(x_1, \tau) \simeq x_1$ we have $g(y) \simeq y$ for large y and $\alpha = \beta$. Inserting Eq. (12) in Eq. (8) we find to leading order

$$\tau^{-\beta} D \left(g'' - \tilde{U}_0 \frac{g'}{y} \right) = \tau^{\beta-1} \beta (g - yg'), \quad (13)$$

where $\tilde{U}_0 = U_0/(k_B T)$ is a key dimensionless parameter. To achieve a t -independent equation, we must have $\beta = 1/2$, typical of Brownian motion. Then,

$$g(y) = c_1 y^{1+\tilde{U}_0} e^{-(y^2)/(4D)} M\left(\frac{3}{2}, \frac{3+\tilde{U}_0}{2}, \frac{y^2}{4D}\right) \quad (14)$$

where $M(a, b, x)$ [also denoted ${}_1F_1(a; b; x)$] is the Kummer M function [13] and we rejected a second solution in terms of the Kummer U function since it does not satisfy the boundary condition $E(x_1, \tau) \rightarrow 0$ when $\tau \rightarrow \infty$ (i.e., relaxation to equilibrium). The constant c_1 is found by matching the solution in the $y \rightarrow \infty$ limit which corresponds to short times. Using $M(a, b, x) \sim \exp(x)\Gamma(b)x^{a-b}/\Gamma(a)$ and $g(y) \sim y$, we find $c_1 = \{\Gamma(3/2)/\Gamma[(3+\tilde{U}_0)/2]\}(4D)^{-\tilde{U}_0/2}$. In particular, for long times, $E(x_1, \tau) \sim \tau^{-\tilde{U}_0/2}$, so as we claimed, $\xi(x_1)$ diverges for $\tilde{U}_0 < 2$.

Steady state cannot be used to obtain the correlation function.—To complete the calculation, we must have $P(x_1, t_1 | 0, 0)$ which was recently obtained [14]. The equilibrium probability density function (PDF), since it decays as a power law $P^{\text{eq}}(x) \propto |x|^{-\tilde{U}_0}$ would give, for $1 < \tilde{U}_0 < 3$, $\langle x(t_2)x(t_1) \rangle = \infty$ for $t_1 = t_2$. This is an unphysical behavior: at finite time one cannot have an infinite value for the correlation function, since the particle cannot travel faster than diffusion permits. Specifically in the limit of long t_1 we have [14]

$$P(x_1, t_1 | 0, 0) \sim P^{\text{eq}}(x_1) \frac{\Gamma\left(\frac{1+\tilde{U}_0}{2}, \frac{(x_1)^2}{4Dt_1}\right)}{\Gamma\left(\frac{1+\tilde{U}_0}{2}\right)}. \quad (15)$$

Since $\Gamma(a, 0) = \Gamma(a)$ as $t \rightarrow \infty$, thermal equilibrium is reached. Nevertheless, for the calculation of correlation functions one must take into account the finite time correction which is represented by the ratio of Γ functions.

Aging correlation function.—Inserting Eqs. (14) and (15) in Eq. (11), we find the nonstationary correlation function for the temperature range $1 < \tilde{U}_0 < 3$:

$$\langle x(t_2)x(t_1) \rangle \sim \langle x^2(t_1) \rangle f_{\tilde{U}_0}\left(\frac{t_2 - t_1}{t_1}\right) \quad (16)$$

where

$$f_{\tilde{U}_0}(s) = \frac{\sqrt{\pi}(3 - \tilde{U}_0)}{2\Gamma\left(\frac{3+\tilde{U}_0}{2}\right)} s^{(3-\tilde{U}_0)/2} \times \int_0^\infty dy y^2 e^{-y^2} M\left(\frac{3}{2}, \frac{3+\tilde{U}_0}{2}, y^2\right) \Gamma\left(\frac{\tilde{U}_0+1}{2}, y^2 s\right). \quad (17)$$

The behavior in Eq. (16) is very different than the stationary case where the correlation function is a function of the time difference $t_2 - t_1$. In this temperature regime the equilibrium mean-square displacement diverges, $\langle x^2 \rangle_{\text{eq}} = \infty$, while the time dependent solution Eq. (15) gives [14] $\langle x^2(t_1) \rangle = a^2 c_2 (4Dt_1/a^2)^{(3-\tilde{U}_0)/2}$, $c_2 = 2(a/Z)[\Gamma(1/2 + \tilde{U}_0/2)(3 - \tilde{U}_0)]^{-1}$. We find $f_{\tilde{U}_0}(0) = 1$ which implies that $C(t_1, t_1) = \langle x^2(t_1) \rangle$, as it should. In the opposite limit $t_2 \gg t_1$, we obtain

$$\langle x(t_2)x(t_1) \rangle \sim c_3 \langle x^2(t_1) \rangle \left(\frac{t_2}{t_1}\right)^{-(\tilde{U}_0)/2} \quad (18)$$

with $c_3 = (3/2 - \tilde{U}_0/2)\sqrt{\pi}\Gamma(2 + \tilde{U}_0/2)/3\Gamma(3/2 + \tilde{U}_0/2)$. In Fig. 2 we compare our analytical Eq. (17) with Langevin simulations showing excellent agreement for various measurement times.

As mentioned we assume that the partition function Z is finite and hence the steady state $P^{\text{eq}}(x)$ is normalizable. This excludes the well-known Bessel process [15] which

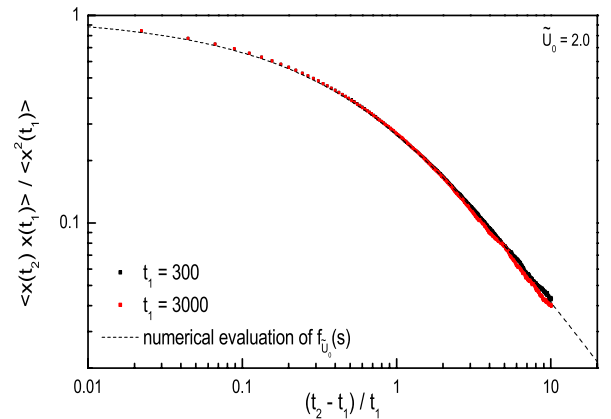


FIG. 2 (color online). The aging correlation function Eq. (16) perfectly matches numerical simulation of the Langevin Eq. (1).

can be mapped onto $U(x) = U_0 \ln|x|$ with its singularity at the origin. It is important to emphasize that $\langle x(t_2)x(t_1) \rangle \sim 1/Z$ depends on the shape of the potential *in the whole space* through Z . Hence for the calculation of the correlation function the regularity of the potential at the origin is vital. Interestingly this is not the case for all observables; e.g., $E(x_1, \tau)$ Eqs. (12) and (14), is Z independent and hence related to the Bessel process [15].

Ergodicity of the dynamics.—Ergodicity of the dynamics is classified in four domains which are controlled by temperature. (a) The most interesting case is the regime $1 < \tilde{U}_0 < 3$. As we showed, a normalized steady state exists and from symmetry $\langle x \rangle_{\text{eq}} = 0$. If we naively assume ergodicity $\bar{x} \rightarrow \langle x \rangle_{\text{eq}} = 0$ and $\lim_{t \rightarrow \infty} \langle \bar{x}^2(t) \rangle = 0$. Rather, from Eqs. (3) and (16), we find [16]

$$\langle \bar{x}^2(t) \rangle \sim \frac{2\langle x^2(t) \rangle}{t^2} \int_0^t dt_1 \int_{t_1}^t dt_2 \left(\frac{t_1}{t}\right)^{(3-\tilde{U}_0)/2} f_{\tilde{U}_0}\left(\frac{t_2-t_1}{t_1}\right). \quad (19)$$

Changing variables to $s = t_2/t_1 - 1$ and $w = t/t_1 - 1$, we find

$$\langle \bar{x}^2(t) \rangle \sim c_4 \langle x^2(t) \rangle \propto t^{(3-\tilde{U}_0)/2} \quad (20)$$

where $c_4 = 4 \int_0^\infty dw (1+w)^{(\tilde{U}_0-7)/2} (7-\tilde{U}_0)^{-1} f_{\tilde{U}_0}(w)$. We see that the fluctuations grow with time; hence, ergodicity is broken. We find that $c_4 \approx 0.2397$ for $\tilde{U}_0 = 1$ and that it decreases monotonically to $c_4 = 0$ at $\tilde{U}_0 = 3$. (b) For lower temperature, $3 < \tilde{U}_0 < 5$, the integrals in Eq. (10) still diverge, and $\langle \bar{x}^2(t) \rangle$ decays as $t^{(3-\tilde{U}_0)/2}$, indicating an anomalously slow approach to ergodicity. (c) For $\tilde{U}_0 > 5$ the temperature is low enough that Eq. (10) is now valid. For $U(x) = 0.5U_0 \ln[1 + (x/a)^2]$ we find

$$\langle \bar{x}^2(t) \rangle \sim \frac{2(\tilde{U}_0 - 4)}{(\tilde{U}_0 - 2)(\tilde{U}_0 - 3)} \frac{a^4}{(\tilde{U}_0 - 5)Dt} \quad (21)$$

which diverges when $\tilde{U}_0 \rightarrow 5$. (d) Finally, for very high temperatures $\tilde{U}_0 < 1$, the equilibrium state Eq. (2) is not defined as the partition function Z diverges. Here $\langle \bar{x}^2(t) \rangle \propto t$, exactly the diffusive behavior of a free particle, $U_0 = 0$ [12]. These four different behaviors are confirmed via numerical simulations presented in Fig. 3, which illustrates convergence on reasonable computer time scales. A summary of the scaling regimes is presented in Table I.

Relation with experiment.—After the submission of this Letter, an experiment on anomalous diffusion of ultracold atoms which employs the well known Sisyphus cooling scheme was reported [10]. In the semiclassical approximation, the atomic velocity distribution follows Fokker-Planck dynamics in an asymptotically logarithmic potential [4,14,17]. Our work provides the theoretical mean-square displacement in this experiment by identifying our position x with the velocity v of the atoms. The measured atomic position is $x(t) = \int_0^t v(t)dt$ and hence

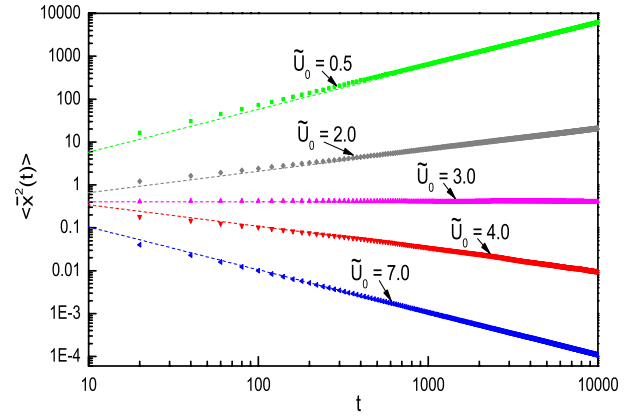


FIG. 3 (color online). In the nonergodic phase $k_B T > U_0/3$ ($\tilde{U}_0 < 3$), $\langle \bar{x}^2(t) \rangle$ increases with time. For the critical point $k_B T = U_0/3$ ($\tilde{U}_0 = 3$), the fluctuations are constant. The dashed curves are theoretical predictions Eqs. (20) and (21), which agree very well with the numerical simulations.

$x(t)/t$ corresponds to the time averaged velocity. The PDF of the atoms in the experiment has been described with a Lévy distribution, with a divergent variance. However, our results show that the mean-square displacement is finite for any finite measurement time. These seemingly contradicting findings are related to the well-known dilemma of whether Lévy flights are at all physical, since they predict diverging mean-square displacement, which must be tamed [4,18]. We speculate that the Lévy distribution found in the experiment describes the center part of the packet, which eventually is cut off to give a finite mean-square displacement. Furthermore, using our results one can estimate the time in which the atoms remain within a finite domain, which is of course crucial for experiments. Experimentally one may also control the depth of the optical potential, here modeled with \tilde{U}_0 , and hence explore the nontrivial dependence of our results on this parameter. We will elaborate on these interesting points in a longer publication.

Discussion.—Aging correlation functions and ergodicity breaking typically describe glassy dynamics [19,20] (and Refs. therein). Our work shows that aging and ergodicity breaking can be found also for simple Markovian dynamics, without the need to introduce heavy-tailed waiting times into the kinetic scheme, nor disorder or many-body physics. The aging correlation function (16) has a signature

TABLE I. Scaling behavior of $\langle \bar{x}^2(t) \rangle$ and $\langle x^2(t) \rangle$ for various values of $\tilde{U}_0 = U_0/(k_B T)$.

	$\langle \bar{x}^2(t) \rangle$	$\langle x^2(t) \rangle$
$\tilde{U}_0 < 1$	t	t
$1 < \tilde{U}_0 < 3$	$t^{(3-\tilde{U}_0)/2}$	$t^{(3-\tilde{U}_0)/2}$
$3 < \tilde{U}_0 < 5$	$t^{(3-\tilde{U}_0)/2}$	t^0
$5 < \tilde{U}_0$	t^{-1}	t^0

that is very different than most previous work. The prefactor $\langle x^2(t_1) \rangle$ grows with time, and hence we call it superaging. This is in contrast to normal aging where the correlation function is of the form $C(t_2, t_1) = \langle x^2 \rangle_{\text{eq}} f(t_2/t_1)$ with a finite equilibrium value $\langle x^2 \rangle_{\text{eq}}$. A similar non-normal aging behavior, albeit with a logarithmic time dependence, has been found in Sinai's model of diffusion in a random environment [19]. Unlike previous scenarios to ergodicity breaking, the amplitude of the stochastic process $x(t)$ in our work increases with time, since the particle explores more and more of the tails of the equilibrium PDF as time goes on. Thus rare events where the amplitude $x(t)$ of the Markovian process attains a large value are responsible for the nonergodic behavior. This is clearly related to the power-law tail of the equilibrium steady state $P^{\text{eq}}(x) \propto |x|^{-\tilde{U}_0}$. More importantly, physical systems with fat tailed equilibrium states are common and hence this type of ergodicity breaking may find broad applications.

This work was supported by the Israel Science Foundation, the Emmy Noether Program of the DFG (Contract No LU1382/1-1) and the cluster of excellence Nanosystems Initiative Munich.

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