Field-Induced Dispersion in Subdiffusion

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We discuss the response of continuous-time random walks to an oscillating external field within the generalized master equation approach. We concentrate on the time dependence of the two first moments of the walker's displacement. We show that for power-law waiting-time distributions with $0 < \alpha < 1$ corresponding to a semi-Markovian situation showing nonstationarity, the mean particle position tends to a constant; namely, the response to the external perturbation dies out. On the other hand, the oscillating field leads to a new additional contribution to the dispersion of the particle position, proportional to the square of its amplitude and growing with time. These new effects, amenable to experimental observation, result directly from the nonstationary property of the system.

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Continuous-time random walks (CTRWs) with on-site waiting-time distributions being power laws lacking the first moment have been shown to provide a powerful tool to describe systems which display subdiffusion [1]. These subdiffusive CTRWs are non-Markovian (semi-Markov) processes characterized by nonstationarity. Examples are dispersive charge transport in disordered semiconductors, contaminants transport by underground water, motion of proteins through cell membranes and many others (see, e.g., [2–4] for reviews and popular accounts). The nonstationarity mentioned above underlies the effects of aging which have been extensively studied in recent years [5–9]. Aging in glassy systems and in systems of quantum dots, as examples, has been formulated in terms of CTRWs. In the absence of time-dependent external perturbations CTRW is a process subordinated to simple random walk, thus leading to the description within a framework of fractional Fokker-Planck equations [2,3,10–12].

Here we discuss a different aspect of aging, based on the response of a CTRW system to a time-dependent field. Such a response to time-dependent fields is a delicate problem, which has not been explored in detail. The results show again that the subdiffusive nature of the problem leads to dramatic deviations when compared to the ordinary Markovian case. Here we investigate the response of a particle which performs a nonstationary CTRW to an external oscillating field by calculation the first two moments of its displacement. New effects are observed which are absent in the Markovian case and should be amenable to experimental observation.

Let us first describe the model adopted throughout this work. In a biased decoupled CTRW a particle arriving to a site i at time t' stays there for a sojourn time t, distributed with the probability density function (PDF) $\psi(t)$ independent of external perturbation. This waiting-time PDF is considered to follow a power-law $\psi(t) \propto \tau_0^\alpha t^{-1-\alpha}$, where τ_0 gives the typical time scale for a jump. Leaving a site it makes a random step of length a in either direction. This step is assumed to be instantaneous on the time scale of typical waiting times and changes of external parameters; the direction of this step can be biased by the time-dependent external force $f(t)$. In what follows we turn to dimensionless units and measure length in units of a and time in units of τ_0. The probabilities of going to the right $w_{i-1,i}(t)$ (from site $i-1$ to site i) and to the left (from site $i+1$ to site i) are assumed to be

$$w_{i-1,i}(t) = \frac{1}{2} + \frac{\mu}{2} f(t); \qquad w_{i+1,i}(t) = \frac{1}{2} - \frac{\mu}{2} f(t). \quad (1)$$

Our description of response of CTRW to external fields is based on generalized master equation (GME) approach. Several different ways to derive the corresponding equations are known in the literature (e.g., [13–17]); the one especially fitted to describing response to time-dependent fields is given in [18]. For the sake of completeness, we give here the sketch of this derivation. The GME follows from two balance conditions: the probability conservation in a given state and under transitions between different states.

The probability balance for the site k reads

$$\dot{p}_k(t) = j_k^+(t) - j_k^-(t), \quad (2)$$

(where the dot denotes the time derivative) with $j_k^\pm(t)$ denoting the gain and loss currents for a site. A particle leaving its site k at time t either was in k from the very beginning or arrived at k at some $0 < t' < t$ so that

$$j_k^-(t) = \psi(t)p_k(0) + \int_0^t \psi(t-t')j_k^+(t')dt'$$
$$= \psi(t)p_k(0) + \int_0^t \psi(t-t')[\dot{p}_k(t') + j_k^-(t')]dt', \quad (3)$$

where in the second line Eq. (2) was used. The formal solution to this equation can be expressed through an integro-differential operator

$$j_k^-(t) = \hat{\Phi} p_k(t) = \frac{d}{dt}\int_0^t M(t-t')p_k(t')dt' \quad (4)$$

with the memory kernel $M(t)$ given by its Laplace transform

$$\tilde{M}(u) = \frac{\tilde{\psi}(u)}{1-\tilde{\psi}(u)}. \quad (5)$$

The probability conservation for transitions between different sites gives the relation between the gain current in the state k and loss currents at neighboring sites:

$$j_k^+ = w_{k-1,k}(t)j_{k-1}^- + w_{k+1,k}(t)j_{k+1}^-. \quad (6)$$

Inserting the corresponding expressions into the first balance equation gives a GME for $p_k(t)$:

$$\dot{p}_k(t) = w_{k-1,k}(t)\hat{\Phi} p_{k-1}(t) + w_{k+1,k}(t)\hat{\Phi} p_{k+1}(t) - \hat{\Phi} p_k(t). \quad (7)$$

Note that the integro-differential operator $\hat{\Phi}$ does not commute with the function of time $w_{ij}(t)$.

Using now Eq. (1) for the transition probabilities and passing to the continuum limit we get a generalized Fokker-Planck equation

$$\frac{\partial}{\partial t}p(x,t) = \left[-\mu f(t)\nabla + \frac{1}{2}\Delta\right]\frac{d}{dt}\int_0^t M(t-t')p(x,t')dt'. \quad (8)$$

For a Markovian random walk process with exponential waiting-time distribution (corresponding to $\alpha = 1$ and thus to $M(t) = 1$) this equation reduces to a usual time-dependent Fokker-Planck equation. For power-law waiting-time distributions $\psi(t) \propto t^{-1-\alpha}$ with $0 < \alpha < 1$ one gets $M(t) \propto t^{\alpha-1}$ and the integro-differential operators on the right-hand side of this equation get proportional to the operator of fractional Riemann-Liouville derivative $\frac{d}{dt}\int_0^t M(t-t')g(t')dt' \propto {}_0D_t^{1-\alpha}g(t)$. This is exactly the case we now concentrate on.

The CTRWs with $0 < \alpha < 1$ are known to show a variety of phenomena connected with nonstationarity (related also to the so-called aging property [5–7,19–22]). One of the manifestations of aging is the decay of response of the system to an alternating or pulsed field in course of the time (i.e., when its age grows); see Ref. [6]. Here we consider the response of the system to a sinusoidal external field $f(t)$. We start from Eq. (8) and consider the moments $m_n(t) = \int_{-\infty}^{\infty} x^n p(x,t)dx$ of the probability distribution of particle's positions. These moments can be easily obtained by multiplying both sides of Eq. (8) by x^n and integration. Assuming the system to be infinite and spatially homogeneous we get by partial integration

$$\int_{-\infty}^{\infty} x^n \nabla p(x,t')dx = -n m_{n-1}(t') \quad (9)$$

and

$$\int_{-\infty}^{\infty} x^n \Delta p(x,t')dx = n(n-1)m_{n-2}(t') \quad (10)$$

(for $n \geq 2$). Thus, general equations for the moments are given by

$$\dot{m}_n(t) = n\mu f(t)\hat{\Phi} m_{n-1}(t) + \frac{n(n-1)}{2}\hat{\Phi} m_{n-2}(t). \quad (11)$$

To be able to use these equations in the whole range of $0 \leq n < \infty$ one can formally put $m_0 = 1$ and $m_{-1}(t) = 0$. The equations for the first moment (mean displacement) and the second (dispersion) read:

$$\dot{m}_1(t) = \mu f(t)\hat{\Phi} 1 = \mu f(t)M(t) \quad (12)$$

and

$$\dot{m}_2(t) = 2\mu f(t)\hat{\Phi} m_1(t) + M(t). \quad (13)$$

Note that for semi-Markovian cases with $0 < \alpha < 1$ $M(t) \propto t^{\alpha-1}$ is a decaying function of time. Therefore, the response $\dot{m}_1(t)$ to the perturbation vanishes in course of the time leading to the effect we call "death of linear response" in systems showing subdiffusion.

The second moment, $m_2(t) = \int_0^t \dot{m}_2(t')dt'$, consists essentially of two contributions, $m_2(t) = \sigma_1^2(t) + \sigma_2^2(t)$, the one depending on the external perturbation (through the first moment m_1)

$$\sigma_1^2(t) = 2\int_0^t dt' \mu f(t')\frac{d}{dt'}\int_0^{t'} M(t'-t'')m_1(t'')dt'' \quad (14)$$

and of the field-independent purely (sub)diffusive contribution

$$\sigma_2^2(t) = \int_0^t M(t')dt'. \quad (15)$$

Let us discuss the overall structure of expressions for the first and the second moment for the case of a periodic force $f(t) = f_0 \sin\omega t$. The first moment is

$$m_1(t) = \mu f_0 \int_0^t dt' \sin\omega t' M(t')dt' = \mu f_0 \int_0^t dt' \frac{e^{i\omega t'} - e^{-i\omega t'}}{2i} M(t')dt'. \quad (16)$$

Turning to the Laplace domain we get

$$\tilde{m}_1(u) = \frac{\mu f_0}{2iu}[\tilde{M}(u-i\omega) - \tilde{M}(u+i\omega)] \quad (17)$$

as it follows from the shift theorem. The asymptotic behavior of the first moment then follows straightforwardly. For $u \to 0$ (corresponding to $t \to \infty$) we have

$$\tilde{m}_1(u) = \frac{\mu f_0}{2iu}[\tilde{M}(-i\omega) - \tilde{M}(i\omega)] \quad (18)$$

being the Laplace transform of a constant $m_1(\infty) = \mu f_0 \,\mathrm{Im}\,\tilde{M}(-i\omega)$. The Laplace transform of the field-dependent contribution to the second moment (again obtained by using the shift theorem) reads:

$$\tilde{\sigma}_1^2(u) = -\frac{\mu^2 f_0^2}{2u}\{\tilde{M}(u-i\omega)[\tilde{M}(u-2i\omega)-\tilde{M}(u)] - \tilde{M}(u+i\omega)[\tilde{M}(u)-\tilde{M}(u+2i\omega)]\}. \quad (19)$$

To obtain the asymptotic behavior of $\tilde{\sigma}_1^2(u)$ it is enough to note that for power-law waiting-time distributions with $0<\alpha<1$, $\tilde{M}(u)\propto u^{-\alpha}$ which diverges when $u\to 0$. Thus, the leading contribution to $\tilde{\sigma}_1^2(u)$ is

$$\tilde{\sigma}_1^2(u) \simeq \mu^2 f_0^2 \frac{\tilde{M}(-i\omega)\tilde{M}(u)+\tilde{M}(i\omega)\tilde{M}(u)}{2u} = \mu^2 f_0^2 \,\mathrm{Re}\,\tilde{M}(i\omega)\frac{\tilde{M}(u)}{u}. \quad (20)$$

This means that the asymptotic growth of the field-dependent contribution to the dispersion is given by $\sigma_1^2(t)\propto \mu^2 f_0^2 \int_0^t M(t')dt' \propto \mu^2 f_0^2 t^\alpha$.

The most important feature of this result is that although the first moment of the distribution stagnates, the field-dependent contribution to the dispersion continues growing, a manifestation of a new effect, specific for nonstationary CTRWs, namely, the field-induced dispersion. This growing contribution is absent for the Markovian and asymptotically Markovian processes ($\alpha = 1$) only due to the fact that the corresponding prefactor $\mathrm{Re}\,(i\omega)^{-\alpha}$ vanishes.

To get an impression on the overall behavior of the first and the second moments let us consider the special case $\alpha = 1/2$. In our numerical example we set $\omega = 1$. Substituting $M(t) = 1/\sqrt{t}$ in Eq. (16) yields

$$m_1(t) = \mu f_0 \sqrt{2\pi} S\left(\sqrt{\frac{2t}{\pi}}\right) \quad (21)$$

involving the Fresnel sinus-integral $S(z) = \int_0^z dt \sin(\pi t^2/2)$. The behavior of $m_1(t)$ is shown in Fig. 1. We see that the susceptibility of the system to an external force decays in course of the time; namely, its linear response dies out. The final value of the first moment is mostly determined by the value of the external perturbation at short times, when the system was "young," the result of what was called "Freudistic" memory of aging systems in Ref. [7].

The behavior of the field-dependent contribution to the second moment follows from Eq. (19). The Laplace transform of $M(t) = 1/\sqrt{t}$ is $\tilde{M}(u) = \sqrt{\pi/u}$. Hence Eq. (19) contains four terms of the form $u^{-1}/\sqrt{(u+a)(u+b)}$, where a, b are 0, $\pm i\omega$, $\pm 2i\omega$. Using the shifting theorem one can complete squares. Using that the Laplace transform of the Bessel function $J_0(at)$ is $1/\sqrt{u^2+a^2}$, it follows that

$$\sigma_1^2(t) = 2\pi\mu^2 f_0^2 \int_0^t J_0\left(\frac{\omega t'}{2}\right)\sin\left(\frac{\omega t'}{2}\right)\sin(\omega t')dt'. \quad (22)$$

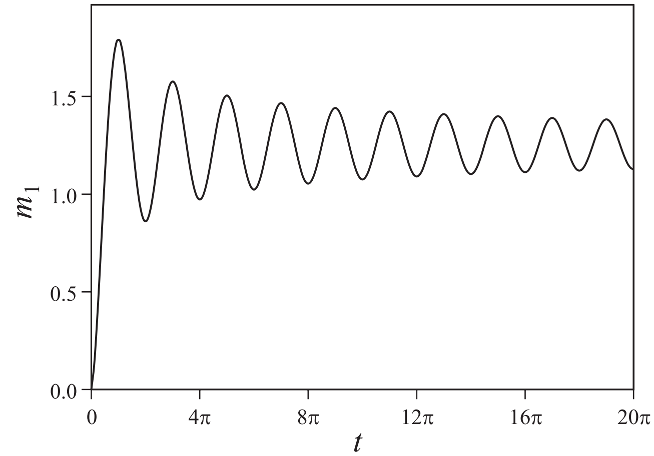

FIG. 1. The mean displacement $m_1(t)$ (measured in units of μf_0) in a CTRW model with $\alpha = 1/2$ as a function of time for $\omega = 1$. Note that for t large the displacement stagnates: the linear response of the system to external sinusoidal field dies.

The numerical evaluation of the integral is given in Fig. 2 showing $[\sigma_1^2(t)]^{1/\alpha}$, i.e., the square of the corresponding expression, Eq. (22). The leading contribution t^α to the overall behavior corresponds to the linear growth of $[\sigma_1^2(t)]^{1/\alpha}$. Interesting is the subleading asymmetric oscillatory behavior overplayed on this overall growth. The amplitude of these oscillations decays on a linear plot but stays constant in the one presenting the square of the function. This means that the decay of the subleading term follows essentially $t^{-1/2}$. This behavior of the leading and subleading terms follows immediately from the asymptotic expansion of $J_0(x)$. The field-independent subdiffusion contribution $\sigma_2^2(t)$ grows as $t^{1/2}$ according to Eq. (15).

Let us summarize our findings. We discussed the behavior of a particle performing continuous-time random walks with a power-law distribution of waiting times lacking the

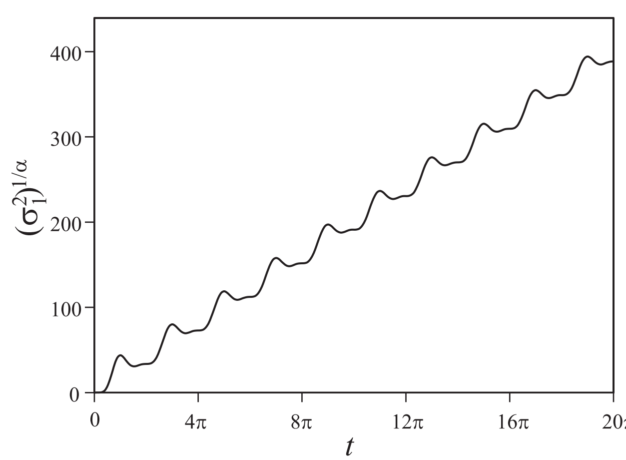

FIG. 2. Shown is the square of $\sigma_1^2(t)$ (measured in units of $\mu^2 f_0^2$) as a function of t for $\alpha = 1/2$ and $\omega = 1$.

first moment ($\psi(t) \propto t^{-1-\alpha}$ with $0 < \alpha < 1$) under the influence of oscillating external field. Using the approach based on the generalized master equation we derive equations for the first two moments of the displacement. The first moment of the displacement stagnates, an effect we term "death of linear response." The second moment, on the contrary, grows as t^{α} and contains, in addition to the normal (sub-)diffusion contribution, a field-induced contribution, proportional to the square of the external field. This new effect which shows up in nonstationary CTRW is absent in the Markovian case ($\alpha = 1$) since the corresponding prefactor vanishes.

The conditions assumed in the Letter are those used in time-of-flight experiments and therefore the predictions derived here are amenable to an experimental verification. All experiments up to date have been done in the presence of time-independent fields. As shown here the generalization to the time-dependent case is far from trivial due to the nonstationary nature of the transport.

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