Death of linear response and 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 displacements. 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, and 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 non-stationary 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. These subdiffusive CTRWs are non-Markovian (semi-Markov) processes characterized by nonstationarity (aging). 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. [1, 2, 3] for reviews and popular accounts). 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 [7, 8, 9, 10, 11]. Here we investigate the response of non-Markov) processes characterized by nonstationarity (aging) to simple random walk, thus leading to the description within a framework of fractional Fokker-Planck equations [7, 8, 9, 10, 11]. Here we investigate the response of continuous time random walks to an oscillating external field within the generalized master equation approach. Several different ways to derive the corresponding equations are known in the literature (e.g. [12, 13, 14, 15, 16]), the one especially fitted to describing response to time-dependent fields is given in [17]. 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

$$\dot{p}_k = j_k^+ - j_k^-,$$  (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

$$j_k^-(t) = \psi(t)p_k(0) + \int_0^t \psi(t-t')j_k^+(t')dt',$$  (3)

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

$$j_k^-(t) = \tilde{\psi}(t)p_k(0) + \int_0^t \tilde{\psi}(t-t') M(t-t')p_k(t')dt',$$  (4)

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

$$M(u) = \frac{\tilde{\psi}(u)}{1 - \tilde{\psi}(u)},$$  (5)
The probability conservation for transitions between different sites give the relation between the gain current in the state $k$ and loss currents at neighboring sites:

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

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

$$\dot{p}_k(t) = w_{k-1,k}(t)\dot{p}_k(t) + w_{k+1,k}(t)\dot{p}_k(t) - \dot{p}_k(t). \hspace{1cm} (7)$$

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

Using now Eq. (6) for the transition probabilities and passing to the continuum limit we get a generalized Fokker-Planck equation

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

For 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 $\frac{d}{dt} \int _0^t \phi(t') dt' \propto 0 D_1^{\alpha-\beta} f(t)$. 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 non-stationarity (related also to the so-called aging property \textsuperscript{2} \textsuperscript{3} \textsuperscript{4} \textsuperscript{5} \textsuperscript{6}). 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. \textsuperscript{8}. Here we consider the response of the system to a sinusoidal external field $f(t)$. We start from Eq. (5) and consider the moments of $p(x,t)$, $m_n(t) = \int_{-\infty}^{\infty} x^n p(x,t) dx$ of the probability distribution of particle’s positions. These moments can be easily obtained by multiplying both sides of Eq. (5) by $x^n$ and integration. Assuming the system to be infinite and spatially homogeneous we get by partial integration

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

and

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

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

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

To be able to use these equations in the whole range of $0 \leq m < \infty$ 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:

$$\dot{m}_1(t) = \mu f(t) \Phi 1 = \mu f(t) M(t), \hspace{1cm} (12)$$

and

$$\dot{m}_2(t) = 2\mu f(t) \Phi m_1(t) + M(t). \hspace{1cm} (13)$$

Note that for semi-Markovian cases with $0 < \alpha < 1$ $M(t) \propto t^{\alpha-1}$ is the 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 $m_1$)

$$\sigma_1^2(t) = 2 \int_0^t dt' \mu f(t') \frac{d}{dt} \int_0^{t'} M(t'-t'')m_1(t'')dt'' \hspace{1cm} (14)$$

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

$$\sigma_2^2(t) = \int_0^t M(t') dt'. \hspace{1cm} (15)$$

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:

$$m_1(t) = \mu f_0 \int_0^t dt' \sin \omega t' M(t') \hspace{1cm} (16)$$

Turning to the Laplace domain we get

$$\tilde{m}_1(u) = \frac{\mu f_0}{2iu} \left[ \tilde{M}(u+i\omega) - \tilde{M}(u+i\omega) \right] \hspace{1cm} (17)$$

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

$$\tilde{m}_1(u) = \frac{\mu f_0}{2iu} \left[ \tilde{M}(-i\omega) - \tilde{M}(i\omega) \right] \hspace{1cm} (18)$$

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

$$\tilde{\sigma}_1^2(u) = -\frac{\mu f_0^2}{4u} \left\{ \tilde{M}(u+i\omega) \left[ \tilde{M}(u-2i\omega) - \tilde{M}(u) \right] - \tilde{M}(u+i\omega) \left[ \tilde{M}(u) - \tilde{M}(u+2i\omega) \right] \right\}. \hspace{1cm} (19)$$
To obtain the asymptotic behavior of $\bar{\sigma}_1^2(u)$ it is enough to note that for power-law waiting time distributions with $0 < \alpha < 1$, $\bar{M}(u) \propto u^{-\alpha}$ which diverges when $u \to 0$. Thus, the leading contribution to $\bar{\sigma}_1^2(u)$ is

$$
\bar{\sigma}_1^2(u) \simeq \mu^2 f_0^2 \frac{\bar{M}(\iota \omega) \bar{M}(u) + \bar{M}(\iota \omega) \bar{M}(u)}{4u} = \mu^2 f_0^2 \text{Re}(\iota \omega) \frac{\bar{M}(u)}{u}.
$$

(20)

This means that the asymptotic growth of the field-dependent contribution to the dispersion is given by $\bar{\sigma}_1^2(t) \propto \mu^2 f_0^2 \int_0^t M(t')dt' \propto \mu^2 f_0^2 t^\alpha$.

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 non-stationary 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 $\text{Re}(\iota \omega)^{-\alpha}$ 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 $\omega = 1$. In this case

$$
\dot{m}_1(t) = \mu f_0 \sin t \frac{d}{dt} \int_0^t (t - t')^{-1/2}dt'
$$

(21)

$$
= \Gamma(1/2) \mu f_0 \sin t \frac{D_t^{1/2}}{1} = \mu f_0 t^{-1/2} \sin t.
$$

Note that the fractional derivative of the constant does not vanish. From this expression we see that the susceptibility of the system to an external force decays in course of the time as $t^{-1/2}$; namely its linear response dies out. Integrating this expression over time we get the analytical form for $m_1(t)$

$$
m_1(t) = \mu f_0 \sqrt{2\pi} S(\sqrt{t})
$$

(22)

involving the Fresnel sinus-integral $S(x) = (1/\sqrt{2\pi}) \int_0^x dt \sin (t)/\sqrt{t}$. The Fresnel integral tends to a constant value for large values of its argument; 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. 3. The behavior of $m_1(t)$ is shown in Fig. 1.

The behavior of the field-dependent contribution to the second moment is

$$
\dot{\sigma}_1^2(t) = \mu f_0 \int_0^t dt' \sin t' \frac{d}{dt} \int_0^t (t' - t'')^{-1/2} m_1(t'')dt''.
$$

(23)

Integrating this expression by parts twice we get:

$$
\sigma_1^2(t) = 2\mu^2 f_0^2 \left[ \sin(t) \Psi(t) - \int_0^t \Psi(t') \cos(t')dt' \right] .
$$

(24)

FIG. 1: The mean displacement $m_1(t)$ (measured in units of $\mu f_0$) 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.

with

$$
\Psi(t) = \int_0^t \sqrt{(t - t')/t'} \sin t' dt'
$$

(25)

The numerical evaluation of the integral is shown in Fig. 2 showing $|\sigma_1^2(t)|^{1/\alpha}$, i.e. the square of the corresponding expression, Eq. (24). The leading contribution $t^\alpha$ to the overall behavior corresponds to the overall linear growth of $|\sigma_1^2(t)|^{1/\alpha}$. Interesting is the subleading asymmetric oscillatory behavior overlaid on this overall growth. The amplitude of these oscillations decays on a linear plot, but seems to stay constant in the one presenting the square of the function. This means that the decay of the subleading term follows essentially $t^{-1/2}$. The field-independent subdiffusion contribution $\sigma_2^2(t)$ grows as $t^{1/2}$ according to Eq. (13).

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 first moment ($\psi(t) \propto t^{-\alpha}$ 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 $t^\alpha$ 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 non-stationary CTRW is absent in the Markovian case ($\alpha = 1$) since the corresponding prefactor vanishes.
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 $\omega = 1$.

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