Aging correlation functions of the interrupted fractional Fokker-Planck propagator

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(Received 1 September 2006; accepted 9 November 2006; published online 29 December 2006)

The authors explore aging in a general semi-Markov process with arbitrary waiting time distributions and discuss the role of trapping phenomenon in glasses in determining the waiting time distributions. In certain limits, they obtain a two-time propagator for the fractional Fokker-Planck equation through coordination, but one can generalize the construction to incorporate cutoffs in the aging of the system, i.e., the interrupted aging. The construction allows the incorporation of cutoffs in the aging of the system, and the exploration of signatures of aging effects in systems that eventually achieve equilibrium. Cutoffs in aging effects are illustrated for interrupted fractional diffusion in a harmonic potential and for the on-time probability of a quantum dot system. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2403874]

INTRODUCTION

Dispersive transport has been observed in experiments as diverse as conduction in amorphous semiconductors, diffusion in glasses and on surfaces, and motions in cells and proteins.[1–7] The general feature of dispersive transport is complex nonexponential temporal relaxation, which can be numerically fit with a power-law tail or a stretched exponential. Phenomenological dispersive transport models often incorporate the nonexponential relaxation into diffusion problems by replacing the exponential waiting time distribution of traditional diffusion processes with a more complex waiting time distribution through subordination, which results in a continuous time random walk (CTRW) or the related fractional Fokker-Planck equation (FFPE) for subdiffusion.[8–10] The origin of the complex waiting time has be extensively explored in glassy systems,[11] and attributed to activation processes with randomly distributed energy barriers.[12–14] The power-law waiting time distribution functions have also been observed in quantum dots, proteins, and other biological systems. However, these systems appear to obey power-law statistics over several decades before achieving an equilibrium state where the time correlation becomes stationary.[15,16] Examples include quantum dot blinking experiments that show a laser power dependent cutoff in the blinking statistics,[15] and photon emission statistics in proteins, when complex correlation function shows non-power-law long time relaxation.[16] Since aging would cause the system behavior to change over time, additional processes that can truncate the aging in the system must always be present. It is therefore not surprising to see a cutoff in the power-law correlations observed in experiments.[15,16] To extend applications of FFPE to real systems, where aging processes are interrupted, requires us to develop a general description of trapping processes and to examine signatures in aging measurements.[12–14,17]

GENERAL TWO-TIME SEMI-MARKOV CONSTRUCTION

A semi-Markov process is the most general description of trapping models with complex waiting times. In a semi-Markov process, the probability distributions of making a transition from a state j to state i at a time t is given by the matrix element $Q_{ij}(t)dt$ with $Q_{ij}(t) \geqslant 0$ for $t>0$ and $\Sigma_i \int dt Q_{ij}(t) \leqslant 1$ for all j.[8] If $\Sigma_i \int dt Q_{ij}(t) < 1$ the particle may stay at site i permanently, i.e., i is a trap. The single time propagator for this process in the Laplace domain, $t \rightarrow s$, is

$$\mathbf{G}_{ij}(s) = \frac{1-\Sigma_k Q_{ki}(s)}{s}[\mathbf{I}-\mathbf{Q}(s)]^{-1}_{ij}, \tag{1}$$

where $\mathbf{I}$ is the identity matrix and $\mathbf{Q}$ is the complete matrix of $Q_{ij}(s)$. The $[\mathbf{I}-\mathbf{Q}(s)]^{-1}_{ij}$ is the Laplace transform of the probability of jumping into the state i at time t given the initial state at j and $[1-\Sigma_k Q_{ki}(s)]/s$ is the probability of not jumping out of the final state.[8] In Eq. (1), the hopping process begins at $t=0$, so the beginning of the hopping process is well defined, e.g., the temperature quenching time in glass forming experiments. More generally, the measurement does not begin at quenching time. In order to explore this scenario, we examine a two-time propagator in double Laplace space $t_1 \rightarrow s_1$ and $t_2 \rightarrow s_2$,[8]

$$\begin{aligned}\mathbf{G}_{ijk}(s_1,s_2) &= \frac{1-\Sigma_l Q_{li}(s_2)}{s_2} \\ &\times \sum_m [\mathbf{I}-\mathbf{Q}(s_2)]^{-1}_{im} \frac{[Q_{mj}(s_1)-Q_{mj}(s_2)]}{s_2-s_1} \\ &\times [\mathbf{I}-\mathbf{Q}(s_1)]^{-1}_{jk} \\ &+ \delta_{ij} \frac{\left[\left\{[1-\Sigma_l Q_{lj}(s_1)]/s_1\right\} - \left\{[1-\Sigma_l Q_{lj}(s_2)]/s_2\right\}\right]}{s_2-s_1} \\ &\times [\mathbf{I}-\mathbf{Q}(s_1)]^{-1}_{jk}. \end{aligned} \tag{2}$$

In the first term on the right hand side of the above equation, $[1-\Sigma_l Q_{li}(s)]/s$ and $[\mathbf{I}-\mathbf{Q}(s)]^{-1}$ correspond to the same processes in Eq. (1), but $[Q_{mj}(s_1)-Q_{mj}(s_2)]/(s_2-s_1)$ corresponds to the jump that spans both time intervals t_1 and t_2. In the second term, the coefficient before the matrix element corresponds to failing to make a jump during the second time interval.

A MICROSCOPIC TRAPPING PICTURE FOR Q(*s*)

A possible origin of $\mathbf{Q}(s)$ is the trapping process observed in glassy systems.[11,13,14] If the experiment monitors a few labeled tracer particles in a glassy matrix, the tracers spend most of their time trapped in cages formed by the matrix and can only move when a trap rearranges through an activation process. If the rearrangements are large enough, we would not expect correlations between the trapping times and/or displacements.[13,14] The process described above violates the fluctuation dissipation theorem on short time scales (high frequency). The traps arrest the motions of the particles so any force applied on time scales shorter than the trapping time will not result in any response. For forces applied on time scales longer than the trapping time and for length-scales larger than the typical tracer particle displacement, the fluctuation dissipation theory (FDT) is valid. The FDT violations of many CTRW processes come from making the mean time larger than the observation time so that the FDT never becomes valid.[10]

The trapping times would be site specific with no directional dependence, $Q_{ij}(t)=W_{ij}\psi_j(t)$. The matrix $\mathbf{W}$ controls the directional aspects of the particle motions and can be approximated as $\mathbf{W}\approx e^{\mathbf{L}_{\mathrm{FP}}\Delta t}$, where Δt is a small time step and $\mathbf{L}_{\mathrm{FP}}$ can be a kinetic matrix or an operator, i.e.,

$$\mathbf{L}_{\mathrm{FP}}\rho(x,t)=\nabla\cdot[D(x)\,\nabla\,\rho(x,t)]+\nabla\cdot[\rho(x,t)\,\nabla\,V(x)],$$

with diffusion constant $D(x)$ and potential $V(x)$. Bouchaud and Georges[13] and Mothus and Bouchaud[14] introduced a waiting time through an activated process with possibly site specific random energy barriers, $\phi_j(t)=\int dE_j \phi(t|E_j)P(E_j)$ with $\phi(t|E_j)\approx\tau_{0j}^{-1}e^{-\beta E_j}\exp(\tau_{0j}^{-1}e^{-\beta E_j}t)$ and τ_{j0} is a weakly temperature dependent prefactor.[13,14] If the energy barriers are exponentially distributed for each site $P(E_j)=\alpha_j e^{-\alpha_j E_j}$ the waiting time has an asymptotic power-law tail $\phi_j(t)\sim\tau_{0j}^{-1}(\alpha_j/\beta)\Gamma[1+(\alpha_j/\beta)][t/\tau_{0j}]^{-(1+\alpha_j/\beta)}$. The distribution of barrier heights $P(E_j)$ is phenomenologically based on extreme value arguments, which are not easily justified due to the sensitivity of the waiting time distribution to the nonuniversal tails of the barrier distribution.[18]

The sensitivity to the nonuniversal barrier height distribution function suggests that the tail of the waiting time distribution should be modified. Xia and Wolynes introduced a cutoff in the free energy barriers of glasses because there are competing processes resulting in a rearrangement with a finite sized barrier height.[12] We introduce a similar cutoff by adding a competing pathway of fixed energy barrier height, i.e., take the fastest times between an exponential process, $k_j e^{-k_j t}$ and $\phi_j(t)$. This approach is similar to the coupling scheme of first order and power-law dynamics discussed elsewhere.[19,20] Having only two such pathways is a simplification that follows if there are two possible classes of rearrangements to the system, ϕ corresponding to a broad distribution of energy barriers and ke^{-kt} corresponding to a narrow distribution of deep energy barriers. For the case of an exponential process competing with power-law waiting time distribution, the interrupted waiting time is[20]

$$\psi_j(t)=\phi_j(t)e^{-k_j t}+k_j e^{-k_j t}\int_t^\infty \phi_j(\tau)d\tau.$$

If $\alpha_j=\alpha$, $\tau_{0j}=\tau_0$, and $k_j=k$ for all lattice sites, we can write an equation for the time evolution of the distribution of energy barrier,[13,14]

$$\partial_t P(E)=-(k+\tau_0^{-1}e^{-\beta E})P(E)+\alpha e^{-\alpha E}\int dE'(k+\tau_0^{-1}e^{-\beta E'})P(E'). \tag{3}$$

The high energy cutoff introduced by k creates a microscopic equilibrium for the distribution of energy barriers and the system achieves equilibrium in a finite amount of time, $t\approx k^{-1}$. The equilibrium distribution for barrier heights, $\partial_t\rho_{\mathrm{eq}}=0$ is $\rho_{\mathrm{eq}}(E)=\eta\alpha e^{-\alpha E}/(k+\tau_0^{-1}e^{-\beta E})$, where η is the normalization. On these long time scales we expect the system to be diffusive. For $\beta>\alpha$ and $k=0$ we recover the trapping result for a lack of equilibrium distribution.[13] The system is microscopically far away from equilibrium for time less than k^{-1} so the system is not in a linear regime for $t<k^{-1}$ and short time violations of the FDT should not be surprising. The traps arrest the motions on these short time scales and the system cannot respond, but the FDT becomes valid for time scales longer than the trapping time ($t>k^{-1}$).

SINGLE TIME PROPAGATOR

In order to study the properties and effects of the modified waiting time distribution, we perform a Laplace transform on the waiting time distribution,[20]

$$\psi_j(s)=\phi_j(s+k_j)+k_j\frac{1-\phi_j(s+k_j)}{s+k_j}. \tag{4}$$

In long time limit, we approximate $\phi_j(s)$ with a Lévy distribution, $\phi_j(s)=e^{-(\tau_j s)^{\gamma_j}}$, where $\gamma_j=\alpha_j/\beta$ is the power-law exponent of the original waiting time distribution and τ_j is a constant that defines the tail amplitude. Equation (4) allows us to write $\mathbf{Q}(s)=\mathbf{W}\boldsymbol{\psi}(s)$ and

$$\mathbf{G}(s)=\frac{\mathbf{I}-\boldsymbol{\psi}(s)}{s}\cdot[\mathbf{I}-\mathbf{W}\boldsymbol{\psi}(s)]^{-1}. \tag{5}$$

Substituting $\mathbf{W}=e^{\mathbf{L}_{\mathrm{FP}}\Delta t}$ and taking the limit Δt, $\tau_j\to 0$, such that $\Delta t/\tau_j^{\gamma_j}\to D_j^{\gamma}$ lead to Green's function for an anomalous diffusion process competing with a first order process,

$$\mathbf{G}(s)\approx[s-\mathbf{L}_{\mathrm{FP}}\mathbf{D}^{\gamma}(s+\mathbf{k})^{1-\gamma}]^{-1}, \tag{6}$$

where $\mathbf{D}^{\gamma}(s+\mathbf{k})^{1-\gamma}=\delta_{ij}D_j^{\gamma}(s+k_j)^{1-\gamma_j}$. If $\gamma_j=\gamma$ and $D_j^{\gamma}=D^{\gamma}$ are the same for all sites, Green's function becomes $\mathbf{G}(s)\approx[s-D^{\gamma}\mathbf{L}_{\mathrm{FP}}(s+\mathbf{k})^{1-\gamma}]^{-1}$ with corresponding Green's function equation, $s\mathbf{G}-\mathbf{I}=D^{\gamma}\mathbf{L}_{\mathrm{FP}}(s+\mathbf{k})^{1-\gamma}\mathbf{G}$. Notice that $\mathbf{L}_{\mathrm{FP}}$ is applied to the interrupted fractional derivative, $(s+\mathbf{k})^{1-\gamma}$ of Green's function, so spatial inhomogeneities in the interrup-

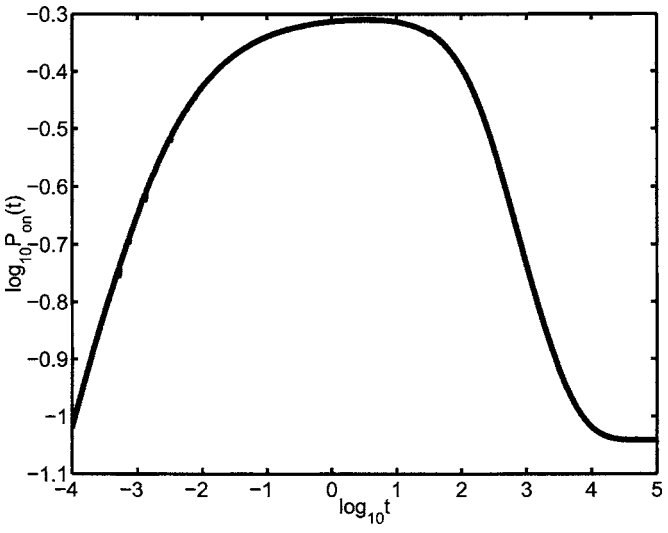

FIG. 1. The on probability for a quantum dot (QD) modeled by the interrupted fractional diffusion propagator. The initial condition assumes the renewal process begins at $t=0$ and the dot is off. Unlike a simple two state kinetic scheme that monotonically approaches equilibrium, the on probability shows a fast rise to an initial transient steady state before decaying to the long time equilibrium distribution.

tion rate $\mathbf{k}$ result in a change in the equilibrium population.

An extensively studied example of this effect on the equilibrium is the quantum dot, which has two states, *on* and *off*. The on time cutoff is experimental verified to be shorter than the *off* time cutoff.[15] If $\gamma_{\mathrm{on}}=\gamma_{\mathrm{off}}=\gamma$ the quantum dot Green's function is

$$\mathbf{G}_{\mathrm{QD}}=\begin{bmatrix} s+D_{\mathrm{on}}^{\gamma}(s+k_{\mathrm{on}})^{1-\gamma} & -D_{\mathrm{off}}^{\gamma}(s+k_{\mathrm{off}})^{1-\gamma} \\ -D_{\mathrm{on}}^{\gamma}(s+k_{\mathrm{on}})^{1-\gamma} & s+D_{\mathrm{off}}^{\gamma}(s+k_{\mathrm{off}})^{1-\gamma} \end{bmatrix}^{-1} \tag{7}$$

and the equilibrium on population is $p_{\mathrm{on}}(t\to\infty)=(D_{\mathrm{off}}^{\gamma}k_{\mathrm{off}}^{1-\gamma})/(D_{\mathrm{off}}^{\gamma}k_{\mathrm{off}}^{1-\gamma}+D_{\mathrm{on}}^{\gamma}k_{\mathrm{on}}^{1-\gamma})$. The $D_{\mathrm{on/off}}^{\gamma}$ term plays the role of determining the amplitude of the power-law tails versus the short time dynamics. When $\gamma=1$ the intensity of the quantum dot (QD) monotonically approaches equilibrium, but for $\gamma\neq 1$, the QD can show an initial rise to an intermediate plateau before monotonic decay to equilibrium, as shown in Fig. 1 for $D_{\mathrm{on/off}}=1$, $\gamma=1/2$, $k_{\mathrm{on}}^{-1}=100$, and $k_{\mathrm{off}}^{-1}=10\,000$.[15] The short time dynamics sufficiently scrambles the on and off probabilities and leads to the transient steady state, whereas the long time asymptotics cause the decay to the equilibrium value. The transient steady state has been observed in experiments and simulations.[21]

Unlike the QD example, if $k_j=k$ is the same for all j, the interrupted fractional derivative commutes with $\mathbf{L}_{\mathrm{FP}}$, and we recover the equilibrium distribution of the normal diffusion process as the equilibrium distribution of the interrupted process. For $k_j=k$, Green's function in Eq. (5) can also be reexpressed as

$$\mathbf{G}(s)=\int\frac{d\omega}{2\pi i}\frac{(s+k)^{\gamma-1}}{s(s+k)^{\gamma-1}+D^{\gamma}i\omega}\mathbf{G}_{\mathrm{FP}}(-\omega), \tag{8}$$

where $\mathbf{G}_{\mathrm{FP}}(\omega)=[-i\omega+\mathbf{L}_{\mathrm{FP}}]^{-1}$ is Green's function for the normal Fokker-Planck equation. This expression is the standard subordination result and can be rewritten in real time as[8]

$$\mathbf{G}(t)=\int_0^{\infty}dt'\,g(t',t)\mathbf{G}_{\mathrm{FP}}(t') \tag{9}$$

where $g(t',t)$ is the inverse Laplace transform, $s\to t$, of

$$g(t',t)=\mathcal{L}^{-1}[(D^{\gamma})^{-1}(s+k)^{\gamma-1}e^{-s(s+k)^{\gamma-1}(D^{\gamma})^{-1}t'}].$$

The interrupted fractional Green function is simply an average of normal diffusion Fokker-Planck Green's function over a stochastic distribution for t' determined by $g(t',t)$.

INTERRUPTED AGING AND THE TWO-TIME PROPAGATOR

The single time propagator assumes that the trapping process begins at $t=0$. More generally, the trapping process will begin at an earlier time than the observation time. Starting from Eq. (2) for the two-time semi-Markov propagator and following the single time propagator derivation gives the two-time propagator,

$$\begin{aligned}\mathbf{G}_{ijk}(s_1,s_2)=&\frac{1-\psi_i(s_2)}{s_2}\sum_m[\mathbf{I}-\mathbf{W}\boldsymbol{\psi}(s_2)]_{im}^{-1}\{\mathbf{W}_{mj}[\psi_j(s_1)\\&-\psi_j(s_2)]/(s_2-s_1)\}[\mathbf{I}-\mathbf{W}\boldsymbol{\psi}(s_1)]_{jk}^{-1}\\&+\delta_{ij}\frac{[\{[-\psi_j(s_1)]/s_1\}-\{[1-\psi_j(s_2)]/s_2\}]}{s_2-s_1}\\&\times[\mathbf{I}-\mathbf{W}\boldsymbol{\psi}(s_1)]_{jk}^{-1}.\end{aligned} \tag{10}$$

After performing the same limits as in Eq. (6), we have

$$\begin{aligned}\mathbf{G}_{ijk}(s_1,s_2)\approx&[s_2-\mathbf{L}_{\mathrm{FP}}\mathbf{D}^{\gamma}(s_2+\mathbf{k})^{1-\gamma}]_{ij}^{-1}\\&\times\frac{(s_2+k_j)^{\gamma_j}-(s_1+k_j)^{\gamma_j}}{s_2-s_1}(s_1+k_j)^{1-\gamma_j}\\&\times[s_1-\mathbf{L}_{\mathrm{FP}}\mathbf{D}^{\gamma}(s_1+\mathbf{k})^{1-\gamma}]_{jk}^{-1}\\&+\delta_{ij}\frac{(s_1+k_j)^{\gamma_j-1}-(s_2+k_j)^{\gamma_j-1}}{s_2-s_1}(s_1+k_j)^{1-\gamma_j}\\&\times[s_1-\mathbf{L}_{\mathrm{FP}}\mathbf{D}^{\gamma}(s_1+\mathbf{k})^{1-\gamma}]^{-1}.\end{aligned} \tag{11}$$

The $[s-\mathbf{L}_{\mathrm{FP}}\mathbf{D}^{\gamma}(s+\mathbf{k})^{1-\gamma}]$ terms are simply the single time interrupted fractional diffusion Green's functions. The $(s+\mathbf{k})^{1-\gamma}$ term is the interrupted version of the fractional derivative operator. The first s_1 vs s_2 difference term $[(s_2+\mathbf{k})^{\gamma}-(s_1+\mathbf{k})^{\gamma}]/[s_2-s_1]$ corresponds to the first jump made during the second time interval, and the second term $[(s_1+\mathbf{k})^{\gamma-1}-(s_2+\mathbf{k})^{\gamma-1}]/[s_2-s_1]$ corresponds to the failure to make a jump during the second time interval. After the first jump, the particle resumes normal interrupted fractional diffusion. We will discuss further details about these two terms after we introduce a memory kernel expression for these terms. Similar to the derivation of the single time propagator, if $k_j=k$, $D_j^{\gamma}=D^{\gamma}$, and $\gamma_j=\gamma$ are independent of the lattice position, the two-time propagator can be rewritten as

$$G_2(s_1,s_2) = \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi} c_1(\omega_1,\omega_2|s_1,s_2)\tilde{G}_{\mathrm{FP}}(-\omega_2) \times G_{\mathrm{FP}}(-\omega_1) + \int \frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi} c_0(\omega_1,\omega_2|s_1,s_2) \times \tilde{G}_{\mathrm{FP}}(-\omega_2) G_{\mathrm{FP}}(-\omega_1), \quad (12)$$

where

$$c_1 = \frac{(s_2+k)^{\gamma-1}}{s_2(s_2+k)^{\gamma-1}+iD^{\gamma}\omega_2}\frac{(s_2+k)^{\gamma}-(s_1+k)^{\gamma}}{s_2-s_1} \times \frac{1}{s_1(s_1+k)^{\gamma-1}+iD^{\gamma}\omega_1}$$

and

$$c_0 = \frac{(s_1+k)^{\gamma-1}-(s_2+k)^{\gamma-1}}{s_2-s_1}\frac{1}{s_1(s_1+k)^{\gamma-1}+iD^{\gamma}\omega_1}.$$

The expression can be used to measure the response to a step potential applied at t_1 by choosing $\mathbf{G}_{\mathrm{FP}} \neq \tilde{\mathbf{G}}_{\mathrm{FP}}$. For the aging correlation function applied to the harmonic oscillator below, $\mathbf{G}_{\mathrm{FP}}=\tilde{\mathbf{G}}_{\mathrm{FP}}$. If we set $k=0$, we recover two-time anomalous diffusion, which coincides with expressions previously derived for translationally invariant systems and harmonic oscillators.[22,23]

A few comments about Eq. (12) are necessary.[24] Consider the evolution during the second time period (t_1, t_1+t_2) for fixed t_1. The propagator corresponds to a random hopping process, where most hops are fast with the occasional long lived trap. The longer the particle hops, the more likely it will find a long lived cage. The expression for c_0 represents the probability that a particle is still in the cage that it found during the first time interval, which results in c_0 being independent of ω_2, while c_1 corresponds to all processes that made at least one hop during the second time interval. If t_1 is short $(s_1 \to \infty)$, the c_0 term goes to zero because the particle did not find a long lived trap. If t_1 is long, but t_2 is short $(s_1 \to 0,\ s_2 \to \infty)$ the c_0 term dominates since the particle found a long lived trap and has not hopped out of it during the second time interval. Eventually, the particle will hop out of the trap found during the first time interval and resume its random walk so the c_0 term decays and the c_1 term dominates for $t_2 > k^{-1}$. The truncation of the c_0 term is the major contrast between interrupted and uninterrupted fractional diffusions.

APPLICATION TO THE HARMONIC OSCILLATOR

As a simple application of Eq. (12), consider fractional diffusion in a one-dimensional harmonic well with unit diffusion constant, viscosity, and force constant. The resulting Green function for normal diffusion is

$$G_{\mathrm{FP}}(x,x',t) = \tilde{G}_{\mathrm{FP}}(x,x',t) = \frac{1}{\sqrt{2\pi(1-e^{-2t})}}\exp\left[\frac{-(x-x'e^{-t})^2}{2(1-e^{-2t})}\right]. \quad (13)$$

For simplicity, the initial condition is $(1/\sqrt{2\pi})e^{-x^2/2}$, and the system is always in macroscopic equilibrium, but not in microscopic equilibrium with respect to the distribution of activation barriers. We are concerned with the correlation function $C(t_2|t_1)=\langle x(t_1+t_2)x(t_1)\rangle$. Setting $D^{\gamma}=1$ (i.e., time and space are unitless) and integrating over ω_i gives the aging correlation function,

$$C(s_2|s_1) = \frac{(s_2+k)^{\gamma-1}}{s_2(s_2+k)^{\gamma-1}+1}\frac{(s_2+k)^{\gamma}-(s_1+k)^{\gamma}}{s_2-s_1}\frac{1}{s_1(s_1+k)^{\gamma-1}} + \frac{(s_1+k)^{\gamma-1}-(s_2+k)^{\gamma-1}}{s_2-s_1}\frac{1}{s_1(s_1+k)^{\gamma-1}}, \quad (14)$$

which recovers the FFPE in the limit of $h \to 0$. Setting $k=0$ recovers the FFPE result.[23] This correlation function is rigorously unity at $t_2=0$. In the limit of no aging, $t_1 \to 0$, one recovers an interrupted version of the correlation function,

$$\lim_{s_1\to\infty} s_1 C(s_2|s_1) = \frac{1}{s_2+(k+s_2)^{1-\gamma}}. \quad (15)$$

Another important feature is the existence of a stationary correlation function in the limit of $t_1 \to \infty$,

$$\lim_{s_1\to 0} s_1 C(s_2|s_1) = \frac{k^{1-\gamma}}{s_2}\left[k^{\gamma-1}-(k+s_2)^{\gamma-1} + \frac{(k+s_2)^{\gamma}(k^{\gamma}-(k+s_2)^{\gamma})}{k+s_2+s_2(k+s_2)^{\gamma}}\right]. \quad (16)$$

As a specific example we choose $\gamma=1/2$ and $k=1/1000$. The solutions for several aging times $t_1=0,1,10,100,\infty$ are plotted in Fig. 2. If the system has not aged, $t_1=0$, the correlation function demonstrates relaxation on many timescales before approaching exponential behavior at the interruption time $k^{-1}=1000$. If the system is allowed to age for a short time,

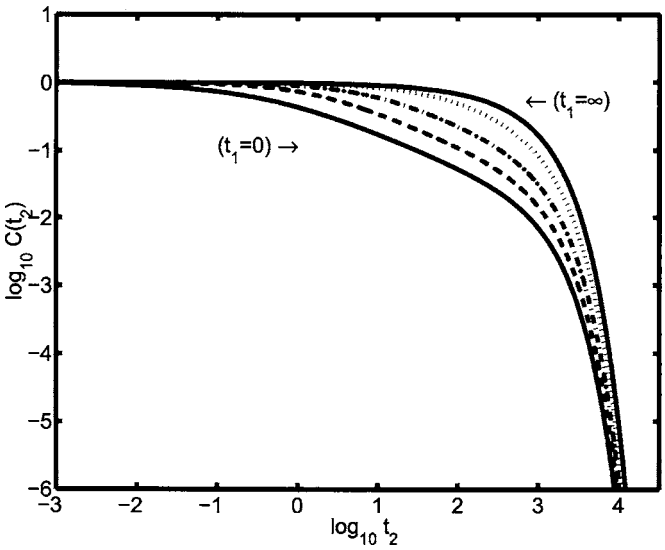

FIG. 2. The correlation function for the interrupted fractional diffusion with $\gamma=1/2$, $k=1/1000$ in a harmonic oscillator with induction times (from bottom to top), $t_1=0,1,10,100,\infty$. The $t_1=0$ solution shows approximately power-law behavior for $1<t_2<1000$, before switching over to exponential behavior in the long time limit. This is in contrast to the nearly exponential behavior (on a log scale) shown by the longer induction times. Aging removes many of the features that one tries to incorporate in the model through a complicated waiting time.

the correlation function remains approximately equal to unity for a short period of time before the onset of the distributed relaxation. The long time exponential behavior is not delayed so one observes a smaller region of multiple time scale relaxation. In the long time limit, $t_1 \to \infty$, the multiple time scale relaxation is absent, and only approximate exponential relaxation is observed. The FDT becomes valid on these longer time scales, and we recover the traditional exponential relaxation associated with the limit.

EXPERIMENTAL MEASUREMENTS

The validity of the FDT at low frequencies and related aging effects must be explored in any experimental system modeled by waiting time processes. If the time of the trapping process cannot be determined, the aged correlation function in Eq. (16) should be used. The longer the first time interval, the longer the c_0 term dominates the correlation function, and the particle does not move. The result is an essential system arrest for periods of time shorter than the characteristic relaxation time determined by the cutoff k^{-1}. This construction of interrupted fractional diffusion makes it apparent that certain quantities are not affected by aging, such as the ordering of events. If the particle is at position $x=0$ at time t_1 and we measure the probability of reaching position $x=1$ before $x=-1$, this probability will not depend on t_1. These invariant measurements should be examined in single molecule experiments where interrupted fractional diffusion appears to fit the correlation function. The measurements may differentiate the simple fractional diffusion memory from a more complex memory effect, such as those expected for the end-to-end distribution of a polymer chain.[25]

Interrupted aging processes are more realistic scenarios for modeling biological systems than the infinite aging of fractional diffusion. Although the stretched exponential approximation to the short time behavior of the nonaged FFPE and a long time power-law have been fit to the correlation functions of several different measurements on single room temperature biomolecules, the systems do not exhibit long time aging processes and appear to achieve equilibrium in a finite amount of time.[16] Similarly, the interruption of aging has been observed in quantum dot blinking statistics and in the aging relaxation of glassy systems, and may prove to be the rule rather than the exception.[15]

ACKNOWLEDGMENTS

This research is supported by the NSF Career Award (No. Che-0093210), Camille Dreyfus Teacher-Scholar Award, and Petroleum Research Fund.

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