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Strong correlations between fluctuations and response in aging transport

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The correlations between the response of a random walker to an external driving field F switched on at time t_a, with the particle's fluctuations in the aging period $(0,t_a)$ are investigated. Using the continuous time random walk and the quenched trap model, it is shown that these correlations remain finite even in the asymptotic limit $t_a \to \infty$. Linear response theory gives a relation between the correlations, the fractional diffusion coefficient, and the field F, thus generalizing the Einstein relation. In systems which exhibit aging, fluctuations in the aging period can be used to statistically predict the nonidentical response of particles to an external field.

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The continuous time random walk (CTRW) [1,2] and the quenched trap model [2–4] are models of anomalous diffusion used to describe a wide variety of physical systems. The CTRW model was suggested in the 1970s by Scher and Montroll [5] to model anomalous non-Gaussian transport of electrons in disordered systems, while Bouchaud [6] used the trap model to describe the phenomenology of glassy dynamics, in particular aging. Consider a random walk process starting at time $t=0$. In the time interval $(0,t_a)$, called the aging period, particles undergo unbiased diffusion, then at time t_a an external field F is switched on which breaks the symmetry of the system, causing a net drift. When the mean displacement of the particles, namely, the averaged response of the particles to the external driving force, depends on t_a even when $t_a \to \infty$, the transport is said to exhibit aging (see discussion below). Aging in the CTRW and the trap model is well investigated [4,7–11]. The aging in these models is related to the power law distribution of waiting or trapping times of the particle, in such a way that the average waiting time is infinite (see details below). When the average waiting time is infinite, the characteristic time scale of the dynamics diverges, and memory effects become very important even when observation times becomes long. Power law trapping times is a wide spread mechanism of aging, for example, in the aging behavior of blinking quantum dots [12,13], where power law waiting times in dark and bright states are observed.

Here we investigate a new aspect of aging. Within the framework of the CTRW and the quenched trap model, we show that the response of the particle to a bias turned on at time t_a is strongly correlated with the fluctuations of the particle in the aging period. Thus in principle knowledge of the fluctuations of the particle in the aging period can be used to statistically predict its response to an external field. Such correlations vanish in the case of nonaging transport, at least asymptotically. Among other things we derive a scaling relation describing these correlations based on linear response theory. As we argue at the end of the paper, similar strong correlations might be found in other models and systems which exhibit aging.

Model 1. We consider the well-known one-dimensional CTRW on a lattice [1,2,14,15]. The lattice spacing is a and the jumps are to nearest neighbors only. Waiting times between jump events are independent identically distributed random variables with a common probability density function (PDF) $\psi(\tau)$. After waiting, the particle has a probability $1/2+h/2$ or $1/2-h/2$ to jump to the right or left, respectively. In the aging period $(0,t_a)$ $h=0$ and the particles follow an unbiased motion, while in the response period $t_a<t$ the bias is $0<h<1$. The total measurement time is $t=t_a+t_r$, where t_r is called the response time. To define the response one has to define the field which is responsible for the bias h. For example, if the particles are coupled to a thermal heat bath with temperature T, and driven by a uniform force field F, standard detailed balance conditions give $h=aF/2k_bT$, when $h \ll 1$ [4,14]. We consider later the generic case

$$\psi(\tau) \sim \frac{A\tau^{-(1+\alpha)}}{|\Gamma(-\alpha)|}, \tag{1}$$

when $\tau \to \infty$ and $0<\alpha<1$, $A>0$. Specific values of α for a wide range of physical systems and models are given in [1,2]. For example, for the annealed version of the trap model $\alpha=T/T_g<1$ [7]. For $0<\alpha<1$, the average waiting time is infinite.

The position of the particle at time t_a+t_r is $X=X_a+X_r$, where $X_a(X_r)$ is the displacement in the aging (response) periods. More specifically, $X_a=\sum_{i=1}^{n_a}\Delta x_i^{(a)}$, $X_r=\sum_{i=1}^{n_r}\Delta x_i^{(r)}$, where $\Delta x_i^{(a)}$ and $\Delta x_i^{(r)}$ are the random jump lengths (of length a) in the aging $(0,t_a)$ and response (t_a,t_a+t_r) periods, respectively, while n_a and n_r are the random number of jumps in the aging and the response periods.

We investigate the correlation function $\langle (X_a)^2 X_r\rangle$, which is a measure for the correlation between the fluctuations in the aging period $(X_a)^2$ and the response to the driving force switched on at time t_a, X_r. We define a dimensionless fluctuation-response (F_{R}) parameter

$$F_{\mathrm{R}}(t_a,t_r) = \frac{\langle (X_a)^2 X_r\rangle}{\langle (X_a)^2\rangle\langle X_r\rangle} - 1, \tag{2}$$

which is equal to zero when the correlations vanish. For the CTRW under investigation one can show that $\langle (X_a)^2 X_r\rangle = ha^3\langle n_a n_r\rangle$ and

$$F_{\mathrm{R}}(t_a,t_r) = \frac{\langle n_a n_r\rangle}{\langle n_a\rangle\langle n_r\rangle} - 1. \quad (3)$$

Thus $\langle n_a n_r\rangle$, the correlations of the number of steps in the aging period with the number of steps in the response period, gives a measure for the correlations between the fluctuations in the displacement in the aging period and the response to the bias.

Let $P_{t_a,t_r}(n_a,n_r)$ be the probability of making n_a jumps in the aging period and n_r jumps in the response period. Knowledge of this function is needed for the calculation of the F_{R} parameter and other high order correlation functions [16], which we shall discuss in a future publication [17]. The paths with $n_a(n_r)$ jump events in the aging period (response period) clearly satisfy $t_{n_a} < t_a < t_{n_a+1}$ and $(t_{n_a+n_r} < t_r + t_a < t_{n_a+n_r+1})$, respectively, where the subscript n in t_n is for the jump number. Hence

$$P_{t_a,t_r}(n_a,n_r) = \langle I(t_{n_a} < t_a < t_{n_a+1}) I(t_{n_a+n_r} < t_r + t_a < t_{n_a+n_r+1})\rangle, \quad (4)$$

where $I(x)=1$ if the event in the parenthesis is true, otherwise it is zero. We define the double Laplace transform $t_a \to u$ and $t_r \to s$ of $P_{t_a,t_r}(n_a,n_r)$, $P_{u,s}(n_a,n_r) = \int_0^\infty dt_r e^{-st_r}\int_0^\infty dt_a e^{-ut_a} P_{t_a,t_r}(n_a,n_r)$. Following the calculation in [17], using the renewal property of the CTRW,

$$P_{u,s}(n_a,n_r=0) = \frac{\hat{\psi}^{n_a}(u)}{s}\left[\frac{1-\hat{\psi}(u)}{u} - \frac{\hat{\psi}(s)-\hat{\psi}(u)}{u-s}\right], \quad (5)$$

while for $n_r \geqslant 1$,

$$P_{u,s}(n_a,n_r) = \frac{\hat{\psi}^{n_a}(u)\hat{\psi}^{n_r-1}(s)}{s(u-s)}[1-\hat{\psi}(s)][\hat{\psi}(s)-\hat{\psi}(u)]. \quad (6)$$

In Eqs. (5) and (6) $\hat{\psi}(u)$ and $\hat{\psi}(s)$ are Laplace transforms of the waiting time PDF. Note that Eqs. (5) and (6) give the proper normalization since $\Sigma_{n_a=0}^\infty \Sigma_{n_r=0}^\infty P_{u,s}(n_a,n_r) = 1/(us)$. Using Eq. (6) and $\langle n_a n_r\rangle_{u,s} = \Sigma_{n_a=0}^\infty \Sigma_{n_r=0}^\infty n_a n_r P_{u,s}(n_a,n_r)$ we find

$$\langle n_a n_r\rangle_{u,s} = \frac{[\hat{\psi}(s)-\hat{\psi}(u)]\hat{\psi}(u)}{s(u-s)[1-\hat{\psi}(u)]^2[1-\hat{\psi}(s)]}, \quad (7)$$

and the averages [13] $\langle n_a\rangle_{u,s} = \hat{\psi}(u)/\{us[1-\hat{\psi}(u)]\}$,

$$\langle n_r\rangle_{u,s} = \frac{\hat{\psi}(s)-\hat{\psi}(u)}{s(u-s)[1-\hat{\psi}(u)][1-\hat{\psi}(s)]}. \quad (8)$$

In principle, once the double Laplace inversion of Eqs. (7) and (8) is made, we can calculate the F_{R} parameter.

If the dynamics is Markovian, namely, the waiting time PDF is exponential $\psi(t) = R\exp(-Rt)$,

$$\langle n_a n_r\rangle = \langle n_a\rangle\langle n_r\rangle = Rt_a Rt_r, \quad (9)$$

and $F_{\mathrm{R}}(t_a,t_r)=0$. For any non-Markovian process with a nonexponential waiting time PDF, the F_{R} parameter is generally not equal to zero.

If the average waiting time $\langle\tau\rangle = \int_0^\infty \tau\psi(\tau)d\tau$ is finite and in the limit $t_a\to\infty$ the fluctuation-response parameter Eq. (2) satisfies

$$\lim_{t_a\to\infty} F_{\mathrm{R}}(t_a,t_r) = 0, \quad (10)$$

and the correlations are lost in this limit. To see this use Eq. (7) in the $u\to 0$ limit, the small u expansion $\hat{\psi}(u) \sim 1 - u\langle\tau\rangle$ to find $\langle(X_a)^2 X_r\rangle \sim ha^3 \frac{1}{u^2\langle\tau\rangle}\frac{1}{s^2\langle\tau\rangle}$. Interestingly, this result is valid for any $s>0$, namely, both for short and long response times t_r. Hence when $t_a\to\infty$ we find $\langle(X_a)^2 X_r\rangle \sim ha^3 \frac{t_a}{\langle\tau\rangle}\frac{t_r}{\langle\tau\rangle}$ and similar calculations for $\langle(X_a)^2\rangle$, and $\langle X_r\rangle$ complete the proof of Eq. (10). Thus, if the aging period is much larger than the *finite* time between jumps the response is not correlated with the fluctuations, since the particles had enough time to equilibrate.

A very different behavior is found in the common situation [1,2] where the average waiting time is infinite, namely, when $0<\alpha<1$ in Eq. (1). Then using Eq. (7), in the limit of small s and u,

$$\langle(X_a)^2 X_r\rangle \sim ha^3 \frac{1}{A^2}\frac{u^\alpha - s^\alpha}{(u-s)u^{2\alpha}s^{1+\alpha}}, \quad (11)$$

where the expansion $\hat{\psi}(u) \sim 1 - Au^\alpha$ was used. Skipping the technical details, we analytically invert the double Laplace transform in Eq. (11) to the double time domain and find

$$\langle(X_a)^2 X_r\rangle \sim ha^3 \frac{t_r^{2\alpha}}{A^2} g\left(\frac{t_a}{t_r}\right), \quad (12)$$

which is valid in the limit of long times t_a and t_r. The scaling function in Eq. (12) is a hypergeometric function

$$g(x) = \frac{x^\alpha {}_2F_1[1,-\alpha;1+\alpha;-x]}{\Gamma^2(1+\alpha)} - \frac{x^{2\alpha}}{\Gamma(1+2\alpha)}. \quad (13)$$

Equation (12) is a main result of this paper, since it shows that even in the long aging time limit a nontrivial correlation between fluctuation and response exists. The hypergeometric function in Eq. (13) is tabulated in MATHEMATICA, hence the solution is not a formal expression. Using $\langle(X_a)^2\rangle \sim a^2 \frac{t_a^\alpha}{A\Gamma(1+\alpha)}$, and the aging response of the model [14] $\langle X_r\rangle \sim ha\frac{(t_a+t_r)^\alpha - t_a^\alpha}{A\Gamma(1+\alpha)}$, the dimensionless fluctuation-response parameter is

$$F_{\mathrm{R}}(x) = \frac{{}_2F_1(1,-\alpha;1+\alpha,-x) - x^\alpha \dfrac{\Gamma^2(1+\alpha)}{\Gamma(1+2\alpha)}}{(1+x)^\alpha - x^\alpha} - 1, \quad (14)$$

where $x = t_a/t_r$. If $\alpha=1$ we have $F_{\mathrm{R}}(x)=0$ indicating that the nontrivial correlations are found in the limit of long times, only for anomalous processes with $\alpha<1$. Equation (14) is valid in the limit $t_a\to\infty$ and $t_r\to\infty$, their ratio x remaining finite.

Comparison between simulations of the CTRW process and Eq. (14) for $\alpha=1/2$ and $\alpha=3/4$ is made in Fig. 1. The figure illustrates that the correlations between fluctuations and response becomes larger as $x=t_a/t_r$ is increased. This is the expected behavior, the larger the aging time t_a compared

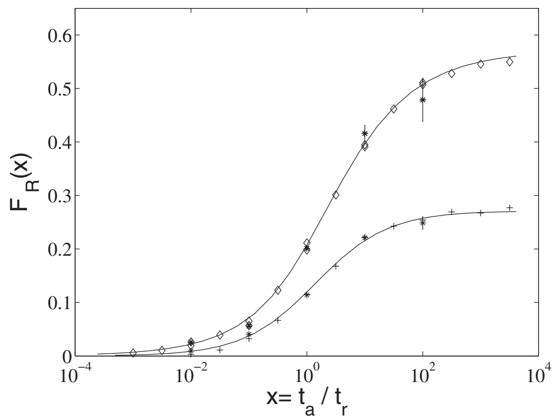

FIG. 1. The fluctuation-response parameter $F_{\mathrm{R}}(x)$ as a function of $x=t_a/t_r$. Simulations and theory (solid curve) agree well without fitting. We use $\alpha=3/4$ (crosses) and $\alpha=1/2$ (diamonds) and see that the correlations are stronger when $\alpha=1/2$. Simulations of a modified CTRW with random bias (stars with error bars) are in good agreement with the CTRW results.

with the response time t_r, the stronger the correlation, since if $t_r \gg t_a$ the particle already "forgot" its behavior in the aging period. Figure 1 also demonstrates that as α is decreased the correlations becomes stronger. In the simulation $t_a=3\times 10^6$, $\psi(\tau)=\alpha\tau^{-(1+\alpha)}$, for $\tau>1$, otherwise it is zero.

Using Eq. (14), we find in the limit $x=t_a/t_r \ll 1$

$$F_{\mathrm{R}}(x) \sim \left(1-\frac{\Gamma^2(1+\alpha)}{\Gamma(1+2\alpha)}\right)x^\alpha + O(x), \tag{15}$$

namely, weak correlations between fluctuations and response. In the opposite limit, the aging regime of $x \gg 1$, the correlations are stronger, and we find

$$F_{\mathrm{R}}(x) \sim \left[\frac{\alpha|\Gamma(\alpha)|^2}{\Gamma(2\alpha)}-1\right]-\frac{1}{1+\alpha}\frac{1}{x^\alpha}, \quad x\to\infty. \tag{16}$$

The leading term gives the nontrivial behavior of the fluctuation-response parameter when $t_a/t_r\to\infty$. We find the bounds $0\le \lim_{x\to\infty}F_{\mathrm{R}}(x)\le 1$, where the lower bound with zero correlations corresponds to $\alpha\to 1$ and the upper bound of strong correlations is found when $\alpha\to 0$.

Applying linear response theory to Eq. (12), yields the connection between the correlation function and physically observable parameters. In this same limit, aging Einstein relations between the ensemble average response and the fluctuations in the absence of the field are valid [4,14]. We find using $h=aF/2k_bT\to 0$,

$$\langle (X_a)^2 X_r\rangle \sim \frac{2FD_\alpha^2 t_r^{2\alpha}}{k_bT} g\left(\frac{t_a}{t_r}\right), \tag{17}$$

where $D_\alpha=a^2/2A$ is the fractional diffusion coefficient, which according to its definition is $\langle X^2\rangle \sim 2D_\alpha t^\alpha/\Gamma(1+\alpha)$ when $F=0$ [1,18]. Equation (17) is important since it shows that the transport coefficient D_α and the exponent α, *describing the fluctuations in the absence of the external driving field*, are the only system parameters needed for the determination of the correlation between fluctuations and the response.

We investigated the F_{R} parameter also for a CTRW with random barriers. Here, the probability of jumping right from site i is $(1+h_i)/2$, where $\{h_i\}$ are independent identically distributed random variables. In our simulations we considered $h_i=h_r$ or $h_i=-h_r$ with probability $1/2$. The sequence $\{h_i\}$ is fixed at the beginning of the simulation. In addition, a bias h is turned on at time t_a, so for time $t>t_a$ the probability of jumping right from site i is $(1+h_i+h)/2$. In Fig. 1 we show that the agreement between this modified CTRW and the standard CTRW with $h_i=0$ is good, at least for the parameters under investigation (we considered $h=h_r=0.1$). The local random bias h_i does not alter the behavior of the F_{R} parameter, since the long tailed waiting times are dominating the aging behavior.

Model 2. As shown by Feigelman and Vinokur [19], transport in disordered systems with quenched disorder may exhibit aging effects. Hence it is natural to check if fluctuations and response are correlated for models of quenched disorder, and if so how do they compare with those we obtained analytically in the annealed CTRW model? In particular do quenched models also exhibit a transition between an aging regime with strong correlations to a regime with vanishing correlations when $t_a\to\infty$? For that aim we consider the quenched trap model on a one-dimensional lattice [3,4]. Each lattice site i has a fixed random energy $E_i>0$, which is the energy barrier the particle has to cross in order to jump from i to $i+1$ or $i-1$. The energy barriers are all independent identically distributed random variables with a common PDF $\rho(E)=(T_g)^{-1}e^{-E/T_g}$. The PDF of escape times from site i is exponential with a mean escape time $\tau_i=\exp(E_i/T)$. Notice that according to this Arrhenius law small fluctuations in the energy may lead to exponentially large fluctuations in the waiting time in site i. After waiting in the trap for a random time the particle has a probability $1/2$ of jumping right or left if the system is not biased. It has a probability $(1\pm h)/2$ of jumping left or right when the bias is not zero. In simulations one lets the system evolve without bias in the aging period $(0,t_a)$ and then a bias is switched on.

It is well known that the model exhibits aging behaviors when $T<T_g$ [6]. Bertin and Bouchaud showed that the aging exhibits linear and nonlinear types of response, depending on the magnitude of h [4]. Here we consider only the linear response regime of $h\to 0$. The trap model is not an exactly solvable model since the effect of the quenched disorder is to induce nonindependent waiting times in the random walk. Hence we investigate the trap model using numerical simulations.

Simulation results for the F_{R} parameter are shown in Fig. 2. We choose model parameters known to exhibit aging [4] $t_a=10^6$, $h=0.008$, and vary t_r. In the high temperature ergodic phase $T/T_g=5/2$, the F_{R} parameter is practically zero with small deviations due to the finite time of the simulation. For $T/T_g=1/2$, namely, in the aging phase, we see strong correlations between the fluctuations and the response especially when $t_r<t_a$. We also plot the $F_{\mathrm{R}}(x)$ parameter for the CTRW model using the exponent $\alpha=2T/T_g/(1+T/T_g)$ $=2/3$ (α is the exponent describing the averaged response

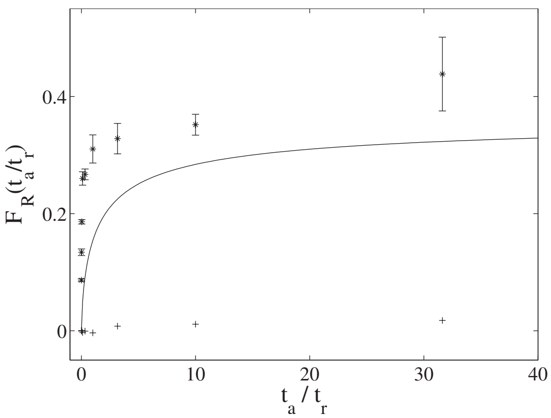

FIG. 2. The F_{R} parameter for the quenched trap model. The plus symbols are for the high temperature phase $T=5T_g/2$, namely, $T>T_g$, which does not exhibit correlations $F_{\mathrm{R}} \simeq 0$, while the star symbols are for the aging phase $T=T_g/2$, which exhibits nontrivial correlations. The solid curve is the CTRW theory.

function [4]). Figure 2 clearly demonstrates that the F_{R} parameter in the CTRW theory is smaller than the corresponding F_{R} parameter of the quenched trap model. In the quenched trap model, unlike the CTRW process, the random walk is correlated in the sense that a particle once returning to a specific trap will recall its waiting time for that trap, hence the F_{R} parameter for the quenched model is larger than the one found for the CTRW.

There are many examples of systems where the correlations discussed in this manuscript may become important, we mention the recent single particle experiments of microbeads diffusing in actin networks [20], which exhibits power law waiting times and anomalous diffusion, very much reminiscent of the trap and CTRW models. However it is important to realize that correlations between fluctuations and transport are not expected to be limited only for systems whose dynamics is characterized with power law waiting time distribution. Consider, for example, the measurement of single LacI repressor protein on DNA [21], where a wide distribution of diffusion coefficients of the proteins was found. This distribution is likely due to the random DNA sequence the single protein explores. Since the single molecules have widely distributed diffusion constants, their response to an external field is likely correlated with their history of diffusion, e.g., particles with small (or large) diffusion, constants have a weak (strong) response, respectively. Further work on the correlations between fluctuations and response, in other models of transport, is left for future work.

To conclude, for any non-Markovian CTRW, the correlations between the fluctuations in the aging period and the response are finite if the aging period is finite. Both for the CTRW with $\alpha<1$ and for the quenched trap model with $T<T_g$, a nontrivial F_{R} parameter was found when the aging period is asymptotically long. These correlations are found when the models exhibit aging and anomalous diffusion. I suspect that other models and systems with aging behavior will exhibit similar nontrivial correlations since aging behavior is related to long memory effect. The wide applications of the CTRW theory, the trap model, and linear response theory make the main equations (14) and (17) of this manuscript relevant to many systems and testable in experiments.

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[1] R. Metzler and J. Klafter, Phys. Rep. **339**, 1 (2000).
[2] J. P. Bouchaud and A. Georges, Phys. Rep. **195**, 127 (1990).
[3] B. Rinn, P. Maass, and J. P. Bouchaud, Phys. Rev. Lett. **84**, 5403 (2000).
[4] E. M. Bertin and J. P. Bouchaud, Phys. Rev. E **67**, 065105(R) (2003).
[5] H. Scher and E. Montroll, Phys. Rev. B **12**, 2455 (1975).
[6] J. P. Bouchaud, J. Phys. I **2**, 1705 (1992).
[7] C. Monthus and J. P. Bouchaud, J. Phys. A **29**, 3847 (1996).
[8] E. Barkai, Phys. Rev. Lett. **90**, 104101 (2003).
[9] P. Allegrini, G. Aquino, P. Grigolini, L. Palatella, and A. Rosa, Phys. Rev. E **68**, 056123 (2003).
[10] I. M. Sokolov, A. Blumen, and J. Klafter, Europhys. Lett. **56**, 175 (2001).
[11] I. M. Sokolov and J. Klafter, Phys. Rev. Lett. **97**, 140602 (2006).
[12] X. Brokmann, J. P. Hermier, G. Messin, P. Desbiolles, J. P. Bouchaud, and M. Dahan, Phys. Rev. Lett. **90**, 120601 (2003).
[13] G. Margolin and E. Barkai, J. Chem. Phys. **121**, 1566 (2004).
[14] E. Barkai and Y. C. Cheng, J. Chem. Phys. **118**, 6167 (2003).
[15] O. Flomenbom and J. Klafter, Phys. Rev. Lett. **95**, 098105 (2005).
[16] F. Sanda and S. Mukamel, Phys. Rev. E **72**, 031108 (2005).
[17] E. Barkai and I. M. Sokolov, e-print arXiv:cond-mat/0705.2857v1, J. Stat. Mech.: Theory Exp. (to be published).
[18] E. Barkai, R. Metzler, and J. Klafter, Phys. Rev. E **61**, 132 (2000).
[19] M. V. Feigelman and V. Vinokur, J. Phys. (France) **49**, 1731 (1988).
[20] I. Y. Wong, M. L. Gardel, D. R. Reichman, E. R. Weeks, M. T. Valentine, A. R. Bausch, and D. A. Weitz, Phys. Rev. Lett. **92**, 178101 (2004).
[21] Y. M. Wang, R. H. Austin, and E. C. Cox, Phys. Rev. Lett. **97**, 048302 (2006).