Rapid Communications

Aging is a log-Poisson process, not a renewal process

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Aging is a ubiquitous relaxation dynamic in disordered materials. It ensues after a rapid quench from an equilibrium "fluid" state into a nonequilibrium, history-dependent jammed state. We propose a physically motivated description that contrasts sharply with a continuous-time random walk (CTRW) with broadly distributed trapping times commonly used to fit aging data. A renewal process such as CTRW proves irreconcilable with the log-Poisson statistic exhibited, for example, by jammed colloids as well as by disordered magnets. A log-Poisson process is characteristic of the intermittent and decelerating dynamics of jammed matter usually activated by record-breaking fluctuations ("quakes"). We show that such a record dynamics provides a universal model for aging, physically grounded in generic features of free-energy landscapes of disordered systems.

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During a quench, when temperature is dropped in a complex fluid or disordered magnet [1–6], density is rammed up in a colloidal system [7–10], or strain is intensified on a granular pile [11,12], amorphous materials begin to jam up such that relaxational timescales exceed experimental capabilities. In the ensuing aging process [13], observables retain a memory of the time t since the quench at $t = 0$, signifying the breaking of time-translational invariance and the nonequilibrium nature of the state. Similar phenomena have been observed also in protein dynamics [14], friction [15], or financial time series [16]. In a jammed disordered system, structural relaxation towards some far-removed equilibrium state proceeds by increasingly rare, activated events [10,17–20] (see Fig. 1). Furthermore, two-time measures of macroscopic observables, taken for some time $\Delta t = t - t_w$ starting at t_w, scale most reasonably as a function of the single variable t/t_w. (In contrast, an equilibrium process would remain time-translational invariant, depending on Δt alone.) For example, experimental data for the thermoremanent magnetization of a glassy magnet [3,18], or for the persistence and the mean-square displacement of colloidal particles [21] as well as their entire displacement probability density (van Hove) function [22], collapse when plotted as function of t/t_w. The wide variety of metastable systems exhibiting this phenomenology suggests its universality [23,24] and call for a unified coarse-grained description, independent of microscopic detail [25–27].

Observations such as this have become the basis for models that treat aging as a renewal process [27–31]. In the trap model [28], for instance, the entire system performs a random walk through a configuration space filled with traps possessing a broad (power-law) distribution of escape times. Thus, the older the system, encountering ever "deeper" traps will become more likely, and the deepest trap encountered dominates all previous timescales. However, once escaped, no memory of previous events informs future events. As noted in Ref. [32], applying this type of description to aging systems violates the system size scaling and self-averaging properties of macroscopic variables, which are universally observed in nature. The problem is avoided in continuous-time random walk models (CTRW), where each particle in a colloid, say, is now endowed with a power-law distribution of times between displacements. Over time, particles perform intermittent jumps, interpreted as them breaking out of their cages formed by surrounding particles, as particle-tracking observations tend to justify [8,24]. In this Rapid Communication we show, however, that any such renewal process is ruled out as an underlying physical mechanism by demonstrating that the transitions from one metastable state to the next follow a log-Poisson process which originates from the record-sized fluctuations needed to relax the aging system.

By their very nature, renewal processes seem antithetical to aging because each renewal resets the history. However, it is the broad distribution of escape times that ensnares an increasing number of particles for indefinitely long times after the quench and overall activity decelerates. To be specific, the probability for observing m such cage-breaking events after the waiting time t_w since the quench in a single CTRW is given by

$$p_m(t_w) = \int_0^{t_w} d\tau\; p_{m-1}(t_w - \tau)\psi(\tau), \tag{1}$$

initiated with $p_m(0) = \delta_{m,0}$, and a interevent-time distribution

$$\psi(\tau) \sim \tau^{-1-\alpha} \quad (0 < \alpha \leqslant 1). \tag{2}$$

For such α, interevent times, i.e., escape times, have a diverging mean and the rate at which events are observed, $\partial_t\langle m\rangle \sim t_w^{-1+\alpha}$, indeed decelerates, so that the accumulated number of events rises sublinearly [see Fig. 1(a)]. Moreover, when such a process has evolved up to time t_w after the quench, the probability to observe the next (escape) event within a time interval $\Delta t = t - t_w$ exhibits the t/t_w-dependence [33] characteristic of most aging phenomena [27]: As the time needed to escape the typical trap entered at t_w is itself $\propto t_w$, then so is $\Delta t \propto t_w$, which constitutes some fraction of the total escape time. These are powerful features of renewal models that have contributed greatly to justify their widespread application to fit data produced in a wide variety of aging

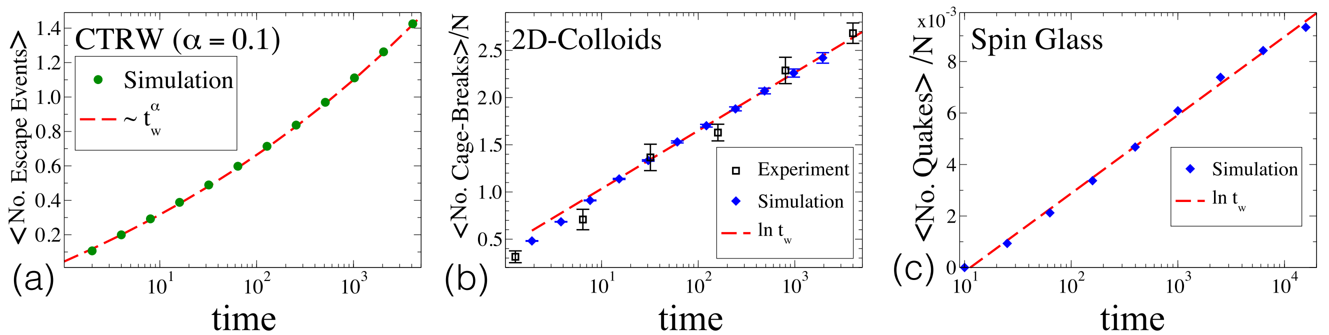

FIG. 1. Accumulated number of relaxation events ("quakes") with time t after a quench during simulations of (a) a continuous-time random walk (CTRW), (b) a jammed 2D colloid, and (c) a 3D Edwards-Anderson spin glass. In (a), we sampled the number of escapes by evolving Eq. (2) for a rather small value of $\alpha = 0.1$, to more closely resemble the physical aging data in (b) and (c). In (b) we measured intermittent cage breaks after a quench, marked by irreversible neighborhood swaps in the molecular-dynamics simulations. Also shown are the data from the colloidal experiment by Yunker *et al.* [10,21]. In (c), we obtained the irreversible barrier crossings in a 3D Edwards-Anderson spin glass. Note that the physical data are growing logarithmically with time, while such a behavior is obtained in the CTRW only for $\alpha \to 0$.

experiments [14,16,24,28,30,34,35]. The exponent α provides a readily available parameter to fit data. However, the *physical* origin of α or $\psi(\tau)$ remains obscure.

In light of its potential benefits, it is most instructive to compare CTRW with a direct measure of the sequence of quake events in realistic aging processes, to assess their statistical properties more fully. This we have undertaken in molecular-dynamics simulations of a two-dimensional (2D) system of bidisperse colloidal particles and a Monte Carlo simulation of three-dimensional (3D) Edwards-Anderson (EA) spin glasses. (Details of these simulations are described in Refs. [22,36].) Each provides a canonical model of aging (and glassy behavior generally) for quite distinct disordered materials [37,38]. In a colloid, disorder is merely structural, arising from the irregular random packing of the particles [37]. In contrast, in a spin glass, dipolar magnets are localized in a lattice of *a priori* fixed but randomly chosen couplings with their neighbors that would frustrate their optimal alignment even in any conceivable—yet dynamically inaccessible—equilibrium arrangement [38]. In both systems we follow the sequence of events after a quench, achieved by either rapidly expanding the colloidal particles to transition from a low-density liquid into a high-density jammed state, or by lowering temperature of the spin glass well below its glass-transition temperature. Sampling over repeated simulations, we have recorded either the cage breakings or the energy barrier crossings (see Fig. 3), which constitute the irreversible events (or jumps) signifying activated relaxation in the respective systems.

In all three cases the rate of events does decelerate as a power law with time. However, in both simulations the rate is essentially hyperbolic, $\partial_t\langle m\rangle \sim t_w^{-1}$, such that the accumulated number of events increases logarithmically with time, as plotted in Fig. 1. This feature of the physical data a renewal process only achieves in the limit of $\alpha \to 0$. That limit is somewhat singular because the distribution in Eq. (2) would become unnormalizable. Arguably, this could be accounted for within numerical accuracy of the data by stipulating logarithmic factors or by simply assuming some small value of α [such as $\alpha = 0.1$, as used in Fig. 1(a)]. Yet, the following will demonstrate that the key discrepancy between a renewal model and the data arises from the actual *sequence* of quake events.

Focusing first on the numerical data, we let t_k denote the kth quake in the time series of measured irreversible events extracted from a trajectory. Figures 2(b) and 2(c) show the statistics of the logarithmic time differences $\Delta\ln = \ln t_{k+1} - \ln t_k = \ln(t_{k+1}/t_k)$, which we treat as identically distributed stochastic variables. To wit, to a good approximation, the statistics is described by an exponential probability density function (PDF) $P_{\Delta\ln}(x) = \exp\{-x/\mu_q\}/\mu_q$, which is shown for a number of different system sizes. The rate of events $1/\mu_q$ increases with system size or simply the number of tracked particles n. That the scaling is indeed linear is shown in the insets, where all data are collapsed by the scaling transformation $\Delta\ln \to \Delta\ln/n$. Thus, the form of the data supports the hypothesis that quake events have a log-Poisson statistic, i.e., the data follow a Poisson distribution whose only parameter—the average—grows *logarithmically* with time, i.e., $\mu_q \propto \ln t$.

The additivity of Poissonian variables now ensures that the number of events is an extensive variable. In CTRW, the number of ongoing independent random walks is a fitting parameter rather than a dynamical consequence of the size of the system at hand. One can nevertheless look at the sequence t_k of renewal events generated by n different random walks. For $n = 1$, the PDF of the time intervals $\tau = t_{k+1} - t_k$ between consecutive events at times t_k and t_{k+1} by definition reproduces the power-law distribution $\psi(\tau)$ in Eq. (2). For $n > 1$, the data from the renewal process retain a power-law distribution, inconsistent with a Poisson process on any timescale. Even if we define a generalized α-Poisson process, based on the observation that the rate of events observed in a renewal process is $\partial_t\langle m\rangle \sim t^{-1+\alpha}$, i.e., $\langle m\rangle(t,t_w) \sim t^\alpha - t_w^\alpha$, the α intervals between events, $t_{k+1}^\alpha - t_k^\alpha$, remain power-law distributed and distinctly non-Poissonian, as demonstrated in Fig. 2(a). Thus, a renewal process must be rejected as a model for aging. With further extensions [41], a nonrenewal CTRW can be designed that evolves many walkers in parallel, one for each future event. Here, the system, after leaving the trap of one walk, immediately enters the already extant trap of another walk, instead of undergoing renewal. While this model provides a log-Poisson statistic for all $0 < \alpha < 1$, any connection to the physical processes we discuss here is tenuous, at best.

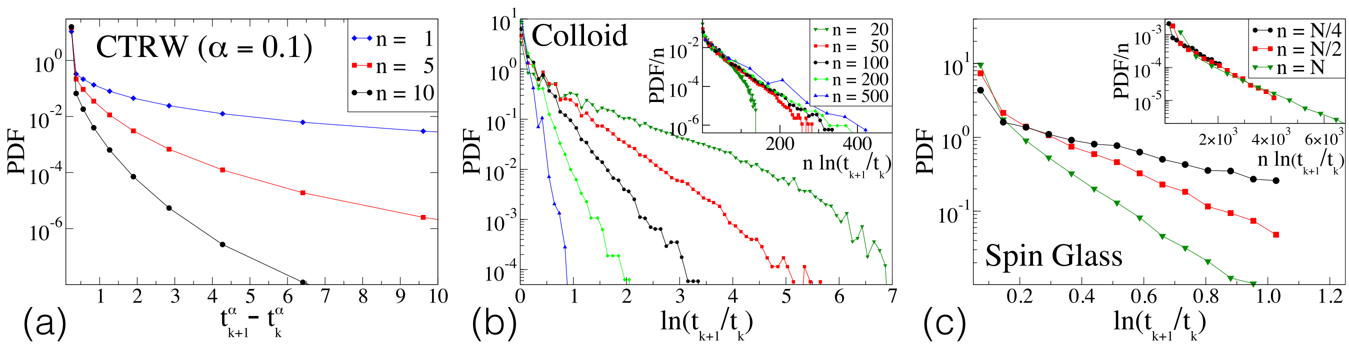

FIG. 2. Poisson statistics of (a) the CTRW in Eqs. (1) and (2) for $\alpha = 0.1$, and for the aging dynamics in simulations of (b) a colloid and (c) a spin glass. Times t_k mark the kth event, i.e., escaping a trap in the CTRW, an irreversible cage break in a colloid, or quakes in a spin glass. The CTRW does not have the exponential form expected for a Poisson process, while both physical aging processes do. Generally, when too many events (i.e., too many degrees of freedom) are blurred together, it hides the local impact of the decelerating activated events and tails weaken. Record dynamics predicts that the rate of events is proportional to the number n of degrees of freedom observed. As the inset for both (b) and (c) shows, the data in the aging simulations indeed collapse when rescaled by n.

In contrast, it is quite fruitful to view aging as a record dynamics (RD) [42–44]. In a statistic of records, a sequence of t independent random numbers drawn from *any* smooth probability density function produces a record-sized number at a rate $\partial_t\langle m\rangle \sim 1/t$. Hence, the number of random events tallied between a time t_w and $t = t_w + \Delta t$ is $\langle m\rangle(t, t_w) \sim \ln(t) - \ln(t_w) \sim f(t/t_w)$. Thus, RD leads to a log-Poisson statistic [44], as found for the physical data above. RD also considers anomalously large events, such as cage breaks in colloids, as being essential to substantially relax the system and having to be viewed as distinct from the Gaussian fluctuations of in-cage rattle. [This distinction is also essential to CTRW, as the discussion before Eq. (2) in Ref. [24] shows]. Yet, such a relaxation must entail a structural change—the physical essence of aging—that makes subsequent relaxation even harder (see Fig. 3). For example, to facilitate a cage break, a certain number of surrounding particles have to "conspire" to move via some rare, random fluctuation [10]. For that event to qualify as an irreversible loss of free energy, the resulting structure must have increased stability, however marginal. A subsequent cage break therefore requires even more particles to conspire. With each fluctuation being exponentially unlikely in the number of particles, cage breaks represent *records* in an independent sequence of random events that "set the clock" for the activated dynamics, resulting in the observed log-Poisson statistics. Then, any two-time observable becomes *subordinate* [18] to this clock: $C(t, t_w) = C[\langle m\rangle(t, t_w)] = C(t/t_w)$. Indeed, much of the experimental colloidal tracking data in the aging regime can be collapsed in this manner [21,45]. In our colloidal simulations this is verified by the mean-square displacement (MSD) of particles between times t_w and $t = t_w + \Delta t$ after a quench at $t_w = 0$ which similarly collapses onto a single function of t/t_w, as shown in Ref. [22] and previously obtained for experiments in Ref. [21]. That function is consistent with a logarithmic growth of MSD with t/t_w, indicative of jumps caused by activations in a decelerating sequence of record events. In fact, the entire van Hove function for particle displacements can be collapsed in this manner [22]. RD furthermore predicts that the rate of events is proportional to the number n of particles observed, as verified by the data. Thus, RD provides an effective model of aging, similarly devoid of microscopic details and hence apt to capture the universality of aging.

Beyond being an effective model, RD allows deep physical insights into the aging dynamics. While ordinary exponential relaxation in the total energy implicates a gradual descent of the material through a smooth (convex) landscape, the temporal and spatial heterogeneity (i.e., intermittency and "dynamic heterogeneity") observed during aging is indicative

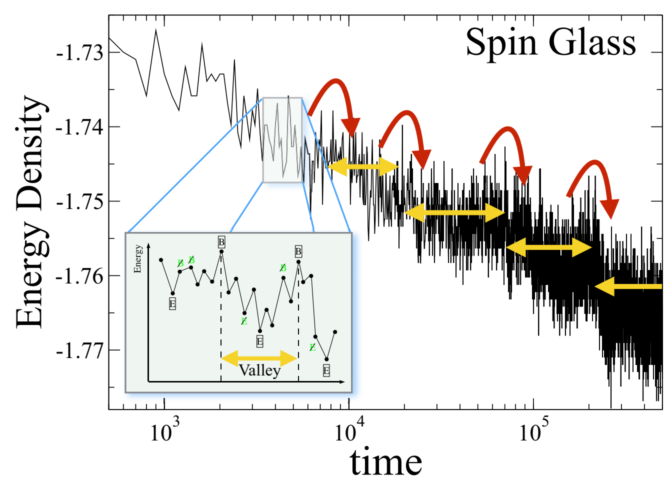

FIG. 3. Energy trace during a single aging process in a 3D EA spin-glass system with time t after a quench. The energy decreases logarithmically with time in a widely fluctuating manner. Unlike in a renewal process, a glass never returns to energy levels visited decades earlier, signifying the gradual but significant structural evolution in the configuration of spins. Energy leaves the systems in intermittent and *irreversible* escape events ("quakes") out of "valleys" (yellow arrows) triggered by a record fluctuation (red arrows). Inset: Tumbling through a complex energy landscape, a time sequence of lowest-energy (E) and highest-barrier (B) records (relative to the most recent "E") is produced [39,40]. Only the highest and lowest records of the "E and "B" are kept to give a strictly alternating sequence "EBEBE…." Then, any "BEB" sequence demarcates entering and escaping a valley. As in-cage rattle within a colloid, each valley represents a local metastable domain in the landscape where the system exhibits quasiequilibrium behavior on timescales shorter than the escape time.

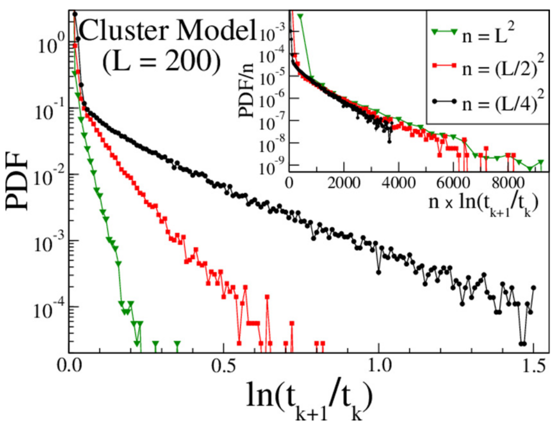

FIG. 4. Log-Poisson statistics for the cluster model [25] on a square lattice of size $L = 200$. Inset: Data collapse when rescaled by n, as in Figs. 2(b) and 2(c) for colloid and spin glass.

of a nonconvex landscape with many local minima trapping the system in a hierarchy of metastable states. However, unlike in a renewal process, leaving such a trap is an irreversible process that has memorable consequences. Free energy (e.g., free volume in the case of a hard-sphere colloid) leaves the system which gets deeper entrenched, calling for a progression of record fluctuations (see Fig. 3).

A phenomenological cluster model of aging has been proposed recently, based on record dynamics [25]. It is an on-lattice model that captures the combined temporal *and* spatial heterogeneity found in a colloidal system: Mobile particles accrete into jammed clusters only to be remobilized in a chance fluctuation (i.e., a quake in free volume) after a time *exponential* in the size h of any cluster which occurs with probability $P(h) \sim e^{-h}$. Following a quench at $t = 0$, clusters form and break up to *irreversibly* distribute their particles to neighboring clusters. The number of clusters reduces by one so that the average size of the remaining clusters (and thus their stability) marginally increases [46]. Their growing size naturally decelerates the dynamics. The algorithm is exceedingly simple and consists of only *two* choices, yet, it readily reproduces experimental data [21]: Particles always completely fill a lattice, one on each site, but each particle either (1) is mobile ($h = 1$), or (2) it is locked in a cluster of size $h > 1$ with adjacent particles. At the time of quench ($t = 0$), all particles are mobile. When picked for an update at time $t > 0$, (1) a mobile particle with $h = 1$ swaps position with a random neighbor and joins its cluster, conversely, (2) a particle in a cluster of size $h > 1$ breaks up the entire cluster with probability $P(h)$. While Ref. [25] already obtained a hyperbolic event rate for the cluster model, consistent with Figs. 1(b) and 1(c), in Fig. 4 we demonstrate its log-Poisson behavior.

In conclusion, our study shows that existing models of aging based on renewal processes are inconsistent with the physical evidence of aging exhibiting jumps or quakes describable as a log-Poisson process. The implications of this finding for other models of aging remain less obvious. Amir *et al.* [11,26] stipulate a convolution of relaxation rates λ with distribution $P(\lambda) \sim \lambda^{-1}$ that implies aging of observables $\sim \log(t/t_w)$, consistent with our description. Earlier theories [47,48], derived from mean-field spin glasses before experiments implicated the importance of intermittency [10,17–20], describe aging merely as a gradual process. Future analysis will reveal its consistency with the evidence in its systemwide averages, but it lacks any notion of *localized* spatiotemporal heterogeneity now considered essential in the understanding of slow relaxation. In contrast, a description of jamming in terms of a random first-order transition (RFOT) [49,50] explains the mechanics of *individual* cage-breaking events in structural glasses in great detail, putting some emphasis on the irreversibility of the event and the structural relaxation it implies. There, the increasing free-energy barriers we stipulated for reaching lower metastable basins in the landscape are explained in terms of an impending entropy crisis: Lower-energy basins are ever harder to find. Yet, we are not aware of any prediction within RFOT about the observed $1/t$ deceleration in the rate of quake events, or any other hallmark of a log-Poisson process. In fact, the need for such a microscopic justification for aging is somewhat antithetical to the rather broad universality found here for both structural as well as quenched glasses, at least for an elementary protocol.

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