Editors' Suggestion

Rejuvenation and Memory Effects in a Structural Glass

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We show numerically that a three-dimensional model for structural glass displays aging, rejuvenation, and memory effects when subjected to a temperature cycle. These effects indicate that the free energy landscape of structural glasses may possess the complex hierarchical structure that characterizes materials such as spin and polymer glasses. We use the theoretical concept of marginal stability to interpret our results, and explain in which physical conditions a complex aging dynamics can emerge in dense supercooled liquids, paving the way for future experimental studies of complex aging dynamics in colloidal and granular glasses.

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The behavior of many disordered materials is dominated by their failure to reach equilibrium, leading to extremely slow relaxations, nonlinear responses, and time-dependent behavior. This aging behavior is observed in a broad variety of condensed-matter systems as microscopically distinct as polymers [1], spin glasses [2], molecular glasses [3,4], colloidal gels [5,6], disordered ferroelectrics [7,8], and crumpled paper sheets [9]. The widespread occurrence of aging phenomena is theoretically understood as a general consequence of frustration leading to a complex free-energy landscape [10,11].

Specific experimental protocols, such as temperature cycles, are used to better characterize the nonequilibrium dynamics of glasses [2,12]. Temperature cycles within the glass phase were first performed in spin glasses, revealing spectacular dynamical effects [13–18]. Aging is reinitialized after a second downward jump in temperature (rejuvenation), but when the first temperature is restored, the system recalls the state reached before that jump (memory). However, when similar protocols are applied to molecular glasses, such as glycerol, no rejuvenation is observed [3], although some memory can be found [19–21]. Both effects were however reported in gelatin gels [22]. Aging is a simple consequence of long relaxation timescales, but rejuvenation and memory effects require a specific, hierarchical organization of the free-energy landscape [23–26]. This is exactly realized in mean field models for spin glasses [27–29], and can directly be confirmed in spin-glass simulations [30–33].

Recently, the mean-field theory for structural glasses predicted the existence of marginally stable glass phases characterized by a hierarchical free energy landscape, with strong similarities with spin glasses [34,35]. Although the existence of a sharp phase transition between normal and marginally stable glass phases remains debated in finite dimensions [36–39], the theory makes crisp predictions regarding the physical conditions where the glassy landscape becomes hierarchical [40–43]. There are numerical evidences that a complex aging dynamics emerges in the hard sphere model [44–46], but simulations of model atomic glasses [47,48] did not find those signatures.

We numerically study the nonequilibrium dynamics of soft repulsive spheres in $d = 3$. This choice is motivated by both theoretical results in the mean-field limit [43] and by a numerical exploration of the complete temperature-density phase diagram to detect the state points where marginal stability can be expected to become physically relevant [49]. By carefully choosing the state points where signs of marginal stability can be observed [49] to perform the present temperature cycles, we successfully observe rejuvenation and memory effects in our model for structural glasses. Our central result is presented in Fig. 1, where we

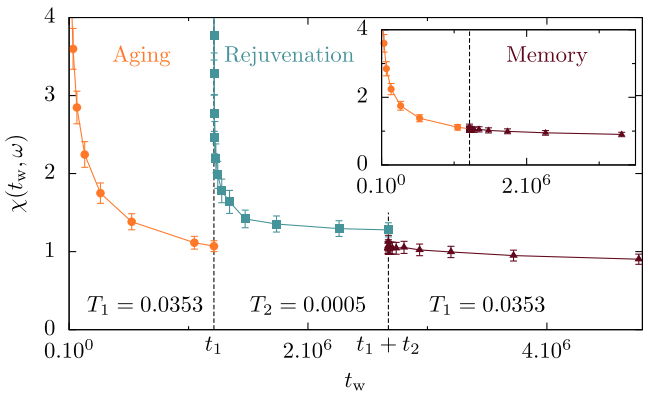

FIG. 1. Aging, rejuvenation, and memory in a structural glass subjected to a temperature cycle. We show the time evolution of $\chi(t_w, \omega = 10^{-5})$ in each step of the cycle, delimited by vertical dashed lines. Aging is observed as the fluid is quenched into the glass phase (circles). The glass rejuvenates as it is cooled further (squares), but retains perfect memory when heated back (triangles). In the inset, the intermediate step is removed to better demonstrate the memory effect.

adopt the same representation as in experiments, showing the evolution of a dynamic susceptibility $\chi(t_w, \omega)$ [Eq. (3)] during the cycle. A high-temperature fluid is rapidly cooled to T_1 in the glass phase. Aging dynamics is signaled by a slowly decreasing χ (circles). The glass is aged for a given time before being cooled to a lower temperature T_2. The glass then rejuvenates, since a strong restart of the aging dynamics takes place at T_2 (squares). When the glass is reheated to T_1 (triangles), it recovers memory of the initial aging (Fig. 1, inset), despite the strong rejuvenation in the intermediate step. We attribute these effects to the hierarchical landscape of structural glasses in a marginally stable phase.

Model and methods.—We study a three-dimensional glass former composed of $N = 3000$ continuously polydisperse particles. Two particles i and j at positions $\mathbf{r}_i$ and $\mathbf{r}_j$ interact via the Weeks-Chandler-Andersen (WCA) potential [50]

$$v(r_{ij}) = 4\epsilon[(\sigma_{ij}/r_{ij})^{12} - (\sigma_{ij}/r_{ij})^{6}] + 1, \qquad (1)$$

only if they are at a distance $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j| < 2^{1/6}\sigma_{ij}$, with a nonadditive interaction rule $\sigma_{ij} = [(\sigma_i + \sigma_j)/2](1 - 0.2|\sigma_i - \sigma_j|)$. The potential and forces are continuous at the physical cutoff distance. For each particle, σ_i is drawn from the normalized distribution $P(\sigma_m \le \sigma \le \sigma_M) \sim 1/\sigma^3$, where $\sigma_m = 0.73$ and $\sigma_M = 1.62$. This model is chosen for its excellent glass-forming ability when simulated either with molecular dynamics, or particle-swap dynamics [51], and represents a canonical model for dense supercooled liquids [52].

The aging dynamics is studied with molecular dynamics (MD). The simulations are performed with a time discretization $dt = 0.003$, within a cubic box of linear size L, using periodic boundary conditions. The temperature is controlled by a Berendsen thermostat with damping parameter $\tau_B = 1$ [53]. We reset the total momentum to zero every 10^6 MD steps. Lengths, times, and energies are expressed in units of $\bar{\sigma} = \int \sigma P(\sigma) d\sigma$, $\sqrt{\epsilon/m\bar{\sigma}^2}$ and ϵ, respectively. The state of the system is determined by temperature T, and packing fraction $\varphi = \pi/(3\sqrt{2}L^3)\sum_i \sigma_i^3$. For this nonadditive polydisperse mixture, the jamming transition occurs near $\varphi_J \sim 0.78$. Here, we focus on a fixed packing fraction $\varphi = 0.85$, and discuss later this choice. At this density, the onset of glassy dynamics is near $T_{\text{onset}} = 0.2$, and at $T_c = 0.07$ the dynamics has slowed down by a factor 10^4, below which conventional MD simulations do not reach equilibrium. In addition, we use a hybrid Swap Monte Carlo method [54] to prepare equilibrated configurations deep in the glass phase, down to $T = 0.035 \sim T_c/2$, to better analyze rejuvenation effects.

Protocol and observables.—We investigate the nonequilibrium dynamics of glasses during a temperature cycle sketched in Fig. 2. In the first step, an equilibrium fluid at

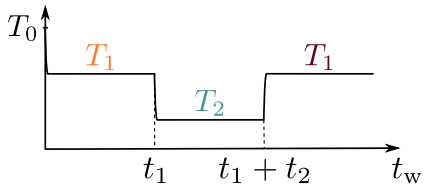

FIG. 2. Protocol: Sketch of the temperature cycle, where T_1 and T_2 are both in the glass phase while T_0 is in the fluid.

$T_0 = 0.36$ is quenched rapidly (with a rate of 3×10^{-3}) to $T_1 = 0.0353 < T_c$. The fluid falls out of equilibrium and slowly ages for a duration t_1. In the second step, the aged glass is rapidly cooled to a lower temperature $T_2 < T_1$. It stays there during a time $t_2 = t_1$, after which the system is heated back to T_1. We measure the mean-squared displacement (MSD):

$$\Delta(t_w, t_w + \tau) = \frac{1}{N}\sum_{i=1}^{N}\langle|\mathbf{r}_i(t_w + \tau) - \mathbf{r}_i(t_w)|^2\rangle, \qquad (2)$$

where t_w is the waiting time after a temperature change. This protocol is repeated using 200 independent equilibrium fluids. The brackets in Eq. (2) represent an average over these independent runs. To make a connection with experiments, we define a dynamic susceptibility [30]

$$\chi(t_w, \omega) = \frac{\Delta(t_w, t_w + \omega^{-1})}{T}, \qquad (3)$$

which plays a role analogous to the ac magnetic or dielectric susceptibility at frequency ω in experiments. This quantity also conveniently compares results at different temperatures, since typical displacements are scaled by T, which is the natural scale for particle motion. Note that our choice does not affect the time dependence in which rejuvenation and memory effects are encoded.

We shall study the role of temperature T_2 on the nonequilibrium dynamics of glasses during a temperature cycle as well as the influence of time t_1 spent at temperature T_1. In particular, we can easily study the limiting case $t_1 \to \infty$, which corresponds to reaching equilibrium at T_1 by generating equilibrium configurations at this temperature using the swap Monte Carlo method. These very stable glasses would be inaccessible by conventional MD.

Aging.—Let us focus on the first step of the temperature cycle, where fluids thermalized at $T_0 = 0.36$ are rapidly cooled to low temperature, $T_1 = 0.0353 < T_c$. The waiting time t_w measures the time spent at T_1. The resulting MSD is presented in Fig. 3, each curve corresponding to a given waiting time t_w. The curves share a similar trend. The MSD increases quadratically at small times τ, before crossing over to a plateau value during a time that depends on t_w, and eventually departs from this plateau at larger times. In terms of microscopic dynamics, this corresponds to a short-time

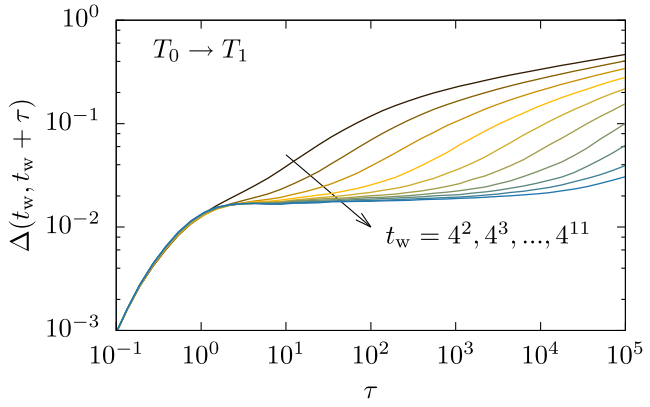

FIG. 3. Aging of the mean-squared displacement after a quench from the fluid $T_0 = 0.36$ to the glass phase $T_1 = 0.0353$. Each curve corresponds to a given waiting time t_w after the quench, and reveals a slower motion in older systems.

ballistic motion, transient trapping within an amorphous cage of neighboring particles, and eventual rearrangement of the cage. Diffusive behavior is not observed within the accessible timescale and particles actually move very little, as the MSD is typically one-tenth of particle diameter or less. At the largest $t_w = 4^{11} \simeq 4 \times 10^6$, the MSD plateaus over 4 orders of magnitude in time, meaning that the amorphous structure of the glass remains frozen over very long times. We observe a clear waiting-time dependence in the dynamics in Fig. 3. The dynamics becomes slower with the age t_w of the system, a typical property of aging systems [55,56]. This slowing down implies that the large τ data obey a subaging τ/t_w^{μ} scaling (we find $\mu \simeq 0.9$), widely known and observed in glasses of various materials [1,10,57].

Aging can be seen as a consequence of the rugged nature of the landscape of glasses. This corresponds to the thermally activated crossing of barriers, which leads the system to slowly relax towards lower energy states, where it stays for longer times [10]. The common wisdom in structural glasses is to view these glassy states as energy minima with no (or simple) internal structure [58], suggesting that no interesting dynamic effect should take place by further cooling the glass. We now present results challenging this view.

Rejuvenation.—We consider the second step of the cycle. The glasses aged during a time t_1 at temperature T_1 are suddenly cooled to $T_2 < T_1$. To investigate the influence of T_2, we present data for $T_2 = 0.01$ and $T_2 = 0.0005$. We also consider glasses of two different ages, $t_1 = 1.2 \times 10^6$ (corresponding to $t_w \simeq 4^{10}$ in Fig. 3) and $t_1 = \infty$, the latter being obtained using the hybrid swap method. As before, we measure the MSD, with t_w now being the time spent at T_2.

We start with a large temperature jump to $T_2 = 0.0005$, and report data for $t_1 = 1.2 \times 10^6$ and $t_1 = \infty$ in Figs. 4(a) and 4(b). In both panels, a strongly aging dynamics is observed, similar to the one observed in the first step in Fig. 3. The MSD evolves continuously over 5 orders of magnitude in time, with strong waiting-time dependence

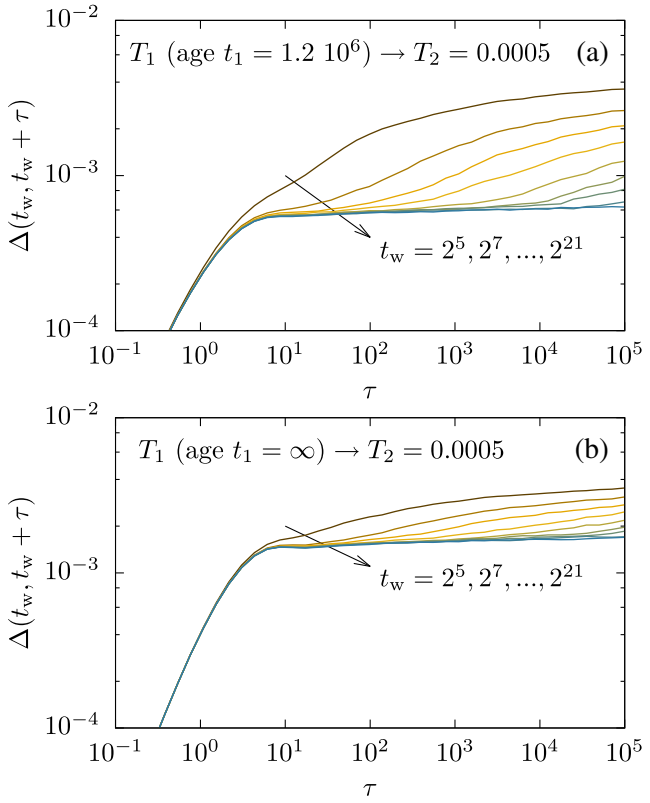

FIG. 4. Rejuvenation effect, or restart of aging dynamics, as glasses aged for (a) $t_1 = 1.2 \times 10^6$ and (b) $t_1 = \infty$ (equilibrium) at $T_1 = 0.0353$ are cooled down to $T_2 = 0.0005$.

and a variation of 1 order of magnitude in amplitude. Such strong aging effects would not be observed if the system simply had to readjust, over a fast timescale, to the new imposed temperature. Remarkably, these strong effects survive in Fig. 4(b) for $t_1 = \infty$. This implies that the aging dynamics at T_2 is not simply the continuation of the one at T_1, but that new slow processes emerge at low temperature. This is precisely the rejuvenation effect first reported in spin glasses, since very old glasses (up to $t_1 = \infty$) behave as young glasses at lower temperatures.

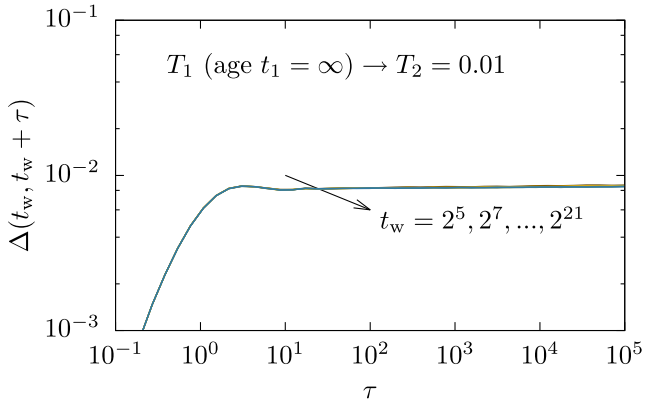

FIG. 5. No rejuvenation if T_2 is too large. Here, glasses aged for $t_1 = \infty$ at $T_1 = 0.0353$ are cooled down to $T_2 = 0.01$.

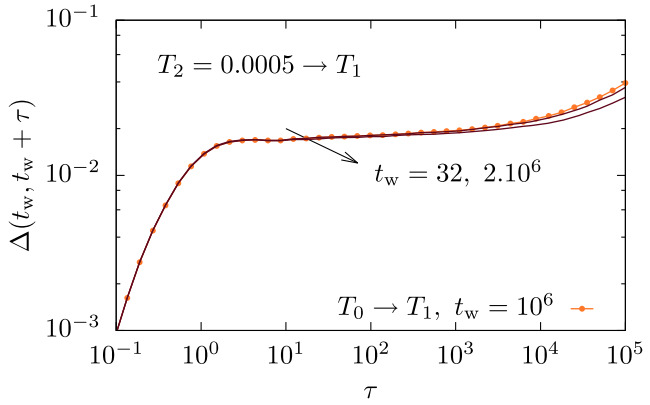

FIG. 6. Memory effect after heating the glass from $T_2 = 0.0005$ (age $t_2 = 1.4 \times 10^6$) back to $T_1 = 0.0353$. We report the MSD after heating for $t_w = 32$ and $t_w = 2 \times 10^6$ (lines). The glass has kept memory of its state at temperature T_1, as the dynamics smoothly continues that of the first cycle shown for $t_w = 10^6$ (circles).

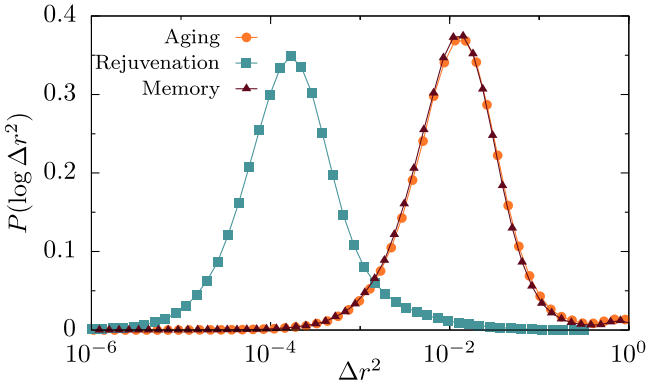

FIG. 7. Hierarchy of length scales in the probability distribution function of particle displacements Δr^2 during aging (circle), rejuvenation (square), and memory (triangle) for $t_w = 2^{20}$ and $\tau = 10^5$. All particles contribute collectively to each step, but at different scales.

Rejuvenation is not observed if T_2 is too high. We show in Fig. 5 the results of cooling from $T_1 = 0.0353$ and $t_1 = \infty$ down to $T_2 = 0.01$. Here, the dynamics does not depend on t_w, signaling the absence of aging. The frozen amorphous structure adjusts over a microscopic timescale at the new temperature T_2, and this process is not slowed down by free energy barriers. This is again similar to observations in spin glasses [26,30].

Memory.—We complete the thermal cycle by heating the glass aged for $t_2 = 1.4 \times 10^6 \simeq t_1$ at $T_2 = 0.0005$ back to $T_1 = 0.0353$. The MSD measured after the heating is shown in Fig. 6, along with the MSD at the last t_w of the first step of the cycle. After going back to T_1, the relaxation dynamics is the direct continuation of the aging which took place in the first step. Despite the strong rejuvenation effect observed at T_2 in the intermediate step, the glass has kept a perfect memory of its age at temperature T_1. The aging dynamics then continues as if the second step had not taken place at all. This is the memory effect [2,12].

We gather all these results in Fig. 1 by reporting the time evolution of $\chi(t_w, \omega = 10^{-5})$, defined in Eq. (3), during the complete temperature cycle. Aging in the first part of the cycle corresponds to a slow decay of $\chi(t_w, \omega)$, while rejuvenation corresponds to a strong restart of a similar aging. Memory is very clear as the third step appears to be the direct continuation of the first one, as emphasized in the inset where the second step is removed. The aging dynamics in the third step proceeds as a simple continuation of the first. This figure mirrors similar results obtained in spin glass materials [2,30,32,59]. The simultaneous observation of both rejuvenation and memory effects is highly nontrivial, and confirms the idea that the landscape inside glassy minima can be rugged and hierarchical in systems of soft repulsive particles that describe structural glasses [43,49].

Separation of length scales.—We have studied the probability distribution function (pdf) of single particle displacements in the three steps of the temperature protocol. At each temperature, we measure the pdf of $\Delta r^2 = |\mathbf{r}(t_w + \tau) - \mathbf{r}(t_w)|^2$ towards the end of the step, for $t_w = 4^{10}$ and $\tau = 10^5$. These distributions give additional information on the typical scale of particle displacements at each temperature (the average value is plotted in Figs. 3–6), and, more importantly on the heterogeneity of the particle displacements.

Results for the three steps are reported in Fig. 7. We observe that the pdf of displacements during aging is broad but relatively featureless, indicating that all particles are involved in the aging dynamics. A similar shape is obtained during the rejuvenation, but at a much smaller scale. This indicates that the aging dynamics in the second step is again due to very collective particle motion involving the entire system, but it involves displacements on much smaller length scales. This explains why memory of the first step is retained, as the structure obtained at the end of the first step is essentially unperturbed during the second step. Dynamics is hierarchical both in timescales and in length scales [26,57].

Discussion.—We have shown that subjecting a three-dimensional model for structural glasses to a temperature cycle reveals rich nonequilibrium dynamical effects, such as rejuvenation and memory effects that were first observed in spin glasses, but not in molecular glasses. Are these effects ubiquitous? Varying more broadly the parameters reported in this work, we find that for $\varphi = 0.85$ and $T_1 = 0.0353$, only quenches below $T_2 \approx 0.001$ will lead to rejuvenation effects in the dynamics. We also analyzed the density dependence of these effects and found that no such rejuvenation effect can be found for packing fractions beyond $\varphi \approx 0.9$. These findings are consistent with a systematic search for marginally stable glassy phases in

the present numerical model [49], which suggest that soft repulsive spheres at packing fractions relevant to describe soft colloids and granular materials are characterized by a complex free energy landscape, which should thus give rise to rejuvenation and memory effects, whereas this physics is absent in the regime describing dense supercooled liquids [47,48]. These conclusions are broadly consistent with mean-field analysis [43], and can explain the absence of rejuvenation reported for glycerol [3], and should guide future experimental studies of the dynamics of glassy materials.

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