

## Asymptotic aging in structural glasses

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Using a nonlocal Monte Carlo algorithm, we study the aging of a fragile glass, being able to follow it up to equilibrium, down to  $0.89T_{MC}$  ( $T_{MC}$  is the mode-coupling temperature), and up to long waiting times at lower temperatures. We show that the fluctuation-dissipation ratio is independent of the dynamics chosen and is compatible with a phase transition and that the scaling behavior of the aging part of the correlation supports the full-aging scenario.

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Aging is found in many complex systems out of equilibrium, such as supercooled liquids,<sup>1</sup> polymers,<sup>2</sup> colloids,<sup>3</sup> or spin-glasses,<sup>4</sup> and understanding it is a necessary step towards a unified description of such systems.<sup>5,6</sup> After a short transient since preparation, a state is reached in which one-time observables (e.g., energy, enthalpy) vary extremely slowly, while two-time quantities (correlations, susceptibilities) strongly depend on the *age* (or *waiting time*  $t_w$ , i.e., the time elapsed since preparation) of the system as well as on frequency  $\omega$  (or the measurement time  $t$ ). Despite recent efforts, our knowledge of aging of real materials is scant in the theoretically important regime of large  $t_w$  and small frequency, where universal features should show up.<sup>5</sup> Two issues still open are the scaling of correlations and the behavior of the fluctuation-dissipation ratio.

Consider observables  $A$  and  $B$  ( $B$  couples to an external field  $h$ ). The susceptibility  $\chi$  [i.e., the time integral of the linear response  $R(t_w, t+t_w) \equiv \delta\langle A(t+t_w) \rangle / \delta h(t_w)|_{h=0}$ ] and the correlation function  $C(t_w, t+t_w) \equiv \langle A(t+t_w)B(t_w) \rangle$  are expected to be of the form<sup>5</sup>

$$C(t_w, t_w + t) = C_{st}(t) + C_{ag}\left(\frac{g(t_w + t)}{g(t_w)}\right), \quad (1)$$

where  $g(t)$  is a monotonic function acting as an “effective” correlation time, and  $C_{ag}$  describes the aging of the system.<sup>7</sup> Most published studies focus on the scaling properties of  $C_{ag}$ : it is generally a function of  $t/t_w^\mu$ , but there is a lack of universality in the values of the exponent  $\mu$ , embarrassing in view of the claimed equivalence of complex systems. *Full aging* ( $\mu=1$ ) has been clearly observed so far only in spin-glasses.<sup>8</sup> For colloids, both *superaging*<sup>9</sup> ( $\mu > 1$ ) and full aging has been reported.<sup>10</sup> Polymers show rather *subaging* ( $\mu < 1$ ),<sup>2,11</sup> as has also been observed in simple liquids.<sup>12</sup> However, the values quoted often correspond to different time regimes, and the regime where  $t_w \rightarrow \infty$  with  $t/t_w$  fixed has not been carefully studied (except for spin-glasses). For example, in glycerol<sup>13</sup> full aging has not been seen either close to the glass temperature  $T_g$  (almost at equilibrium) or at

lower temperatures  $T$ . In both regimes the explored frequencies were much larger than  $1/t_w$ .

Aging is also characterized by a nontrivial behavior of the fluctuation-dissipation ratio (FDR), namely,

$$X(t_w, t+t_w) = \frac{TR(t_w, t+t_w)}{dC(t_w, t_w+t)/dt_w}. \quad (2)$$

The fluctuation-dissipation theorem (FDT) states that  $X=1$  in thermodynamic equilibrium, but this need not be so during aging, and *FDT violations* (i.e.,  $X \neq 1$ ) are observed. Experiments,<sup>14,15</sup> mean-field results,<sup>16</sup> and simulations<sup>17,18</sup> suggest that the FDR depends on time only through the correlation function, i.e.,  $X=X(C(t_w, t_w+t))$ . In structural glasses, in which we concentrate from now on, simulations also show that at fixed  $t_w$ ,  $X$  takes essentially two values:  $X(C)=1$  for  $C$  greater than some  $q_{EA}(T)$  (called the Edwards-Anderson parameter) and  $X(C)=x(t_w) < 1$  for  $C < q_{EA}(T)$ . Since  $T/X$  can be interpreted as an effective temperature  $T_{eff}$ ,<sup>16</sup> it seems that FDT violations in structural glasses can be characterized by a single time-dependent  $T_{eff}(t_w) \equiv T/x(t_w)$ , related to the slowest degrees of freedom. This lacks experimental confirmation. (Note that other definitions of effective temperatures have been explored.<sup>13,19</sup>) Also open is the issue of the behavior of  $T_{eff}(t_w)$  as  $t_w \rightarrow \infty$  (numerical data available cover only very short waiting times in the sense that one-time quantities are still quickly evolving<sup>12,18</sup>), of great theoretical interest because it is related to the possible *thermodynamic* meaning of  $T_{eff}$ .<sup>16</sup>

In this paper we study the aging dynamics down to  $0.53 T_{MC}$  ( $T_{MC}$  is the mode-coupling<sup>20</sup> temperature, below which dynamics slows down dramatically), reaching very large waiting times. This can be achieved through the use of a nonlocal algorithm [Swap Monte Carlo or SMC (Ref. 21)], which greatly accelerates the dynamics. We reach an asymptotic regime where the correlation function shows full aging within errors (supporting the analogy with spin glasses<sup>8</sup>), and where FDT violations are independent of the dynamics and of the age of the system.

We have simulated the soft-sphere binary mixture<sup>22</sup> [pair potential  $V_{AB}(r)=(\sigma_{AB}/r)^{12}$ , diameter ratio 1.2], a simple fragile glass former, using a *nonlocal* Metropolis Monte Carlo algorithm [hereafter SMC (Ref. 21)] which adds swap moves (with probability  $p$ ) to standard *local* Monte Carlo (LMC). Although swap acceptance is very low ( $\approx 3 \times 10^{-3}$ ) the equilibration time is considerably shortened; e.g., at  $0.89T_{MC}$  extrapolations estimate it to be three orders of magnitude larger for LMC than for SMC (note that other nonlocal algorithms have proved useful in simulations of structural glasses<sup>23</sup>). We used the following protocol: Starting from a random configuration, a system of  $N=2048$  particles was instantaneously quenched to the final temperature  $T$ , and allowed to evolve for  $t_w$  steps. This preparation was done with the SMC algorithm with  $p=0.1$ , which gives the faster equilibration for this system size. After  $t_w$ , the correlation and response functions in the presence of an external field  $h$  were computed, mostly in SMC runs with  $p=0.1$ , but also in LMC and SMC runs with different  $p$  in order to assess the dependence of the results on the dynamics. Due to the swap moves, particle diffusion is not a convenient observable. Instead, we divided the simulation box in  $N_c$  cubic subcells and considered the quantity

$$A(t) = \frac{1}{N_c} \sum_{\alpha=1}^{N_c} \epsilon_{\alpha} n_{\alpha}(t), \quad (3)$$

where  $\epsilon_{\alpha} = \pm 1$  randomly and  $n_{\alpha}$  is the occupation number of subcell  $\alpha$ . The side of the subcells was about  $0.35\sigma_{AA}$  so that essentially  $n_{\alpha}=0,1$ . Note that swap moves do not change  $A(t)$ . To measure response, a term  $\lambda NA$  was added to the Hamiltonian, with  $\lambda = h k_B T$  ( $h$  is dimensionless). We considered the correlation  $C(t_w, t_w+t) = \langle NA(t_w)A(t_w+t) \rangle$ , where  $\langle \dots \rangle$  means average over both thermal histories and the  $\epsilon_{\alpha}$ , together with the integrated response  $k_B T \chi(t_w, t_w+t) = \langle A(t_w+t) \rangle / h$ .<sup>24</sup>

With SMC we can equilibrate the system down to  $T = 0.89T_{MC}$ . The correlation  $C(t_w, t_w+t)$  shows aging up to  $t_w = 10^5$ , but does not change between  $t_w = 10^5$  and  $10^6$ , which is approximately the region where the energy reaches a stationary value (Fig. 1). We conservatively estimate the autocorrelation time as the time  $\tau$  needed for  $C$  to reach the asymptotic value  $N/N_c$  ( $\sim 0.04$ ), obtaining  $\tau = 2 \times 10^5$ , much smaller than  $10^6$  (the total length of the simulation). Hence we claim that the system has equilibrated, which is further confirmed by the fact that the FDT holds. In contrast, well below  $0.89T_{MC}$  the system is out of equilibrium up to  $t_w = 2 \times 10^7$  (our largest observational time). A stretched exponential fit of the equilibrium correlation in the late  $\alpha$ -relaxation regime yields a stretching exponent  $\beta \sim 0.3$ . The equilibrium LMC correlation function does not decay to  $N/N_c$  within the simulated times; hence it is still an open point whether SMC changes the shape of the correlations in equilibrium, or whether the two dynamics are related by a simple rescaling of time.

We first address the issue of the scaling of the correlation during aging at  $T = 0.53T_{MC}$  (in general far below  $T_g$ , e.g. for glycerol this corresponds to  $T \sim 140$  K, while  $T_g \sim 190$  K). With SMC we find (Fig. 2) that the correlations for  $t_w = 5$

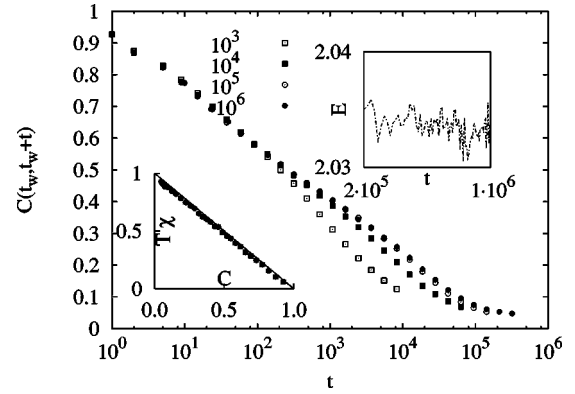


FIG. 1. Correlation function  $C(t_w, t_w+t)$  vs  $t$  for  $T=0.89T_{MC}$  at  $t_w=10^3, 10^4, 10^5, 10^6$  (24 samples). Bottom, left: integrated response  $T\chi$  vs correlation function  $C$  at  $t_w=10^6$ . Top, right: Energy per particle  $E$  vs  $t$  during a SMC quench with  $p=0.1$ . Error bars are of the order of point size.

$\times 10^5$ ,  $5 \times 10^6$  can be made to collapse by plotting them as a function of  $t/t_w^\mu$  with  $\mu=1.05(6)$ , compatible with full aging. The collapse applies to the aging part [ $C_{ag}$ , Eq. (1)], which dominates the correlation for  $t/t_w > 0.1$  ( $\omega t_w < 10$ ), as has also been observed in spin glasses.<sup>8</sup> The two shortest  $t_w$ 's (inset) can instead be scaled with  $\mu \sim 0.85$ . The same value (within errors) was found in molecular dynamics simulations of the Lennard-Jones binary mixture,<sup>12</sup> so we argue that the accelerated dynamics does not affect the scaling. If one insists on scaling all curves, it can be done reasonably well using  $\mu \sim 0.9$ , though this is likely an artifact of mixing two different regimes. The relevant point is that  $\mu \sim 1$  is seen clearly only for  $t_w \gg 1$  and in the  $t \sim t_w$  region, which is where it is expected to hold<sup>9</sup> if structural glasses share the dynamic properties of spin-glasses.<sup>5</sup> The failure of full aging for  $t/t_w \ll 1$  is hence in agreement with dielectric susceptibility measurements in glycerol.<sup>13</sup> We are not aware of experimental studies in the conditions where we find full aging, but such measurements are clearly needed.

A second important result is that although the susceptibility and correlation are affected by the choice of dynamics, the FDR is not. In fact, Fig. 3 shows the ratio  $T_{eff}/T$  (i.e., the inverse of the FDR) at  $T=0.89T_{MC}$  during aging and up to

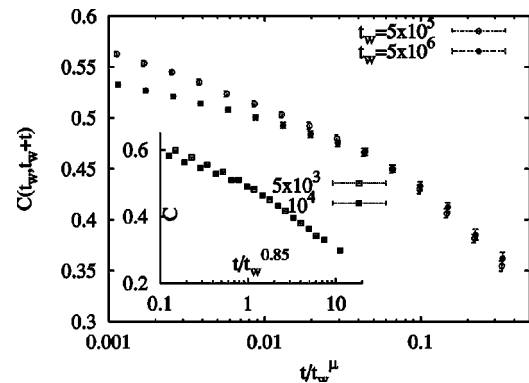


FIG. 2.  $C$  vs  $t/t_w^\mu$ ,  $\mu=1.05(6)$ , for  $t_w=5 \times 10^5, 5 \times 10^6$  at  $T = 0.53T_{MC}$  from SMC runs (24 samples). Inset:  $C$  vs  $t/t_w^{0.85}$  for  $t_w = 5 \times 10^3, 10^4$  (48 samples).

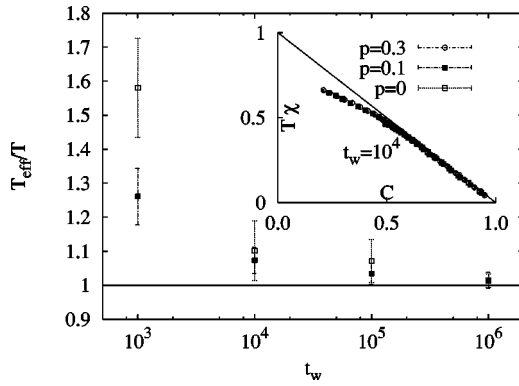


FIG. 3.  $T_{\text{eff}}/T$  vs  $t_w$  for SMC and LMC runs at  $T=0.89T_{\text{MC}}$  (16 samples), obtained by a linear fit of the points of the parametric  $T\chi$  vs  $C$  plots deviating from the FDT line. Errors were estimated with the jackknife method (Ref. 25). Inset:  $T\chi$  vs  $C$  for  $t_w=10^4$  at  $T=0.53T_{\text{MC}}$  for  $p=0$  (LMC) and  $p=0.1, 0.3$  (SMC),  $N=20000$  (8 samples).

equilibration for both SMC and LMC algorithms, obtained measuring the FDR in simulations that used configurations taken along the SMC quench as a starting point. After a short transient ( $\sim 10^4$  steps) the FDR's become indistinguishable within errors. At  $T=0.53T_{\text{MC}}$  and with LMC, we can reach the region of FDT violations only for  $t_w=10^4$ , so we look at the FDR at fixed  $t_w$  for LMC and SMC with  $p=0.1$  and  $0.3$ , obtaining a good agreement (Fig. 3, inset).

Finally, we investigate the FDR for large times at  $T=0.53T_{\text{MC}}$ . In Fig. 4 we plot  $T_{\text{eff}}$  at  $t_w=5 \times 10^3$ ,  $10^4$ ,  $5 \times 10^5$ , and  $5 \times 10^6$  as a function of the instantaneous inherent structure (IS) energy  $E_{\text{IS}}(t_w)$ . We also plot  $T_{\text{eff}}$  computed according to the IS approach,<sup>18</sup>  $T_{\text{eff}}^{-1} = \partial \Sigma / \partial f$ , where  $\Sigma(f)$  is the logarithm of the number of IS with free energy  $f$ , and  $\partial \Sigma / \partial f$  is obtained as in Ref. 18. This idea (which makes no prediction about the  $t_w \rightarrow \infty$  limit of  $T_{\text{eff}}$ ) had previously been confirmed only in the very early aging regime by molecular dynamic simulations.<sup>18</sup> Our results show a reasonable agreement even at quite large times.

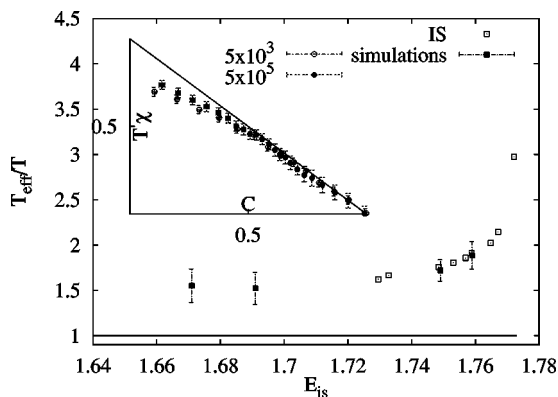


FIG. 4.  $T_{\text{eff}}/T$  for  $T=0.53T_{\text{MC}}$  vs the instantaneous IS energy  $E_{\text{IS}}(t_w)$  measured in SMC runs.  $E_{\text{IS}}(t_w)$  is found by energy minimization starting from 50 instantaneous configurations within a small time window ( $\ll t_w$ ) around  $t_w$ . We also show the  $T_{\text{eff}}$  predicted by the IS approach (Ref. 18). Inset: the  $T\chi$  vs  $C$  plot for two different  $t_w$ .

The limiting value of  $T_{\text{eff}}$  as  $t_w \rightarrow \infty$  is of great theoretical interest. If the system eventually equilibrates, then  $T_{\text{eff}} \rightarrow T$ , as we have found for  $T=0.89T_{\text{MC}}$ . Approaches that consider aging a result of critical slowing down due to the proximity of a critical point which is never reached (because it is located at  $T=0$ ,<sup>26</sup> or because of the impossibility to establish a “liquid” long range order<sup>27</sup>) predict this to be the case for all temperatures. A different view relates the asymptotic value of the FDR to a thermodynamic transition described by replica symmetry breaking.<sup>28</sup> Above the transition,  $X(C)$  is predicted to reach slowly the equilibrium value 1 [so  $T_{\text{eff}} \rightarrow T$  (Ref. 6)], while below the FDR it should remain nontrivial and  $T_{\text{eff}}$  should tend to a constant  $>T$ , since the system never equilibrates. In this scenario the asymptotic FDR is claimed to classify complex systems in universality classes.<sup>5,28</sup> A third possibility is that FDT violations are due to nucleation and slow growth of the crystal phase,<sup>29</sup> in which case at long times one expects the coarsening regime to be reached, and so  $T_{\text{eff}} \rightarrow \infty$ .

Our results for  $0.53T_{\text{MC}}$  do not seem to support this last possibility. The data are instead compatible with the presence of a thermodynamic replica symmetry breaking (RSB) transition,<sup>28</sup> since FDR does not seem to change between  $t_w=5 \times 10^5$  and  $t_w=5 \times 10^6$  ( $E_{\text{IS}}$  are, respectively, 1.691 and 1.671). Note that this is the same regime where the system displays full aging. It cannot be excluded that  $T_{\text{eff}} \rightarrow T$ , but it looks less likely if we note that extrapolating  $E_{\text{IS}}(t_w)$  to  $t_w \rightarrow \infty$  with a power law gives an asymptotic  $E_{\text{IS}}=1.642$ . In the first approximation the RSB approach predicts that  $T_{\text{eff}}$  equals the transition temperature, which unfortunately has been only roughly estimated.<sup>28</sup> We just observe the fact that, at the qualitative level, the measured  $T_{\text{eff}}/T$  in Fig. 4 levels off at a value greater than 1 in the late aging regime supports the RSB scenario.

In summary, we have studied numerically the late aging regime of a simple glass-forming liquid using local and non-local Monte Carlo (SMC). We find that the scaling of the correlation functions and the FDR during aging do not depend on the dynamics. This is a strong generalization of the previous finding<sup>30</sup> that equilibrium relaxation in the Lennard-Jones mixture is qualitatively identical under different *local* dynamics (except, as here, for very short times). We have found that correlation functions in the late aging regime show within errors full-aging scaling, suggesting an equivalence between the aging dynamics of structural and spin glasses. This should be searched experimentally at frequencies comparable or shorter than  $1/t_w$ . We also measured the FDR while taking one-time quantities closer to asymptotic values than in previous studies. FDT violations do not imply a thermodynamic transition. However, if a transition does exist, there should be a correspondence between the asymptotic  $T_{\text{eff}}$ , which is accessible to experiments, and the order parameter, which is not.<sup>31</sup> The FDR's measured in experiments<sup>14,15</sup> and simulations<sup>18</sup> up to now depend strongly on the age of the system; hence their utility in investigating the existence of a transition is still an open point.

Here, we have been able to reach a regime where  $T_{\text{eff}}$  has no noticeable time dependence. Interestingly enough, it coincides with the full-aging regime. At the lowest temperature, the  $T_{\text{eff}}$  measured over a time window of 3 orders of magnitude approaches a finite value, different from the equilibrium temperature. This seems only slightly compatible with a critical slowing down ( $T_{\text{eff}} \rightarrow T$ ) or the growth of a crystal phase ( $T_{\text{eff}} \rightarrow \infty$ ) and favors instead the phase transition scenario. Our result suggests that the relevant information for an understanding of aging in structural glasses has to be looked

for in a regime that so far had not been investigated, either in experiments or in simulations.

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