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## Real-space model for activated processes in rejuvenation and memory behavior of glassy systems

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We offer an alternative real-space description, based purely on activated processes, for the understanding of relaxation dynamics in hierarchical landscapes. To this end, we use the cluster model, a coarse-grained lattice model of a jammed system, to analyze rejuvenation and memory effects during aging after a hard quench. In this model, neighboring particles on a lattice aggregate through local interactions into clusters that fragment with a probability based on their size. Despite the simplicity of the cluster model, it has been shown to reproduce salient observables of the aging dynamics in colloidal systems, such as those accounting for particle mobility and displacements. Here, we probe the model for more complex quench protocols and show that it exhibits rejuvenation and memory effects similar to those attributed to the complex hierarchical structure of a glassy energy landscape.

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### 1. Introduction

The morphology of energy landscapes in high-dimensional configuration spaces is at the heart of complex dynamics for a broad range of statistical systems.<sup>1,2</sup> Examples are legion in disparate systems: glassy materials like amorphous fluids,<sup>3</sup> jammed grains,<sup>4,5</sup> colloids,<sup>6,7</sup> disordered magnets,<sup>8,9</sup> crumpling sheets,<sup>10,11</sup> or entangled polymers,<sup>12–15</sup> all face frustration while relaxing their free energy. Due to competing variables exerting geometric or energetic constraints on each other, a complex, multimodal landscape is imposed on the space of all possible configurations. A universal framework of how a system traverses its landscape can elucidate its collective states, and the transitions between them that result in different phases of behavior. Such a framework is therefore of wide ranging scientific interest.

A standard approach to gain insight into the complexity of the landscape of a glassy system, whether in experiment or in simulation, is through a hard quench<sup>12</sup> from the liquid-like high-temperature (or low-density) to a low temperature (or high density) regime, initiating a non-equilibrium relaxation dynamics known as aging.<sup>3,5,16–28</sup> Such a quench takes the system instantly deep into the glassy landscape. There, a hierarchy of barriers emerges that quite naturally calls for an effective description of the ensuing dynamics in terms of a sequence of activated events that is called record dynamics (RD),<sup>29</sup> since that hierarchy renders all but the largest fluctuations ineffectual and relaxation is characterized by timescales

for barrier crossings that exceed all others.<sup>9,30–32</sup> There is significant experimental evidence indicating the dominance of such large, intermittent events in driving the relaxation dynamics.<sup>10,11,26,33–37</sup> Alternative approaches to describe aging in terms of intermittent events.

As discussed in ref. 38 RD derives its generality from its small set of assumptions about the properties of the energy landscape for a generic glassy system. Energy landscapes are a widely applicable concept across many areas in science and engineering,<sup>2</sup> describing the configuration space of systems with a large number of degrees of freedom. For RD within that concept, we merely need to stipulate (1) that a complex energy landscape has a rapidly (say, exponentially) growing number of meta-stable states for increasing energy and (2) that lower-energy meta-stable states are more stable, *i.e.*, have higher energy barriers against escape, than those at higher energy. Then, a hard quench entrenches the system with high probability in one of the far more prolific meta-stable states of higher energy, which the system explores through (reversible) quasi-equilibrium fluctuations. Only progressively larger (and increasingly rare), record-sized fluctuation events allow the system to overcome ever larger barriers to tumble irreversibly into the next, marginally more stable local energy minimum.

Accordingly, in RD incremental relaxation is coarse-grained into a sequence of record barrier crossing events that are required to unlock farther reaches in the landscape.<sup>32,39</sup> These records drive the dynamics (*i.e.*, “set the clock”) in disordered materials, generically, reminiscent of the concept of “material time”.<sup>40,41</sup> This “clock” for records decelerates at a rate  $\lambda(t) \propto 1/t$ , as new records are ever harder to achieve. Dynamics

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proceeds homogeneously in  $\log t$  instead of in linear time,<sup>31,42</sup> as observed in many experiments for polymers,<sup>15,43</sup> colloids,<sup>26,44,45</sup> granular piles,<sup>5,46</sup> or crumpling sheets,<sup>10</sup> obtaining for the accumulation of events  $\langle n(t, t_w) \rangle \sim \int_{t_w}^t \lambda(\tau) d\tau \sim \log(t/t_w)$ . Then, any two-time correlations become subordinate<sup>36</sup> to this clock:  $C(t, t_w) = C[n(t, t_w)] = C(t/t_w)$  for times  $0 < t_w < t$  after the quench, as shown, e.g., in ref. 38.

As a real-space incarnation of RD, a simple on-lattice “cluster model” has been designed<sup>47</sup> that captures the combined temporal and spatial heterogeneity found in a generic aging system. Despite its simplicity, the model has already been shown to reproduce<sup>38,48</sup> salient experimental<sup>26</sup> and simulational<sup>49</sup> results for quenches in colloids.

Further subjecting an aging system to a protocol of temperature shifts should trigger rejuvenation and memory effects. In these protocols, the aging process restarts after a second quench (rejuvenation), and resumes the dynamics prior to the second quench upon reheating, thus having memory. In spin glasses, it is easy to demonstrate rejuvenation and the imprinting of entire histories<sup>50–55</sup> under small variations of temperature and fields after a quench. Similarly, in polymeric and colloidal systems, memory and rejuvenation effects have been known for a long time.<sup>56–58</sup> Recently, using extensive MD simulations of a structural (colloidal) glass, it was argued that these effects validate mean-field predictions from spin glass theory.<sup>59</sup> Here, following the protocol of ref. 59, we demonstrate similar rejuvenation and memory effects in the cluster model of RD. Reproducing almost the entire phenomenology in such a minimalistic setting highlights the role of rare activated processes, which are the only driving mechanism in the cluster model. Merely the lack of a realistic equilibrium state in this model leads to unphysical behavior at infinitely long times.

## II. Methods

### A. Cluster model

In the cluster model,<sup>47</sup>  $N = L^d$  particles completely fill a (hyper-cubic) lattice of base-length  $L$  in dimension  $d$ , one on each site at all times. Yet, each particle by itself is either isolated and forms a cluster of size  $h = 1$ , or it is jammed in with adjacent particles as a member of a cluster of size  $h > 1$ . Isolated particles ( $h = 1$ ) possess independent mobility, those in clusters with  $h > 1$  are locked in and require activation to become mobile. At the time of the quench,  $t = 0$ , all particles are mobile, owing to the prior “liquid” high- $T$  or low-density state of the system. Each update step  $s$ , one randomly chosen site is picked for an update. (Time is measured in units of lattice sweeps,  $t = s/N$ .) There are two possible outcomes, depending on the state of the particle on that site:

(1) A mobile particle interacts with a randomly chosen neighbor and both exchange position, the basic unit of mobility in the model. Whether that neighbor itself was mobile ( $h = 1$ ) or already part of a larger cluster ( $h > 1$ ), the addition of the mobile particles now leads to a (jammed and thus immobile) cluster with  $h' = h + 1 > 1$ .

(2) An immobile particle jammed inside a cluster of size  $h > 1$  may activate a barrier-crossing event with an  $h$ -dependent probability per sweep,<sup>60</sup>

$$P(h) \propto e^{-\beta h}. \quad (1)$$

If it occurs, such an event will break the cluster and create  $h$  newly mobilized particles.

Thus, following a quench out of the initial liquid state of mobile particles, clusters form and break up irreversibly to remobilize and re-distribute their particles to neighboring clusters. For a sufficiently large value of the external control parameter  $\beta$  (that acts as a density or an inverse temperature), a large fraction of particles soon accrete into jammed clusters that only intermittently break up and almost instantaneously feed their particles into ever fewer – and thus ever larger – neighboring clusters, which in turn necessitate ever larger and thus ever more rare fluctuations, requiring a time exponential<sup>9,61</sup> in the size of those clusters. The effect of all regular fluctuations that only rarely achieve such a significant event beyond reversible in-cage rattle is coarse-grained into  $P(h)$  in eqn (1). Cluster growth ultimately decelerates the dynamics, since only larger and fewer clusters remain, which signifies the slow structural changes that characterize aging.<sup>10,26</sup> Note that high or low “density” in this model is dictated *via* the choice of the temperature-like parameter  $\beta$  in eqn (1), not by the actual (and always uniform) filling of the lattice.

In ref. 47 the two-time mean-square displacement (MSD),

$$\Delta(t, t_w) = \frac{1}{N} \sum_{i=1}^N \langle |\vec{r}_i(t) - \vec{r}_i(t_w)|^2 \rangle, \quad (2)$$

was shown to grow logarithmic as  $\Delta(t, t_w) \sim A \ln(t/t_w)$ , depending on the waiting time  $t_w$  after the quench when the measurement commences. In RD, this is a direct consequence of the  $\sim A/t$  decline<sup>62</sup> in the rate of cluster break-up events (such a rate for irreversible events was explicitly verified in experimental data for aging colloids<sup>38</sup>). In Fig. 1, we demonstrate that the proportionality factor  $A$  is a function of  $\beta$ , similar to what has been observed for domain growth in spin-glass simulations,<sup>63</sup> granular compaction,<sup>5</sup> but also for MSD in colloidal experiments at different densities.<sup>38</sup>

### B. Waiting-time method

In the following, we use two-step quench temperature cycles in the cluster model to elicit rejuvenation and memory effects. To that end, we will simulate the model on a two-dimensional lattice of base length  $L = 64$  throughout, employing periodic boundary conditions. Such investigations require measurements over many decades in time, which is conveniently accomplished in the cluster model using a waiting-time algorithm.<sup>47,64</sup> This method orders all possible events in a chronology based on their probability of occurring in eqn (1), thus avoiding rejected moves that occur in conventional Monte Carlo algorithms. Given eqn (1), newly freed particles will join clusters on sub-sweep time scales, meaning that each cluster break-up event will typically move many particles nearly



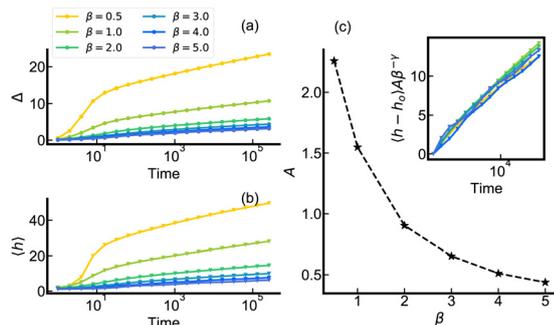


Fig. 1 Increase of (a) MSD and (b) average cluster size with the logarithm of time  $t$  for  $t_w = 1$ , for different values of  $\beta$  in eqn (1). The results show that motion slows systematically with increasing  $\beta$ . At the shortest times, fast transient effects resulting from the quench predominate, leading to an instant jump  $\Delta_0$  in MSD or  $h_0$  in cluster size, before the logarithmic scaling sets in. Panel (c) shows the dependence of the log-slope  $A$  on  $\beta$  in fitting  $\Delta \sim A \ln(t)$  to the cluster sizes  $\langle h \rangle$  in (b), yielding  $A \approx \beta^{-\gamma}$  with  $\gamma \approx 0.7$ . The inset demonstrates the collapse of the appropriately rescaled data from (b).

simultaneously. That said, this method can rapidly telescope into the future within just a few update steps when break-ups become rare. Specifically, each of the  $k$  clusters is assigned a survival time,  $\{\delta t_{ij}\}_{i=1}^k$ , based on its current size  $h_i$  according to<sup>64</sup>

$$\delta t_i = -\log(X_i)/P(h_i), \quad (3)$$

with  $P(h_i)$  as given in eqn (1) and made stochastic by employing a random number  $X_i$  sampled from a uniform distribution. Then, the event with the lowest  $\delta t_{\min} = \min_i \{\delta t_{ij}\}$  is selected, updating the global time to  $t + \delta t_{\min}$  and assigning a new  $\delta t$  to the most recently modified clusters.

### III. Results and discussion

In Fig. 2, we illustrate that our simple model is capable of exhibiting both rejuvenation and memory effects. Following ref. 59, we employ the MSD given in eqn (2) to define a dynamic susceptibility function

$$\chi(t_w, \omega) = \beta \Delta(t_w + \omega^{-1}, t_w), \quad (4)$$

where  $\tau = \omega^{-1}$  sets a time-window over which the decay of the instantaneous mobility at time  $t_w$  is assessed. The initial quench occurs from an infinite temperature ( $\beta = 0$ ) to  $\beta_1 = 0.5$ . At that point,  $\chi$  drops as a function of  $t_w$ , using a window size of  $\tau = 2$  (in sweeps), while the system is aged up to  $t_1 = 50$  sweeps. At that time the system has developed a Poissonian cluster-size distribution with average cluster size reaching about  $\langle h \rangle \approx 30$ , see Fig. 1(b), leaving a number of the smallest and most marginally stable clusters below that size most likely to break. In fact, as illustrated in Fig. 2(c), a fraction of those clusters are in the process of steadily collapsing at time  $t_1$  in a large enough system. (Since clusters, as well as correlations between clusters, only grow very slowly, there is little difference in averaging over the evolution of one large or many smaller systems, as long as  $\langle h \rangle^{1/d} \ll L$ .)

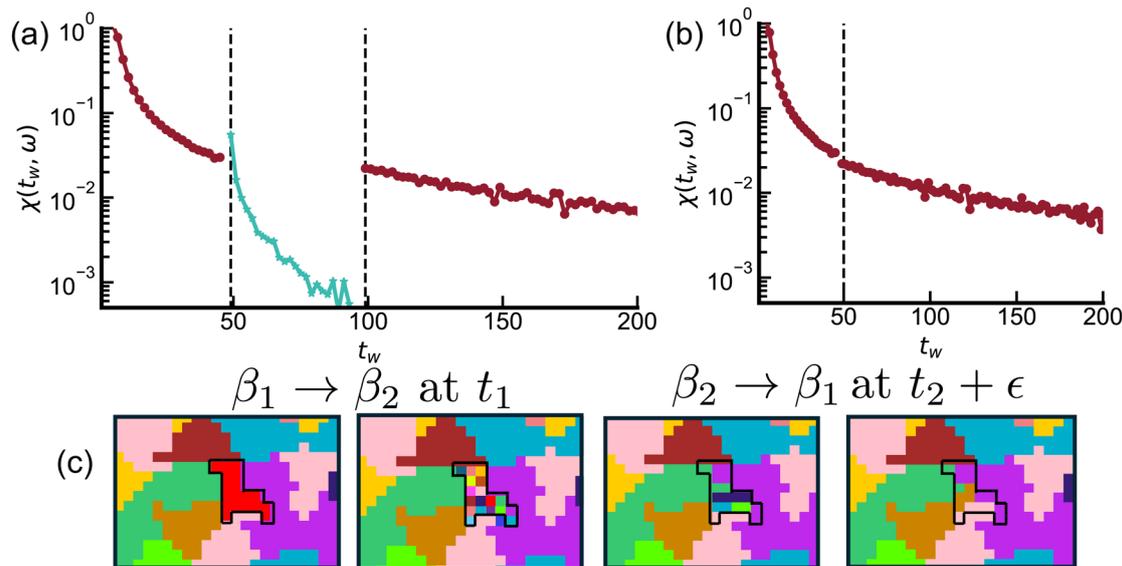
At time  $t_1$ , we perform a further quench of the system, down to  $\beta_2 = 5$ . At this much reduced temperature, only clusters below the corresponding average cluster of  $\langle h \rangle \approx 3$ , as taken from Fig. 1(b), would qualify as unstable on this time-scale, *i.e.*, susceptible to breaking up in a time of the order of  $t_1$ . Clearly, all of the existing clusters are much too large and are completely frozen at this temperature. Only those currently freed particles from the cluster break-ups can contribute to the instantaneous mobility in this part of the temperature cycle. This small but extensive fraction of mobile particles, in turn, relives the entire history of an aging system freshly quenched to  $\beta_2$ , within the background of otherwise frozen clusters. As in ref. 59, the overall reduction in mobility  $\Delta$  is partially compensated by the relative factor of  $\beta$  in the definition of  $\chi$  in eqn (4): Here, we have  $\beta_2/\beta_1 = 10$ , while it is  $\approx 70$  in ref. 59. Thus,  $\chi$  “rejuvenates”, immediately jumping up above the previous level reached before  $t_1$ , before decaying itself. When the temperature is then reset to  $\beta_1 = 0.5$  after  $t_1 + t_2 = 100$  sweeps, the impact left by the rejuvenating sub-system had a minimal effect on the entire system. Merely those clusters in the process of breaking up at  $t_1$  already have advanced minutely. Accordingly, its instantaneous mobility returns to the level frozen in at  $t_1$ .

As a further validation of rejuvenation, ref. 59 compared the age-dependent (two-time) MSD observed following the initial quench to  $\beta_1$  with the MSD found after the second quench to  $\beta_2$  while using its starting point  $t_1$  as the new origin of time. Indeed, in their Fig. 3 and 4(a), they demonstrate that in both measurements the two-time MSD behaves analogously, as if  $t_1$  was an entirely independent quench.

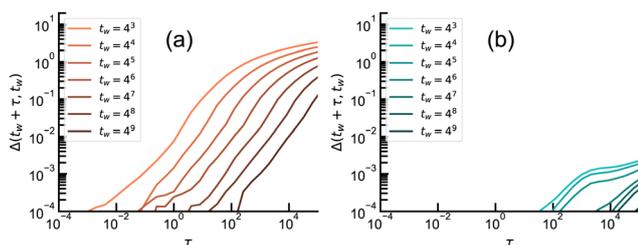
To replicate these results in RD, we employ for the cluster model the same setting as in Fig. 2 but with a simple quench to  $\beta = 0.5$ . Now, the system is aged (without second quench) up to various waiting times  $t_w$  to measure MSD  $\Delta(t_w + \tau, t_w)$  for the lag-time  $\tau = t - t_w$ . This data is plotted in Fig. 3(a), which reproduces Fig. 3 of ref. 59. It demonstrates that a system that was aged up to a time  $t_w$  remains confined for a corresponding time  $\propto t_w$  before exhibiting any discernible MSD. Incidentally, this fact, as well as a collapse of this data as function of  $t/t_w$ , was previously explained for experiments on colloids in terms of RD in ref. 38. Although mean-field arguments would suggest that MSD after a transient should saturate at long times,<sup>65</sup> the existence of activated dynamics in real systems induce further (logarithmic) growth.

More importantly, the rejuvenation effect seen in Fig. 4(a) of ref. 59 is captured for the cluster model in Fig. 3(b) which presents the two-time MSD of particles for several  $t_w$  during the second stage of the temperature cycle. Having undergone the initial quench to  $\beta = 0.5$ , the dynamics are evolved up to time  $t_1 = 50$  sweeps, at which time the system is cooled down even further to  $\beta = 5$ . Once the particles are quenched to the second temperature, they are aged up to a given waiting time  $t_w$ , now taking  $t_1$  as the new origin of time. As above, the dynamics are measured as a function of lag-time  $\tau = t - t_w$  for each  $t_w$ . While the MSD after the second quench differs by a magnitude compared to Fig. 3, the  $t_w$  dependence shows that rather than continuing the dynamics from the prior quench, the



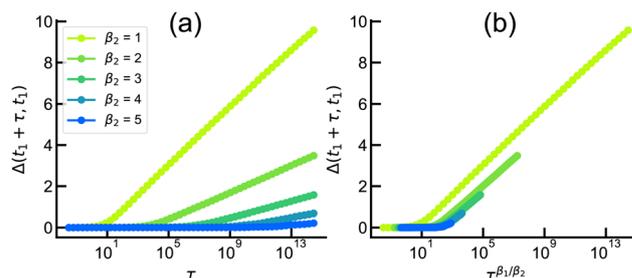


**Fig. 2** Rejuvenation and memory effects produced on an  $L = 64$  square lattice subject to a temperature cycle. The system at  $t_w = 0$  undergoes a hard quench to  $\beta_1 = 0.5$ , ages until time (in sweeps)  $t_1 = 50$ , when temperature is reduced once more to  $\beta_2 = 5$ . After aging further until  $t_2 = 100$ , it is reset to  $\beta_1$ . In (a), the susceptibility  $\chi$  defined in eqn (4) is plotted as a function of  $t_w$  using  $\tau = \omega^{-1} = 2 \ll t_1$ . In turn, (b) shows that  $\chi$ , when reheated at  $t_2$ , is a continuation of the dynamics from the system prior to the second quench at  $t_1$ . Both can therefore be “stitched together”. In (c), a physical depiction of the situation is provided. This row shows the cluster formation by zooming in on a small part of the lattice (different colors indicate distinct clusters). The region most affected by the quench at  $t_1$  is outlined in all the snapshots. There, some cluster of size  $h = 18$  happens to be in the process of breaking up. Solely its freed particles are able to move during a time window of size  $\tau = 2$  after  $t_1$ . A few of them attach to neighboring clusters, the remaining ones form small clusters that can survive for a long time at  $\beta_2 = 5$ . When the lattice is reheated to  $\beta_1$  at time  $t_2$ , those small cluster almost instantly (*i.e.*, in a small time interval  $\epsilon \ll t_2$ ) break up and their particles integrate into the surrounding clusters, as they would have done without the second quench at  $t_1$ . Thus, the cluster-size distribution at  $t_1$  is virtually identical to that at  $t_2$ , once reheated, which is why the dynamics in panel (b) appears to pick up where they left off prior to the second quench.



**Fig. 3** MSD (a) after quench to  $\beta_1 = 0.5$  (without subsequent temperature changes) and (b) after a subsequent quench at  $t_1 = 50$  to  $\beta_2 = 5.0$ . In both cases, the quenched system is aged up to certain waiting time  $t_w$ , before the dynamics of the particles are measured relative to the configuration at  $t_w$  as a function of lag-time  $\tau = t - t_w$ . Both (a) and (b) show the characteristic dependence of MSD on the age  $t_w$ . For (b) this implies that the second quench actually rejuvenated the system, albeit at a much lower mobility due to the lower temperature. Note that large fluctuations which ensue due to a rare event account for the large gap between MSD curves measured at  $t_w = 4.5$  and  $t_w = 4.6$ .

process re-initializes and dynamics are refreshed based on  $t_w$ , the age of the system following the second quench at  $t_1$ . It is apparent from this analysis that in the cluster model the intervening quench to  $\beta_2$  (if it is not excessively long, see below) leaves little mark on the large fraction of frozen-in clusters, which on re-heating at  $t_1 + t_2$  continue their mobility where it froze in at  $t_1$ .



**Fig. 4** Demonstration for the end of memory. In (a), we measure the MSD  $\Delta(t_1 + \tau, t_1)$  for particles in the cluster model initially quenched to  $\beta_1 = 1.0$ , then aged for  $t_1 = 25$  sweeps, when it undergoes the second quench to  $\beta_2$ . The system remains entrenched in its meta-stable state attained at  $t_1$  for a time  $\tau = \tau_2$  that depends on  $\beta_2$ , before significant displacement occurs that erases the memory of that state. In (b), this data collapses when  $\tau$  is rescaled according to eqn (5).

We note that the coarse-grained motion in our model by design eliminates both, the (trivial) initial ballistic motion and the subsequent rattle particles experience at the shortest times while confined within their cages. Such in-cage rattle contributes to a visible plateau in the MSD of experiments or of continuum MD simulations, as seen in Fig. 3 and 4(a) of ref. 59. Accordingly, such a plateau is absent in our study, in which particles are bound to discrete lattice sites until an actual event occurs.



One aspect of rejuvenation in spin glasses<sup>66</sup> or the MD simulations<sup>59</sup> that the cluster model can not reproduce concerns the infinite time limit  $t_1 \rightarrow \infty$ . Even in that case, rejuvenation – albeit in a very weak form – is observed. This may not be too surprising, since this situation parallels the original quench that puts a glassy system out of equilibrium into an aging state, only that this quench to  $\beta_2 > \beta_1$  commences from a temperature below the glass transition,  $\beta_1 > \beta_g$ , instead of from  $T = \infty$  ( $\beta = 0$ ). Nonetheless, the glassy system is dislodged from equilibrium into a non-equilibrium state, however minutely, and aging ensues. In the cluster model, such an equilibrium state does not exist: at infinite time for  $\beta_1 > \beta_g$ , there would be just one large cluster spanning the system, whose eventual break-up would erase all memory of  $\beta_1$  and any distinction with  $\beta = 0$ .

Finally, we point out that the cluster model reproduces other properties predicted for systems exhibiting rejuvenation and memory effects. For instance, for spin glasses it was shown in ref. 66 that the memory effect may diminish for a very long rejuvenation stage. In Fig. 2, the system ages from the initial quench at temperature  $\beta_1$  and at  $t_1$  has entrenched itself in a meta-stable state of some typical free-energy barrier  $\Delta F$ . To escape the memory of that state at  $\beta_1$ , a record fluctuation is needed, which according to RD typically occurs at time  $\tau_1 \approx t_0 \exp\{\beta_1 \Delta F\}$  with  $\tau_1 \sim t_1$ , where  $t_0$  is some system-specific microscopic time. Quenching anew at  $t_1$  from  $\beta_1$  to  $\beta_2$  leaves the system even deeper entrenched within that state, now needing a time  $\tau_2 \approx t_0 \exp\{\beta_2 \Delta F\}$  to escape and lose its memory. With or without second quench at  $t_1$ , the clusters formed at  $t_1$  with high probability remain stable for  $\tau \ll \tau_{1,2}$  but dissolve for  $\tau \gg \tau_{1,2}$ , allowing their particles in the process to displace as  $\Delta(t_1 + \tau, t_1) \sim A \ln(\tau/\tau_{1,2} + 1)$ .<sup>47</sup> Thus, if reheating is forestalled until a time  $t_2 \gg \tau_2$ , *i.e.*, a time beyond

$$\tau_2 \sim B t_1^{\beta_2/\beta_1}, \quad (5)$$

with some microscopic constant  $B = t_0^{1-\beta_2/\beta_1}$ , memory will have been lost. In Fig. 4, we demonstrate this effect in the cluster model, evolving the system with the waiting time method over 15 decades.

## IV. Conclusions

In conclusion, we have demonstrated that the cluster model reproduces the macroscopic observable rejuvenation behavior of the structural glass studied in ref. 59. However, we would like to qualify some of their conclusions. For one, that their results fit well with predictions of mean-field theory should not necessarily be taken as evidence that all aspects of the theory apply to real systems. As our dramatically simplified model suggest, an elementary description of the requisite hierarchical landscape features<sup>38,67</sup> may exist. That mean-field theory also happens to provide the same elementary features does not imply that all aspects of that theory apply. Rejuvenation and memory by themselves are not even sufficient to imply glassy behavior.<sup>55</sup>

Furthermore, we disagree with the conclusion, based on the pdf of particle displacements,  $\mathcal{P}(\log \Delta r^2)$ , shown Fig. 7 in ref. 59, that “all particles are involved in the aging dynamics” which occurs “due to very collective particle motion involving the entire system...”. We observe that these pdf are each distributed around the respective plateau values of MSD in Fig. 3 and 4(a) of ref. 59, thus merely representing ordinary in-cage rattle. While this may appear as “featureless” and therefore homogeneous, it has been shown that the actual irreversible events that drive relaxation during aging are highly intermittent and localized,<sup>30</sup> and are likely hidden deep within the large- $\Delta r^2$  tail of those pdf. (Note, *e.g.*, the minute bump near  $\Delta r^2 \approx 10^0$  in Fig. 7 of ref. 59.) Ultimately, this heterogeneity is exactly what is captured by the break-up of clusters in our model, after coarse-graining out the in-cage rattle, as that rattle only rarely amounts to meaningful (record-sized, irreversible) displacements.<sup>32</sup>

## Conflicts of interest

There are no conflicts to declare.

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