Self-Organized Criticality Below The Glass Transition

Katharina Vollmayr-Lee\textsuperscript{1,2} and Elizabeth A. Baker\textsuperscript{1}

\textsuperscript{1}Department of Physics and Astronomy, Bucknell University, Lewisburg, Pennsylvania 17837, USA
\textsuperscript{2}Institut für Theoretische Physik, Universität Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany\textsuperscript{*}

(Dated: March 26, 2006)

We obtain evidence that the dynamics of glassy systems below the glass transition is characterized by self-organized criticality. Using molecular dynamics simulations of a model glass-former we identify clusters of cooperatively jumping particles. We find string-like clusters whose size is power-law distributed not only close to \( T_c \) but for all temperatures below \( T_c \), indicating self-organized criticality which we interpret as a freezing in of critical behavior.

PACS numbers: 64.70.Pf, 02.70.Ns, 61.43.Fs

Although the relaxation dynamics of glass-forming liquids has been studied for decades, there are still many unresolved questions [1]. Especially for the dynamics of the glass out of equilibrium it is still an open and hotly debated question what characterizes the relaxation. We present in this Letter work on the dynamics below the glass transition where we focus on cooperative motion. Cooperative rearranging regions have been studied mostly above the glass transition and are the basis of Adam-Gibbs theory [2]. Above the glass transition two kinds of cooperative motion have been identified: (i) string-like motion [3–5] where of the order of ten particles move along a Conga-line and where each particle is significantly more mobile than an average particle, and (ii) very cooperative motion where of the order of 40 particles participate and where each particle undergoes only a small displacement [6].

To study cooperative motion below the glass transition we use molecular dynamics simulations, which have the advantage of providing us with the microscopic information of every particle’s position at all times. Using these particles trajectories we first search each simulation run for jump events where a particle jumps out of its cage of neighbors. Then we identify clusters of cooperatively jumping particles, i.e., jump events which are correlated in space and time. We find that the cluster size distribution follows a power law independent of details of the cluster definition. Furthermore, we find string-like clusters as they have been found above the glass transition.

A similar cluster definition for the system under study but at a temperature slightly above the glass transition has also revealed a power law distribution [3]. Such distributions are a signature of criticality, such as the cluster size distribution in percolation theory at the critical point [7]. However, contrary to these simulations and percolation theory, we find a power law not only close to a critical point but for all temperatures below \( T_c \) that we have investigated (see Fig. 1). We thus find a type of self-organized criticality.

Our data are consistent with the following scenario: a glass cooled down to \( T_c \) develops critical behavior, and then upon further cooling the criticality remains frozen in. Mode-coupling theory for glasses predicts the development of critical fluctuations [8] when \( T_c \) is approached from above and a recent extension of mode-coupling theory to temperatures below the transition has predicted such a freezing in of critical behavior [9]. The signature of criticality in these theories is the power law of a two-time correlation function [9–11]. Our data, while consistent with the mode-coupling freezing in scenario, find criticality in a spatial structure rather than a relaxation exponent. We believe this represents the first direct observation in glasses of self-organized criticality, that is, criticality for all temperatures below \( T_c \).

Our system is a well-studied binary Lennard-Jones (LJ) mixture of 800 A and 200 B particles. We refer the reader for details of the model to [12] and for de-

*Electronic address: kvollmay@bucknell.edu

FIG. 1: Distribution \( P(s) \) of cluster sizes of simultaneously jumping particles for temperatures \( T = 0.15 \) – 0.43. The inset shows \( P(s) \) at \( T = 0.43 \) where the jump events have been divided into five equal time windows. The black solid line represents the first window.
tails of the molecular dynamics simulations to [13]. Previous simulations have shown that this system exhibits the main features of glass-forming liquids and is thus a good simple model for glass-formers [12]. For present-day computer simulations the system falls out of equilibrium in the vicinity of $T_g = 0.435$ (in reduced LJ units) [12]. Whereas Donati et al. [3] studied cooperative motion of this system above the glass transition, we study here the same system but below the glass transition at 10 temperatures ranging from 0.15 to 0.43. We use 10 independent, well-equilibrated configurations at $T = 0.5$ and then instantly quench the system to the desired temperature, e.g. $T = 0.15$. After an (NVT) run of 2000 time units we then run the (NVE) production run for 10³ time units.

For the definition of jump events we use the trajectory $\mathbf{r}_n(t)$ of each particle $n$ and take time averages over 800 time units to obtain its thermal fluctuation $\sigma_n$ and average positions $\langle \mathbf{r}_n \rangle(t)$ at times $t_l = 800(l - 0.5)$ where $l = 1, 2, \ldots, 125$. We define a particle $n$ to undergo a jump if its change in average position $\Delta \mathbf{r}_n = |\mathbf{r}_n(t_l) - \langle \mathbf{r}_n \rangle(t_{l-4})|$ satisfies $\Delta \mathbf{r}_n > \sqrt{20} \sigma_n$ [14]. We thus identify for the whole simulation run all jump events $(n, i, \langle \mathbf{r}_n \rangle_i, l_f, \langle \mathbf{r}_n \rangle_f)$ of jumping particles $n$, jumping from average position $\langle \mathbf{r}_n \rangle_i$ at time $t_i$, the time associated with bin $i$, to average position $\langle \mathbf{r}_n \rangle_f$ at time $t_f$ [15].

To address the question of cooperative motion we investigate how these single particle jump events are correlated in time and space. To identify correlations in time, we group the jump events according to the bin index $l$. We thus obtain $N_l$ simultaneously jumping particles for each time bin $l$. To investigate how these $N_l$ particles are spatially correlated, we identify clusters where particles $n$ and $m$ are defined to be neighbors (and therefore members of the same cluster) if their distance $|\mathbf{r}_n - \mathbf{r}_m|$ is smaller than the position of the first minimum $r_{\text{min}}$ of the corresponding radial pair distribution function of the complete system ($r_{\text{min}} = 1.4$ for AA, 1.2 for AB and 1.07 for BB independent of temperature). This analysis gives us for each time $K_l \geq 0$ distinct clusters. The clusters are numbered by $k = 1, 2, \ldots, K_l$ and we denote by $N_{l,k}$ the set of particle labels composing the $k$th cluster in time bin $l$ with $N_{l,k}$ particles (i.e. $\sum_{k=1}^{K_l} N_{l,k} = N_l$). We now look at the size distribution $P(s)$ of all clusters $N_{l,k}$, with $s$ being the number of cluster members, i.e.

$$P(s) = \frac{\sum_k K_l \delta(s, N_{l,k})}{\sum_l K_l} \quad (1)$$

where $\delta(x, y)$ is the Kronecker delta function.

In Fig. 1 we show a log-log plot of $P(s)$ for temperatures $T = 0.15 - 0.43$. We observe essentially a straight line, i.e. a power law $P(s) \sim s^{-\tau}$, indicating scale invariance. The exponent for different temperatures ranges from $\tau = 2.2 - 2.7$ with the trend of $\tau$ increasing with temperature. The average is $\tau = 2.50 \pm 0.05$. A power law of $P(s)$ has also been found in simulations of the same binary Lennard-Jones system slightly above but close to $T_c$ with $\tau = 1.86$ [3]. In percolation theory the size distribution $n_s$ (where $P(s) = n_s / \sum_s n_s$) follows also a power law where the mean-field exponent $\tau = 2.5$ and in three dimensions $\tau = 2.2$ [7]. However, in these simulations and in percolation theory the power law occurs only at $T_c$. In contrast, we find a power law for all temperatures (see Fig. 1), i.e. we find scaling invariance for the whole temperature range below $T_c$. As mentioned above, this is consistent with the scenario of critical behavior being frozen in, and remaining for all temperatures below $T_c$.

This scale-invariance independent of a control parameter is usually found in systems with self-organized criticality [16] and is, to our knowledge, a new phenomenon for the cluster size distribution of structural glass formers. The occurrence of self-organized criticality is usually associated with being out of equilibrium and having widely separated time scales. Our system is consistent with these requirements: single particle jumps take of the order of 10 time units, the time between successive jumps is of the order of 30000 [13], and the equilibrium relaxation time is significantly longer than the simulation run [12].

Due to the importance of implications for relaxation dynamics below the glass transition, the question arises if our results for the cluster size distribution are specific to details of the analysis. We therefore investigate how robust $P(s)$ is with respect to different definitions. Since the system is out of equilibrium, we first ask the question if the jump dynamics changes during the simulation run. We therefore divide the whole simulation run into five time windows of equal size and determine $P(s)$ with Eq.(1) by restricting the sum over $l$ accordingly. The inset of Fig. 1 shows the resulting $P(s)$ for $T = 0.43$. We find that in the first time window the most collective processes occur: up to approximately 250 particles jump simultaneously and spatially correlated. The distribution $P(s)$ however seems to follow a power law with $\tau = 2.5 \pm 0.1$ not only independent of temperature but also independent of the time window (i.e. waiting time). We find the same waiting time independence also for all other temperatures.

To further check the sensitivity on details of the power law of $P(s)$ we next modify the definition of a cluster. Whereas usually cluster connections are defined in space (and therefore avalanche-like correlations are tested in space), we generalize now the definition of a cluster by treating space and time similarly (and thus allowing for avalanche-like correlations in time also). Instead of requiring as before that two jump events $\alpha$ and $\beta$ occur simultaneously ($l_{i,\alpha} = l_{i,\beta}$) we define extended clusters by allowing two jump events to occur at neighboring time bins (i.e. $|\Delta| \leq 1$). We show in Fig. 2 results for two different definitions of extended clusters (I & II). The difference between these definitions is due to the usage of time and position before or after the jump. We define two jump events $\{n^\alpha, l_{i,\alpha}, \langle \mathbf{r}_n \rangle_i^\alpha, l_f^\alpha, \langle \mathbf{r}_n \rangle_f^\alpha\}$ and $\{m^\beta, l_{i,\beta}, \langle \mathbf{r}_m \rangle_i^\beta, l_f^\beta, \langle \mathbf{r}_m \rangle_f^\beta\}$ to be connected if
def. I: $|l^2_i - l^2_f| \leq 1$ and $|\langle r_n \rangle_i^2 - \langle r_n \rangle_f^2| \leq r_{\text{min}}$

def. II: $|l^2_i - l^2_f| \leq 1$ and $|\langle r_n \rangle_i^2 - \langle r_n \rangle_f^2| \leq r_{\text{min}}$ or $|\langle r_n \rangle_i^2 - \langle r_n \rangle_f^2| \leq r_{\text{min}}$

As before for simultaneously jumping particles, definitions I and II result in a cluster size distribution which follows a power law, however, with a smaller exponent of $\tau = 2.13 \pm 0.04$ and $\tau = 2.22 \pm 0.04$ for clusters I and II respectively. As shown in Fig. 2 for $T = 0.30$ and 0.43 we find again a power law for all temperatures (similar results are obtained for other temperatures).

![Fig. 2](image)

**FIG. 2:** Distribution of cluster size for extended clusters I & II for temperatures $T = 0.30$ and 0.43.

To further investigate these clusters we next characterize their geometric shape directly via coordination numbers (instead of displacement-displacement correlations as in [3]). We determine for each cluster $N_{l,k}$ the average coordination number

$$z_{l,k} = \frac{1}{N_{l,k}} \sum_{n \in N_{l,k}} z_n$$

(2)

where $z_n$ is the number of neighboring particles $m \in N_{l,k}$ of particle $n$. Fig. 3 shows the average

$$\langle z(s) \rangle = \sum_l \sum_{k=1}^{K_l} \delta(s, N_{l,k}) z_{l,k} / \sum_l \sum_{k=1}^{K_l} \delta(s, N_{l,k})$$

(3)

as a function of $s$. We observe no temperature dependence of $\langle z(s) \rangle$ and therefore an additional average over simulation runs at different temperatures has been included in Fig. 3. The comparison with an ideal string and the most compact cluster (sphere) indicates that both the clusters of simultaneously jumping particles as well as the extended clusters are string-like. This is similar to the results of cooperative motion above the glass transition [3–5] and below the glass transition [17].

Our results are consistent with the following scenario: above the critical temperature $T_c$ string-like clusters are found. Close to $T_c$ the distribution of cluster sizes follows a power law. Below the glass transition this critical behavior gets frozen in. Independent of details of the cluster definition and independent of waiting time, we find string-like clusters with a cluster size distribution which follows a power law for all investigated temperatures. We expect this self-organized criticality to occur also for other glasses out of equilibrium and leave the test thereof for future work.

KVL thanks the Institute of Theoretical Physics, University Göttingen, for hospitality and financial support. EAB gratefully acknowledges support from NSF Grant No. REU-0097424. The authors thank J. Horbach, W. Kob and K. Binder for comments on an earlier version of this manuscript and also A. Latz, and A. Zippelius for helpful discussions.

[1] K. Binder and W. Kob, “Glassy Materials and Disordered Solids: An Introduction to Their Statistical Mechanics,” Word Scientific, Singapore 2005.
[2] G. Adam and J. H. Gibbs, J. Chem. Phys. 43, 139 (1965).
[3] C. Donati, J. F. Douglas, W. Kob, S. J. Plimpton, P. H. Poole, and S. C. Glotzer, Phys. Rev. Lett. 80, 2338 (1998); C. Donati, S. C. Glotzer, P. H. Poole, W. Kob, and S. J. Plimpton, Phys. Rev. E 60, 3107 (1999); M. Vogel, B. Doliva, A. Heuer, and S. C. Glotzer, J. Chem. Phys. 120, 4404 (2004).
[4] E. R. Wees, J. C. Crocker, A. C. Levitt, A. Schofield and D. A. Weitz, Science 287, 627 (2000).
H. Miyagawa, Y. Hiwatari, B. Bernu, and J. Hansen. J. Chem. Phys. 88, 3879 (1988); G. Wahnström, Phys. Rev. A 44, 3752 (1991); D. N. Perera and P. Harrowell, J. Chem. Phys. 111, 5441 (1999); Y. Gebremichael, M. Vogel, and S. C. Glotzer, Mol. Simul. 30, 281 (2004); Y. Gebremichael, M. Vogel, and S. C. Glotzer, J. Chem. Phys. 120, 4415 (2004) and references therein; V. Téboul, A. Monteil, L. C. Ai, A. Kerrache, and S. Maabou, Eur. Phys. J. B 40, 49 (2004); M. Vogel and S. C. Glotzer, Phys. Rev. Lett. 92, 255901 (2004);

G. A. Appignanesi, J. A. Rodriguez Fris, R. A. Montani, and W. Kob, e-print cond-mat/0506577 (2005);

D. Stauffer, Phys. Rep. 54, 1 (1979).

W. Götze and L. Sjögren, Rep. Prog. Phys. 55, 241 (1992).

A. Latz, preprint cond-mat/0106086.

A. Latz, J. Phys.: Condens. Matter 12, 6353 (2000).

J.-P. Bouchaud, L. Cugliandolo, J. Kurchan, M. Mézard, Physica A 226, 243 (1996).

W. Kob and H. C. Andersen, Phys. Rev. Lett. 73, 1376 (1994); Phys. Rev. E 51, 4626 (1995); Phys. Rev. E 52, 4134 (1995).

K. Vollmayr-Lee, J. Chem. Phys. 121, 4781 (2004).

As reported in [13] $\sigma_n$ and $\Delta r_n$ are of the order of 0.1 and 0.7 respectively.

For more details of the jump definition see [13]. Therein we distinguish irreversible and reversible jumps. In this Letter we include for all presented results both irreversible and reversible jumps.

P. Bak, C. Tang, and K. Wiesenfeld, Phys. Rev. Lett. 59, 381 (1987).

C. Oligschleger and H. R. Schober, Phys. Rev. B 59, 811 (1999); H. Teichler, J. Non-Cryst. Solids 312-314, 533 (2002); R. E. Courtland and E. R. Weeks, J. Phys.: Condens. Matter 15, S359 (2003); M. Kluge and H. R. Schober, Phys. Rev. B 70, 224209 (2004); H. Teichler, Phys. Rev. E 71, 031505 (2005).