EUROPHYSICS LETTERS 15 December 2006

Europhys. Lett., **76** (6), pp. 1130–1136 (2006)
DOI: 10.1209/epl/i2006-10405-1

Self-organized criticality below the glass transition

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received 27 April 2006; accepted in final form 31 October 2006
published online 24 November 2006

PACS. 64.70.Pf – Glass transitions.
PACS. 02.70.Ns – Molecular dynamics and particle methods.
PACS. 61.43.Fs – Glasses.

Abstract. – 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.

Introduction. – 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 [3–7] 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 [8].

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 [4] and also for other systems such as SiO_2 and polymer melts [9–11] have also revealed a power law distribution but at a temperature slightly above the glass transition. Such distributions are a signature of criticality,

such as the cluster size distribution in percolation theory at the critical point [12]. 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. 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 [13] 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 [14]. The signature of criticality in these theories is the power law of a two-time correlation function [14–16]. 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.

Model. – 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 [17] and for details of the molecular dynamics simulations to [18]. 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 [17]. The mode-coupling critical temperature is $T_c = 0.435$ (in reduced LJ units) [17]. Whereas Donati *et al.* [3,4] 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 $2 \cdot 10^4$ time units.

Jump definition. – For the definition of jump events we use the trajectory $\boldsymbol{r}_n(t)$ of each particle n and take time averages over 800 time units to obtain its thermal fluctuation σ_n and average positions $\overline{\boldsymbol{r}_n}(t_l)$ at times $t_l = 800(l - 0.5)$ where $l = 1, 2, \ldots, 25$. We define a particle n to undergo a jump if its change in average position $\Delta\overline{r_n} = |\overline{\boldsymbol{r}_n}(t_l) - \overline{\boldsymbol{r}_n}(t_{l-4})|$ satisfies $\Delta\overline{r_n} > \sqrt{20}\,\sigma_n$ [19]. We thus identify for the whole simulation run all jump events $\{n, l_i, \langle \boldsymbol{r}_n \rangle_i, l_f, \langle \boldsymbol{r}_n \rangle_f\}$ of jumping particles n, jumping from average position $\langle \boldsymbol{r}_n \rangle_i$ at time t_{l_i}, the time associated with bin l_i, to average position $\langle \boldsymbol{r}_n \rangle_f$ at time t_{l_f} [20].

Cooperative motion. – 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_i. 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 $|\langle \boldsymbol{r}_n \rangle_i - \langle \boldsymbol{r}_m \rangle_i|$ is smaller than the position of the first minimum $r_{\min}$ of the corresponding radial pair distribution function of the complete system ($r_{\min} = 1.4$ for AA, 1.2 for AB and 1.07 for BB independent of temperature) [21]. 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 $\mathcal{N}_{l,k}$ 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 $\mathcal{N}_{l,k}$, with s being the number of cluster members, *i.e.*

$$P(s) = \sum_l \sum_{k=1}^{K_l} \delta(s, N_{l,k}) \Big/ \sum_l K_l, \tag{1}$$

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

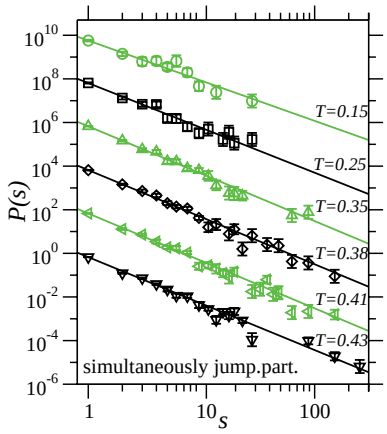

Fig. 1 – Distribution $P(s)$ of cluster sizes of simultaneously jumping particles for temperatures $T = 0.15$–0.43. For clarity, the distributions have been shifted by successive factors of 100. Linear fits to $\log P(\log s)$ are included. The waiting time defined in the text, is $t_{\rm wait} = 0.2 \cdot 10^4$.

In fig. 1 we show a log-log plot of $P(s)$ for temperatures $T = 0.15$–0.43. For clarity only a subset of all temperatures is shown and the curves have been shifted by a factor of 100^n ($n = 0, 1, 2, \ldots$). Error bars have been determined via the ten independent simulation runs. Weighted linear fits to $\log P(\log s)$ are included. We observe essentially a straight line, *i.e.* a power law $P(s) \sim s^{-\tau}$, indicating scale invariance. In fig. 2 we show the exponents τ as a function of temperature. (Corresponding to fig. 1 are the circles and the bold straight line which serves as guide to the eye.) Error bars have been obtained via the slopes of the 10 independent weighted linear fits to $\log P(\log s)$. We find that τ is increasing with increasing temperature, and approaching approximately the value $\tau \approx 1.86$, which has been found for the same binary Lennard-Jones system slightly above but close to $T_{\rm c}$ [4]. 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 $\tau = 2.2$ [12]. However, in these simulations and in percolation theory the power law occurs only *at* $T_{\rm c}$. In contrast, we find a power law for all temperatures (see fig. 1, and similarly for not included temperatures), *i.e.* we find scaling invariance

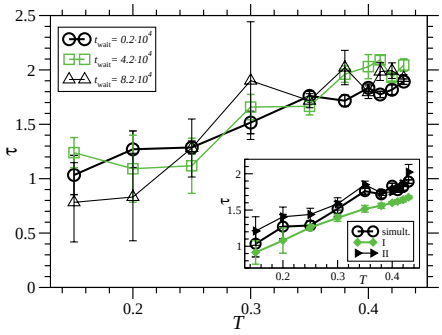

Fig. 2 – Exponent τ as a function of temperature T for simultaneously jumping particles and for varying waiting time $t_{\rm wait}$. The inset shows τ for $t_{\rm wait} = 0.2 \cdot 10^4$ for different cluster definitions given in the text.

for the *whole* temperature range below T_c [22]. 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 [23] 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 [18], and the equilibrium relaxation time is significantly longer than the simulation run [17].

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. To 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 too). Instead of requiring as before that two jump events α and β 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.* $|\Delta l| \leq 1$). We show in fig. 3 results for two different definitions of extended clusters (I and 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 $\{n^\alpha, l_i^\alpha, \langle \boldsymbol{r}_n \rangle_i^\alpha, l_f^\alpha, \langle \boldsymbol{r}_n \rangle_f^\alpha\}$ and $\{m^\beta, l_i^\beta, \langle \boldsymbol{r}_m \rangle_i^\beta, l_f^\beta, \langle \boldsymbol{r}_m \rangle_f^\beta\}$ to be connected if

– def. I: $|l_i^\alpha - l_i^\beta| \leq 1$ and $|\langle \boldsymbol{r}_n \rangle_i^\alpha - \langle \boldsymbol{r}_m \rangle_i^\beta| \leq r_{\min}$

– def. II: $\big(|l_i^\alpha - l_f^\beta| \leq 1$ and $|\langle \boldsymbol{r}_n \rangle_i^\alpha - \langle \boldsymbol{r}_m \rangle_f^\beta| \leq r_{\min}\big)$ or

$\big(|l_f^\alpha - l_i^\beta| \leq 1$ and $|\langle \boldsymbol{r}_n \rangle_f^\alpha - \langle \boldsymbol{r}_m \rangle_i^\beta| \leq r_{\min}\big)$.

As before for simultaneously jumping particles, definitions I and II result in a cluster size distribution which follows a power law. As shown in fig. 3 for $T = 0.30$ and 0.42 we find again a power law for all temperatures (similar results are obtained for other temperatures). This power law is different than previous results slightly above the glass transition [3, 5, 9, 11] which find an exponential string length distribution, where a string is defined similar to our

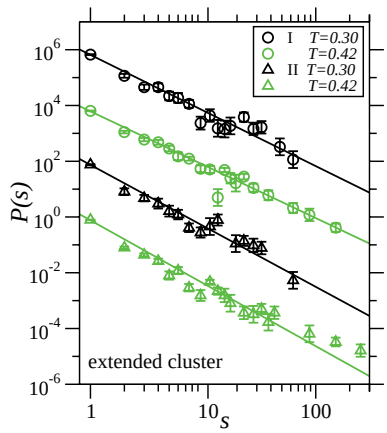

Fig. 3 – Distribution of cluster size for extended clusters I and II for temperatures $T = 0.30$ and 0.42. The waiting time defined in the text, is $t_{\text{wait}} = 0.2 \cdot 10^4$.

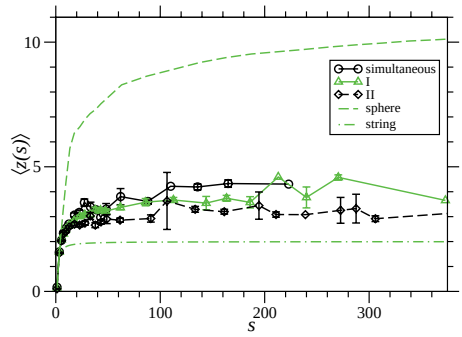

Fig. 4 – Average coordination number z within a cluster as a function of cluster size s.

definition II [24]. The exponents τ for the extended cluster definitions I and II are of the same order and show the same temperature dependence as for simultaneously jumping particles (see inset of fig. 2).

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

$$z_{l,k} = \frac{1}{N_{l,k}} \sum_{n \in \mathcal{N}_{l,k}} z_n, \tag{2}$$

where z_n is the number of neighboring particles $m \in \mathcal{N}_{l,k}$ of particle n [21]. Figure 4 shows the average

$$\langle z(s) \rangle = \sum_l \sum_{k=1}^{K_l} \delta(s, N_{l,k}) z_{l,k} \Big/ \sum_l \sum_{k=1}^{K_l} \delta(s, N_{l,k}) \tag{3}$$

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. 4. The

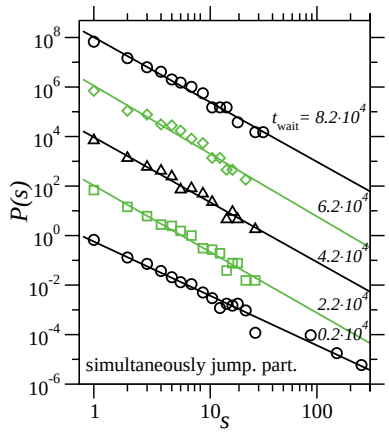

Fig. 5 – Distribution of cluster size for simultaneously jumping particles at $T = 0.43$ and varying waiting time t_{wait}.

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 [3–7] and below the glass transition [25].

Since the system is out of equilibrium, we next ask the question if the above results change if we increase the waiting time $t_{\rm wait}$ after the temperature quench [26]. We therefore run the (NVE) production run for 10^5 instead of $2 \cdot 10^4$ time units and analyze five time windows of equal size by adjusting the sum over l in eqs. (1) and (3) accordingly. Counting the initial (NVT) production run, we thus study waiting times $t_{\rm wait} = 0.2 \cdot 10^4, 2.2 \cdot 10^4, \ldots, 8.2 \cdot 10^4$. Figure 5 shows the resulting $P(s)$ for simultaneously jumping particles at $T = 0.43$ [27]. 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 not only for all temperatures below $T_{\rm c}$ but also for all waiting times. As shown in fig. 2 the exponents seem to be independent of the waiting time, indicating possibly $t_{\rm wait}$-independent distributions $P(s)$. We obtain similar results for simultaneously jumping particles at other temperatures and also for the extended cluster definitions I and II. Also the average coordination number $\langle z(s) \rangle$ seems independent of $t_{\rm wait}$.

Conclusions. – Our results are consistent with the following scenario: above the critical temperature $T_{\rm c}$ string-like clusters are found. Close to $T_{\rm c}$ 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. In the simulation the finite number of particles sets an upper limit on the cluster size s and thus on the range over which this power law is observed. We therefore plan further investigations with a system of significantly more particles. The experiments of Weeks *et al.* [6] result in a power law of $P(s)$ for one $\phi > \phi_{\rm c}$ (which corresponds to $T < T_{\rm c}$). The power law for a glass out of equilibrium seems to be robust, since Weeks *et al.* used definitions of mobile particles (jumping particles) and clusters that differ from the definitions presented in this paper. We therefore expect this self-organized criticality to occur also for other glasses out of equilibrium and therefore suggest further investigations of $P(s)$ for glasses out of equilibrium.

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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.

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