Avalanches and Dynamical Correlations in supercooled liquids

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We identify the pattern of microscopic dynamical relaxation for a two dimensional glass forming liquid. On short timescales, bursts of irreversible particle motion, called cage jumps, aggregate into clusters. On larger time scales, clusters aggregate both spatially and temporally into avalanches. This propagation of mobility, or dynamic facilitation, takes place along the soft regions of the systems, which have been identified by computing isoconfigurational Debye-Waller maps. Our results characterize the way in which dynamical heterogeneity evolves in moderately supercooled liquids and reveal that it is astonishingly similar to the one found for dense glassy granular media.

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Identifying the physical mechanisms responsible for the slowing down of the dynamics of supercooled liquids is still an open problem despite several decades of intense research. While traditional descriptions of glassy systems have mainly focused on energy landscape concepts[1] and spatially averaged quantities, recent work has centered on the real space properties reflected in the dramatically heterogeneous dynamics shared by nearly all glass-forming materials. Concomitantly, investigations of the behavior of dense driven granular media have uncovered tantalizing similarities with the dynamics of supercooled liquids[2, 3, 4, 5] and provided new inspirations for research on the glass transition. Among the most notable findings related to the real space dynamical properties in supercooled liquids as well as granular media is the evidence that dynamic facilitation [6, 7] and sizable dynamic correlations [8] play an important role. This is supported by the detailed analysis of microscopic dynamics which has identified correlated particle motion in clusters, strings and other motifs. [9, 10, 11]. A natural question [12] related to these findings is what, if any, structural features are correlated with the heterogeneity noted in the real space dynamics. Important progress in this direction has been obtained [13], through the introduction of the quantitative notion of "propensity", and then later in [14, 15, 16], where it has been shown that irreversible motion is correlated with the spatial characteristics of soft modes.

In this paper we address the following three questions: First, do supercooled liquids exhibit the same hierarchical organization of dynamics (i.e. cage escapes within clusters within avalanches) as recently reported in granular materials [10, 11]? Second, to what degree are these different scales of collective motion determined by the underlying structure? And reciprocally, to what extend is the evolution of the structure related to the relaxation events within a given realization of the dynamics?

We shall address these questions by performing computer simulations on a new two-dimensional model of glass-forming liquid and applying the cluster analysis developed in [10]. This new model is distinguished from previous 2D mixtures [17] in that supercooled liquid dynamics may be simulated without the formation of palpable crystalline micro-domains. Our main results are that the glassy dynamics of dense driven granular systems [10] and supercooled liquids turn out to be astonishingly similar even at the microscopic level. *This is remarkable given the fact that granular systems are driven non-equilibrium systems with dissipative contact interactions while supercooled liquids are equilibrium conservative systems.* Quasi-instantaneous clusters of nearby relaxing particles are typically followed by adjacent clusters showing how long term dynamical correlations emerge. This dynamic facilitation leads to the formation of finite size and finite duration avalanches located on the "soft" regions of the configuration as probed by the isoconfigurational average of the Debye-Waller factor. Finally the clusters of relaxing particles induce non-local reorganisation of the structure as probed by the dynamics itself of the Debye-Waller factor.

As a model for a supercooled liquid we focus on a $2D$ non-additive binary mixture of $N = 5,760$ particles enclosed in a square box with periodic boundary conditions, interacting via purely repulsive potentials of the form $u_{ab}(r) = \varepsilon(\sigma_{ab}/r)^{12}$. The mole fraction of the smaller particles is taken to be $x_1 = 0.3167$. All units are reduced so that $\sigma_{11} = \varepsilon = m = 1.0$, m being the mass of both types of particle. We use non-additive potentials, namely $\sigma_{12} = 1.1 \times \sigma_{11}$ and $\sigma_{22} = 1.4 \times \sigma_{11}$ to avoid the formation of crystalline domains. The temperature dependence of the structural and dynamical properties of this model were characterized in [18]. Molecular dynamics simulations were carried out at constant NVT (T=0.4) using the Nose-Poincare Hamiltonian [19] after equilibration at constant NPT as described in [14]. All time units are scaled in such a way that the structural re-

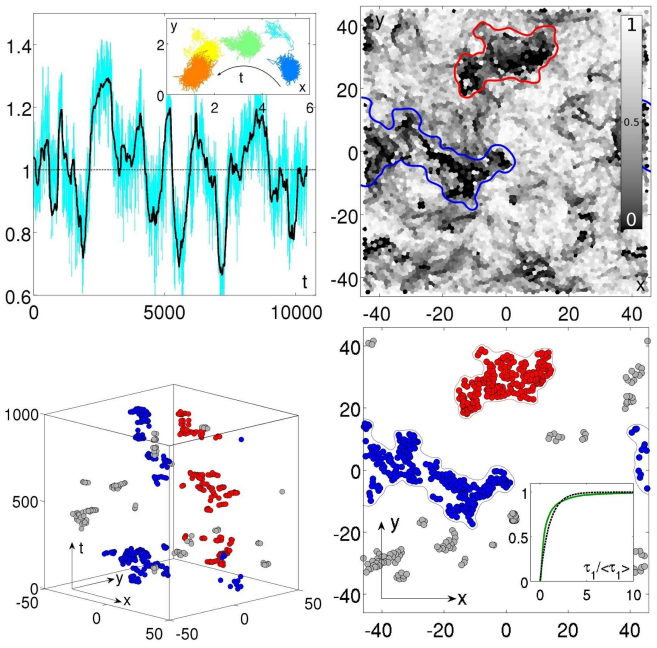

FIG. 1: (color online) Cooperative cage jumps form large decorrelation patterns. Top left: Comparison between the relative averaged relaxation $Q_t(a^*,\tau^*)/\langle Q_t\rangle_t$ (cyan) and the relative percentage $P_t(\tau*)/\langle P_t\rangle_t$ of particles that have not jumped between t_0 and $t_0+\tau^*$ (black). *Inset* Trajectory of a single particle over $14\tau_\alpha$. Color changes when the particle jumps. Top right: Map of $Q_t(\tau^*)$. Bottom left: Spatio-temporal view of the cage jumps between t and $t+\tau^*$. The jumps corresponding to two arbitrarily chosen avalanches are painted in blue and red. Bottom right: Map of the cage jumps occurring between t and $t+\tau^*$. Note that the blue and red avalanches lie in distinct regions of space. *Inset* Cumulative Pdf of the reduced time lag between adjacent clusters τ_1 (green), compared to the corresponding curve for a random distribution of clusters (black).

laxation time τ_α, defined as the time required for the self intermediate scattering function to decay of $1/2$, equals 10^3. The typical collision time is 0.12 in these units.

We choose as a measure of the local mobility (or relaxation) of a particle p:

$$Q_{p,t}(a,\tau) = \exp\left(-\frac{||\Delta\vec{r}_p(t,t+\tau)||^2}{2a^2}\right), \qquad (1)$$

where $\Delta\vec{r}_p(t,t+\tau)$ is the displacement of the particle p between t and $t+\tau$ and a is the length scale over which the motion is probed. A global measure of the dynamics is provided by the correlation function, $Q_t(a,\tau) = \frac{1}{N}\sum_p Q_{p,t}(a,\tau)$, and its fluctuations $\chi_4(a,\tau) = N\mathrm{Var}\left(Q_t(a,\tau)\right)$. As in [20], we focus on the values of a and τ corresponding to maximal dynamic heterogeneity, i.e. highest value of $\chi_4(a,\tau)$ (see [20] for details). This leads to $a^*=0.29$ and $\tau^*=1078$. Note that the latter is very close to the relaxation time $\tau_\alpha=1000$.

In order to analyze the microscopic dynamics and study possible connections with the dynamics of dense driven granular media, we follow the same procedure as in [10]. This allows one to separate the dynamics along a given trajectory into periods of inefficient vibrational motion separated by relaxation events also called cage jumps. (see inset of the top left panel of fig. 1). One has to bear in mind that a particle undergoing a cage jump does not necessarily change neighbors. In the top left panel of fig. 1 we compare the relative values of $Q_t(a^*,\tau^*)$ to those of $P_t(\tau^*)$, which is the percentage of particles that have not jumped during the time τ^*. The two curves track each other, showing that cage jumps provides a powerful coarse grained description of the dynamics. In addition, we also show that the cage jumps are exactly located in the areas where the decorrelation is maximal (compare fig. 1 right top and bottom). We repeat the same spatio-temporal analysis performed for the two dimensional granular media studied in [10]. The outcome is remarkably similar. First, cage jumps aggregate into clusters, which are formed by cage jumps adjacent in space (as measured by the neighboring particles) and time (separated by less than $\tau_{th}=28$, which is twice the precision of the cage detection algorithm). The size of these clusters are largely distributed with an average value of 7.6 cage jumps per cluster. Second, clusters aggregate into avalanches in which the first cluster triggers the appearance of successive clusters nearby shortly after, see fig. 1 bottom left. This is clearly demonstrated as in [10] by focusing on the cumulative *Pdf* of the lag times τ_1 separating each cluster from the nearest adjacent one, normalized by its average value $\langle\tau_1\rangle$. See the inset of fig. 1,bottom right, where this *Pdf* is compared to the equivalent distribution for randomly distributed clusters in space and time. One can see a clear excess of both small and large lags: the *Pdf* can be extremely well fit by the union of two data sets corresponding to Poissonian processes with two different timescales $\tau_S=240$ and, $\tau_L=1746$. The short time scale corresponds to the existence of a correlation among adjacent clusters. The large one is related to the average time spent in a cage. This leads to a very peculiar type of dynamical correlation, which in the literature is often called dynamical facilitation [6, 7]: local relaxations are followed closely in space and in time by other local relaxations. The concatenation of these events leads to the dynamical heterogeneity observed on the timescale τ_α. However, at least for the degree of supercooling considered here, we find that facilitation is not conserved in the following sense: avalanches are well separated, indicating that there are relaxation events, which cannot be explained by the facilitation mechanism.

It is interesting to compare the actual values of these

	a^*	ξ_4	τ_α	τ^*	τ_S	τ_L
Supercooled Liquid	0.29	2.9	1000	1078	240	1746
Dense Granular Media	0.12	3.1	1000	915	155	1384

TABLE I: Comparison of length and time scales normalized so that $\tau_\alpha=1000$. See definitions in the text.

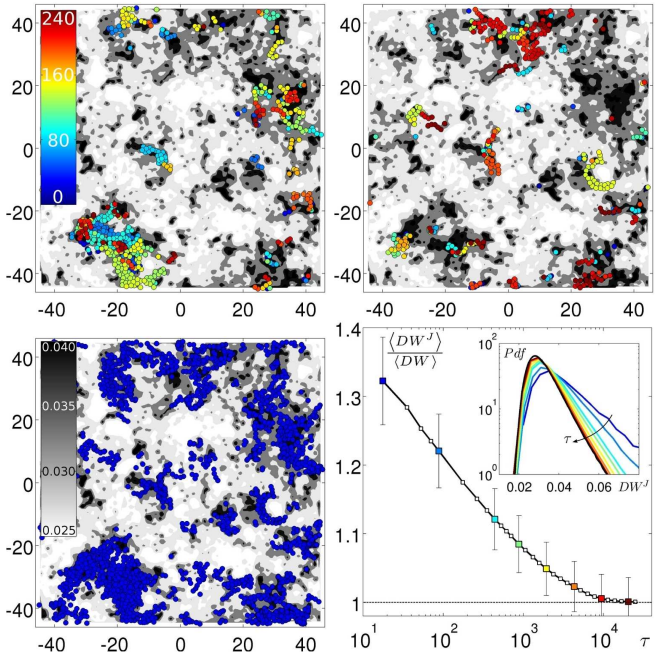

FIG. 2: (color online) Top: Cage jumps occuring between t (blue) and $t + \tau_S$ (red) for two different isoconfigurational trajectories, on top of a DW factor map computed at time t (in grey). Bottom left: Cage jumps occurring in 6 isoconfigurational trajectories between t and $t + \tau_S$ (in blue) tile the high DW regions. Colorbar indicates the DW values in levels of grey. Bottom right: Average $\langle DW^J \rangle$ over the particles having jumped between t and $t + \tau$, divided by the average $\langle DW \rangle$ over all particles, as a function of the lag time τ. *Inset* Pdf of DW^J for the particles jumping in $[t; t + \tau]$ for several values of τ. The black curve is the Pdf for all particles.

parameters to those of the granular system investigated previously. This comparison is performed in Table I, where we also report the value of the dynamical correlation length ξ_4, obtained from the spatial range of the dynamical correlator G_4(whose integral is equal to χ_4), see e.g. [21]. The dynamics are strikingly similar, a non-trivial result given the difference between an equilibrated thermal liquid and a non-equilibrioum steady state of vibrated grains. One difference we find is that the average distance between avalanches is somewhat smaller in the liquid case than in the granular one: ~ 10 as compared to ~ 27. Recent results [11] obtained by changing the density of the granular sample show that our model of a supercooled liquid would compare with a granular system characterized by a slightly smaller density.

We shall now investigate whether one can find a property closely connected to the structure at time t, which would allow one to predict where clusters will appear in the future and even some aspects of avalanche evolution. On the basis of previous work [14, 15] a natural candidate for such a feature are the so-called soft modes. Here we will use another means of identifying the location of "soft" regions or modes by using the isoconfigurational Debye-Waller (DW) factor[14, 22]. Starting from the system configuration at time t, one computes the local Debye-Waller factor for particle i: $DW_i = \langle [\vec{r_i}(t) - \langle \vec{r_i} \rangle_{\delta t}]^2 \rangle_{\delta t, \mathcal{C}}$, where the average is over the isoconfiguration ensemble as well as over a short time interval δt which in this work is taken to be 25.

Starting from the same equilibrated configuration, we have run 6 isotrajectories and have obtained the cage jumps for all of them. Remarkably, all of the cage jumps occurring in the interval of time $[t, t + \tau_S]$ fall on top of high DW areas, see fig. 2-top. Note that $\tau_S \gg 25$, thus the correlation between the DW map at time t (a nearly instantaneous structural quantity), and the dynamics taking place at longer times, is non-trivial.

Comparing the two top panels of Fig 2, we find that different isoconfigurational trajectories lead to cage jumps that take place at different times and in different regions although they are always located on top of high DW areas. This means that although clusters are very likely to be in soft regions, when and where they exactly appear is a stochastic event. The two top panels of Fig 2 strongly imply that a significant part of the avalanche structure of facilitated motion, and not just the initial cluster in an avalanche, occurs on top of the real-space geometric structure encoded in the soft mode map. Remarkably, we find that merging all cage jumps that occur in the interval of time τ_S in the 6 isoconfigurational trajectories cover nearly all the high DW areas, as shown in fig. 2 (bottom left). A similar comparison with localized low frequency normal modes, along the lines of [14], shows less, but still significant, correlation. We interpret this as a signature of anharmonic effects appearing in the vibrational structure of our model of a supercooled liquid. Indeed, it is likely that there are several potential energy minima in the basin in which the liquid is confined at short times. The local DWs allow one to overcome this difficulty and still provide a measure of local softness. The above results are in agreement with the previous conclusion of Berthier and Jack [23], who found that structural properties are better predictors of dynamics on large as opposed to short length scales.

In order to present a more quantitative proof of the correlation between DWs and cage jumps, we have computed the DW at time t averaged only over particles that jump between t and $t + \tau$ as a function of the lag time τ. This quantity, normalized with respect to $\langle DW(t) \rangle_t$ for all particles, is shown in fig. 2 (bottom right). We find that at short times the average DW for the jumping particles is substantially higher than the DW averaged over all particles. This correlation disappears for larger times comparable to times over which the DW maps decorrelate, which we find to be roughly of the order of $\tau_\alpha/3$.

A final issue worth investigating concerns the relation between cage jumps and DW map renewal. We find that decorrelation is a distinctly non-local process. More precisely, we have discovered that a cage jump at time t correlates with changes of the DWs that happen shortly after and extend quite far away. This is demonstrated

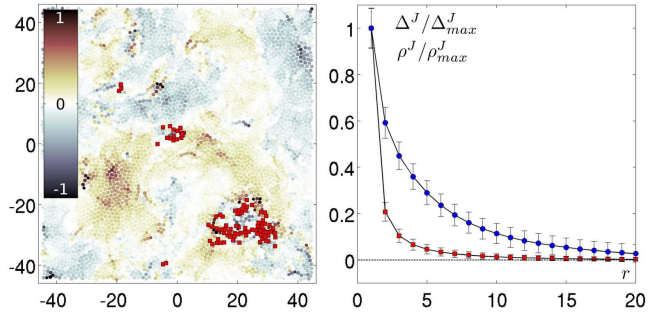

FIG. 3: (color online) Left: Cage jumps occurring in $\tau = 17$ on top of a map of the relative difference $(DW(t+\tau) - DW(t))/\langle DW\rangle$. Right: Normalized $\Delta^J(r) = \langle|\delta DW_i|\rangle^J_r - \langle|\delta DW_i|\rangle^J_\infty$ (blue circles) where $\langle|\delta DW_i|\rangle^J_r$ is the absolute difference of DW over $\tau = 17$ averaged over the particles in the disk of radius r around a given cage jump. The analogous quantity for the density of jumps $\rho^J(r) = \langle\delta_i\rangle^J_r - \langle\delta_i\rangle^J_\infty$ (red squares) where δ_i is 1 if particle i jumps between t and $t+\tau$ and 0 otherwise. Error bars are given by the standard deviation.

visually in fig. 3(left). In order to provide a quantitative proof we consider $|DW(t) - DW(t+\tau)|$ averaged over all particles, that are at distance r from a cage jump taking place at time t and subtract from that quantity its $r = \infty$ value. In fig. 3(right) we show this quantity, called $\Delta^J(r)$, for $\tau = 17$. One finds that $\Delta^J(r)$ is quite long ranged, in particular much more than the cage jump correlation function $\rho^J(r)$, see fig. 3(right) and its caption for a precise definition of $\rho^J(r)$. What is mediating the non-local interaction between cage jumps and DWs is an intriguing question. One possibility is that a slowly varying spatial field, like the thermal strain discussed in [24], plays an important role by providing long ranged dynamical interactions.

The picture that emerges from our study is that the dynamics occurs, as in the dense granular system studied in [10, 11], via a two time scale process that gives rise to dynamical heterogeneities and induces macroscopic relaxation. At short times, the particles collectively jump within clusters whose sizes are very widely distributed. These clustered jumps trigger other ones nearby, leading to well separated large scale avalanches.

We find that this dynamical facilitation process is coupled to the structure : mobility preferentially follows the soft regions and has a non-local influence on the evolution of the topography of hard and soft areas. The resulting picture of facilitation is quite different from the one based on the propagation of a conserved mobility field.

Studying the evolution of dynamical properties with decreasing temperature following the same analysis would allow for direct tests of prominent theories of the glass transition. For example, in the picture based on kinetically constrained models of glasses [7] facilitation should become more relevant and conserved upon lowering the temperature. In the random first order transition theory [25], the dynamics should be correlated with soft regions for moderately supercooled liquids but, closer to the glass transition, the relaxation should be dominated by other processes. Three of us [11] have performed such analysis for granular media and found that facilitation becomes *less* conserved as the density is increased. Performing a similar analysis for our model of supercooled liquids would be extremely important. Work in this direction is in progress.

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