Possible Universal Relation Between Short time $\beta$-relaxation and Long time $\alpha$-relaxation in Glass-forming Liquids

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Relaxation processes in supercooled liquids are known to exhibit interesting as well as complex behavior. One of the hallmarks of this relaxation process observed in the measured auto correlation function is occurrence of multiple steps of relaxation. The shorter time relaxation is known as the $\beta$-relaxation which is believed to be due to the motion of particles in the cage formed by their neighbors. One the other hand longer time relaxation, the $\alpha$-relaxation is believed to be the main relaxation process in the liquids. The timescales of these two relaxations processes dramatically separate out with supercooling. In spite of decades of researches, it is still not clearly known how these relaxation processes are related to each other. In this work we show that, there is a possible universal relation between short time $\beta$-relaxation and the long time $\alpha$-relaxation. This relation is found to be quite robust across many different model systems. Finally we show that length scale obtained from the finite size scaling analysis of $\beta$ timescale is same as that of length scale associated with the dynamic heterogeneity in both two and three dimensions.

Dynamics of supercooled liquids are very complex in nature. The decay of two point density-density auto correlation shows two steps relaxations as the liquid is supercooled. In spite of decades of research the complex dynamical behaviours associated with putative glass transition, is still poorly understood. The density auto correlation function decays to a plateau at shorter time and then at much longer time it finally decays from the plateau to zero in a stretched exponential manner. The short time relaxation in the plateau region is known as $\beta$-relaxation whereas the longer time relaxation is called the $\alpha$-relaxation. Although a lot of efforts have been made to understand the nature of $\alpha$ relaxation and its microscopic origin, far less researches are done to understand the same for shorter time $\beta$-relaxation and its possible connection with the $\alpha$-relaxation.

At short times, it is believed that the particles get trapped in transient cage formed by their neighboring particles and they undergo a kind of rattling motion in those cages. Eventually they hop out of the cage and probably after successive such cage breaking processes, the liquid finally relaxes. It is also not clearly known whether the rattling in a cage and subsequent breaking of the cage is the $\beta$-relaxation. If one assumes such an event to be a $\beta$-relaxation and multiple such events leads to $\alpha$-relaxation, then one can expect that short time and long time relaxation processes will be intimately related to each other. Such a scenario is indeed suggested in some studies.

In [10], author have proposed a correlation between $\beta$-relaxation time $\tau_\beta(T_g)$ calculated at the experimental glass transition temperature ($T_g$), and the Kohlrausch-Williams-Watts (KWW) exponent $(1-n)$ of $\alpha$-relaxation at $T_g$. $T_g$ is defined experimentally at the temperature where the relaxation time of the liquid becomes 100s. The KWW exponent is the stretching exponent of the decay profile of the two-point density-density auto correlation function or the self intermediate scattering function as

$$Q(t) = \exp \left[-\left(\frac{t}{\tau_\alpha}\right)^{1-n}\right], \quad 0 \leq n \leq 1, \quad (1)$$

where $Q(t) = \left(\sum_{i=1}^{N} w(|\vec{r}_i(t) - \vec{r}_i(0)|)\right)$. The window function $w(x) = 1.0$ if $x \leq 0.3$ and 0 otherwise (see SI for further details). $(\ldots)$ represents ensemble average. The $\alpha$ relaxation time is defined as $Q(\tau_\alpha) = 1/e$. At high temperature, the relaxation in liquid is exponential, that is $n = 0$, thus one can expect that the nonzero value of $n$ will be related to many body or cooperative nature of the $\alpha$-relaxation process.

The Coupling Model (CM) proposed in [13–15], suggests a relation between primitive relaxation time, $\tau_0$ with that of the the $\alpha$-relaxation time as

$$\tau_\alpha = \left[\tau_0 \tau^{-n}_c\right]^{1/(1-n)}, \quad (2)$$

where $\tau_c$ is the microscopic time scale. $\tau_0$, the primitive relaxation time is argued to be close to $\beta$-relaxation time as both of them are assumed to be the precursors of the long time $\alpha$-relaxation process. This relationship has been tested for many glass forming liquids near the experimental glass transition temperature and found to agree with the above relation to varying degree. Fujimori and Ouni’s correlation index $c$ [18], defined as $c \equiv 1 - \frac{\tau_0}{\tau_\beta}$, and the coupling parameter $n$ of CM was also shown subsequently to be linearly proportional to each other for many experimental glass-forming liquids. This clearly suggests that although there are proposals and reports of possible inter-relation between $\tau_\alpha$ and $\tau_\beta$, a general consensus is still missing.

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In a recent study [1], it has been shown that the system size dependence of $\tau_3$ in three dimensional glass-forming liquids is controlled by the dynamic heterogeneity length ($\xi_\alpha$) that are obtained from the finite size scaling [19, 20] of peak height of the four-point dynamic susceptibility ($\chi_4(t)$, see SI for definition). $\chi_4(T)$ [3, 19]. The peak of $\chi_4(t)$ appears at $\alpha$-relaxation time scale, suggesting a very strong inter-relation between these two relaxation processes. Thus a possible universal relation between $\alpha$ and $\beta$ time scale and its origin can be connected to the growth of different length-scales in the system. The main objective of the present work is to revisit this possible relationship between $\tau_3$ and $\tau_\alpha$ and try to explore existence of an universal relationship between these two timescales using more microscopic quantities like dynamic heterogeneity length scale ($\xi_\alpha$) and static length scale ($\xi_s$) that grow with supercooling [3, 4, 21, 22].

In this article, we propose a new universal relation between $\beta$-relaxation and $\alpha$-relaxation in model glass-forming liquids and try to rationalize the results within the framework of the well-known Random First Order Transition (RFOT) [2, 23, 24] theory of glass transition. Rest of the paper is arranged as follows. First we briefly discuss some of the details of the simulation methods and the models and then we define some relevant correlation functions that are used to calculate different relaxation times and length-scales. A set of new quantities are defined to analyze the data particularly for two dimensional systems. In two dimensions, there will be contribution from long-wave length phonon mode and appropriate corrections need to be made to disentangle the effect due to glass transition and long wavelength density fluctuations on the measured quantities [25–31]. We then discuss about the relation between $\tau_\alpha$ and $\tau_3$ for both two and three dimensional systems. Effects of finite size on these results are then discussed. Finally we rationalize our observation within the framework of RFOT theory and propose a new universal relationship between $\tau_\alpha$ and $\tau_3$.

We have studied different model systems in two and three dimensions with somewhat different inter particle potentials to make sure that the results obtained are generic and applicable for wide variety of systems. First model is the well known Kob-Andersen binary model where particles interact via Lennard-Jones potential. We refer the model as 3dKA [32]. The second model studied is also a binary mixture of particles but with pure repulsive inter particle interactions and this is referred here as 3dR10 [32]. Other models are 3dIPL [34], 3dHP [35, 36], 3dBMLJ [32, 37]. We have done very large system size simulations in three dimensions to remove finite size effects (see SI for further details). We have done simulations for system sizes in the range $N \in [1000, 108000]$. The data reported in the article is for $N = 108000$ only. In two dimensions, we study same Kob-Andersen model and refer it as 2dKA.

A slightly modified version of 2dKA model is also studied and will be referred as 2dPoly. The system size in two dimensions ranges from $N \in [100, 10000]$. All the details regarding these different models and the simulations details are given in the SI.

![FIG. 1: Top panel: Mean square displacement (MSD) and its log-derivative are plotted against time for 3dKA model. In the inset we show the temperature dependence of $\tau_3$ for the same model. The temperature dependence is close to Arrhenius in agreement with previous experimental results. Bottom panel: Similar plot for 3dR10 model. In the inset we show the temperature dependence of $\tau_3$ for 3dR10 model. For this model also the temperature dependence of $\tau_3$ follows Arrhenius behavior.](image_url)
below as

$$\langle |\Delta r(t)|^2 \rangle = \left\langle \frac{1}{N} \sum_{i=1}^{N} |\vec{r}_i(t) - \vec{r}_i(0)|^2 \right\rangle. \quad (3)$$

MSD shows a point of inflection at an intermediate time, thus if we plot the log-derivative of MSD with time, $d\log \langle |\Delta r(t)|^2 \rangle/d\log t$, it will show a dip at that inflection point. $\tau_\beta$ is defined as the time where the point of inflection appears. This procedure is shown for 3dMKA (top panel) and 3dR10 (bottom panel) models in Fig.1. One can clearly see the minimum in $d\log \langle |\Delta r(t)|^2 \rangle/d\log t$ vs $t$ plots and also the minimum shifts to higher and higher values of $t$ with decreasing temperature.

![Graph](image1)

**FIG. 2:** Data collapse of finite size dependence of $\tau_\beta$ for 2dR10 mode. Here $\tau_\beta$ is obtained from normal MSD (see text for details). Inset shows the variation of length scale with temperature. The scale obtained here are very different than the dynamic heterogeneity length scale of this model system.

In [1], it was shown that the system size dependence of $\tau_\beta$ is controlled by the dynamic heterogeneity length scale in three dimensions. To check the validity of the same results in two dimensional models, we have done the finite size scaling (FSS) analysis of $\tau_\beta$ for 2dR10 model. The results are shown in Fig.2. $\tau_\beta$ has fairly large system size dependence (shown in SI) and the dependence becomes very strong at lower temperatures. Although the data collapse observed is quite good, the obtained length scales as shown in the inset, is found to be very different from the dynamic heterogeneity length scale of this model obtained via different methods.

The variation of the length scale in the studied temperature range is very large suggesting a possible contribution coming from long wavelength density fluctuations which are prevalent in two dimensional systems due to Marmin-Wagner theorem [29, 38-40]. Thus disentangling contributions coming from these long wavelength fluctuations and the glass transition is very important to understand glass transition in two dimensions [31, 40, 41]. To overcome such a problem in two dimensional system we have calculated cage-relative MSD (crMSD) following [31, 30, 41]. The crMSD is defined as follows. First we define the cage related displacement of particle $i$ as

$$\Delta r_{i,CR}(t) = \Delta r_i(t) - \frac{1}{N_{nn}} \sum_{j \in n, n} \Delta r_j(t), \quad (4)$$

where $N_{nn}$ are the number of nearest neighbor of $i^{th}$ particle, and $\Delta r_j(t) = r_j(t) - r_j(0)$. Particles are defined as the neighbors of $i^{th}$ particle if they satisfy $r_{ij}(t) = \sum_{n=1}^{N} |\vec{r}_n(t) - \vec{r}_n(0)|^2$. In inset shows the Arrhenius temperature dependence of $\tau_\beta$. Bottom panel: Similar plot for the 2dR10 model. Similar Arrhenius temperature dependence of $\tau_\beta$ is shown for this model in the inset.

![Graph](image2)

**FIG. 3:** Top panel: Cage-relative mean square displacement (crMSD) and its log-derivative is plotted against time for 2dMKA system. Inset shows the Arrhenius temperature dependence of $\tau_\beta$. Bottom panel: Similar plot for the 2dR10 model. Similar Arrhenius temperature dependence of $\tau_\beta$ is shown for this model in the inset.
Some quasi-universal relation is observed in two dimensions. After calculating $\tau_\beta$, one is able to extract all the relevant information from MSD related only to glass transition in two dimensions.

In the left panel of Fig.4, we show the system size dependence of $\tau_\beta$ obtained from the log-derivative of cage-relative mean square displacement (crMSD) with dynamic heterogeneity length scale as the cutoff (see text for details) for the 2dR10 model. Right panel: Data collapse to obtain the length scale which is found to be same as the dynamic heterogeneity length scale.

$$|r_j(t) - r_j(0)| < A \sigma_{\alpha\beta}.$$ A is the cutoff used to define the neighbors. It is usually taken as the distance where first minimum of the radial distribution function $g(r)$ appears. The crMSD is then defined as,

$$\langle |\Delta r_{CR}(t)|^2 \rangle = \left( \frac{1}{N} \sum_{i=1}^{N} |\Delta r_{i,CR}(t)|^2 \right)$$

(5)

As we are measuring relative displacements, it will not be affected by the long wavelength phonon modes and thus one should be able to extract the relevant information from MSD related only to glass transition in two dimensions.

It turns out that the results very much depend on how the neighboring particles are chosen, for example, if one chooses a cutoff at the second minimum in $g(r)$, then one finds a week system size dependence of $\tau_\beta$ compare to almost no system size dependence if first minimum of $g(r)$ is chosen. This is somewhat puzzling and seems to constraint the usefulness of the crMSD. One can rationalize these results from the understanding that if one defines a cage relative motion using particles which are its immediate neighbors then one is basically removing even local cooperative motions in the systems. This local cooperative motion has nothing to do with the long wavelength phonon mode.

To keep the cooperative motions undisturbed over the dynamic heterogeneity length scale, we choose the cutoff length for defining the neighbors to be same as the dynamic heterogeneity length. This procedure gives us an estimate of $\tau_\beta$ which is not affected by the long wavelength density fluctuations at the same time any possible contributions coming from cooperative motions will not be washed away. In our subsequent analysis we have followed this method to calculate $\tau_\beta$. In the top panels of Fig.5, we have shown $d \log \langle |\Delta r_{CR}(t)|^2 \rangle / d \log t$ vs $t$ plots for 2dmKA and 2dR10 models. In the inset we show the Arrhenius temperature dependence of $\tau_\beta$ for these model systems.

In the left panel of Fig.5, we show the system size dependence of $\tau_\beta$ for 2dR10 model and in the right panel we show the finite size scaling of the same data using dynamic heterogeneity length scale, $\xi_t$ taken from Ref.[42]. The data collapse is indeed reasonable. Thus it can be concluded that finite size scaling of $\tau_\beta$ is governed by the dynamic heterogeneity length scale in two dimensions also. This is very similar to the observation reported for three dimensional model [8]. This suggests that glass transitions in two and three dimensions rely on the possible universal relation between these two timescales. As discussed earlier, in Ref.[10], a power law relationship has been proposed between $\tau_\alpha$ and $\tau_\beta$ as $\tau_\alpha \sim \tau_\beta^{1/(1-n)}$. As KWW stretching exponent $1-n$ decreases from 1 with supercooling in a manner which is
similar for different model systems, one expects to be able to obtain a master curve by plotting $\tau_\alpha$ as a function of $\tau_\beta$ for all the temperatures with appropriate choice of the pre-factor in the power law relation. In Fig.5, we have tested the same proposal by plotting $\tau_\alpha$ as a function of $\tau_\beta$ in log-log plot for both two and three dimensional models.

One can see that the power law relation is not very robust in the studied temperature range for three dimensional systems and deviates strongly especially at lower temperatures. In two dimensions also one sees similar results but data seem to fall on a quasi universal master curve. The observed data collapse although is not very satisfactory. In this work, we propose a simple but robust relation between $\alpha$-relaxation and $\beta$-relaxation time. The form of the proposed relationship between these two timescales can be rationalized within the framework of Random First Order Transition (RFOT) theory \cite{2,23,24}. In RFOT long time structural relaxation time $\tau_\alpha$ is connected to static length scale, $\xi_s$ via,

$$\tau_\alpha(T) = \tau_\infty \exp \left( \frac{\mu c_v}{K_B T} \right).$$

(6)

As shown in \cite{9} and in present work, finite size effects of $\tau_\beta$ can be understood using the dynamic heterogeneity length scale, $\xi_d$ in both dimensions, we expect from dynamic scaling arguments that $\tau_\beta$ will be related to $\xi_d$ as

$$\tau_\beta \sim \xi_d^z,$$

(7)

where exponent $z$ is found to be close to 0.8 for all three
FIG. 8: Top left panel: $\tau_\alpha$ is plotted against $\tau_\beta^3/T$ for all the studied two dimensional model systems. Top right panel: Similar plot for all the studied three dimensional model systems. Both in two and three dimensions data follow a master curve which suggests the observed dependence to be very robust. Bottom left panel: $\ln(\tau_\alpha(T)/\tau_\alpha(T_0))$ is plotted against $A[(T_0/T)(\tau_\beta(T)/\tau_\beta(T_0))^{-\gamma}-1]$ for all the two dimensional model systems. All the data are clearly on a single master curve with $\gamma = 0.450$. Bottom right panel: Similar plot for all the model three dimensional model systems with $\gamma = 0.600$.

dimensional model systems and $z \simeq 1.25$ for all the studied models in two dimensions as shown in Fig.6. If one assumes that static and dynamic length scales are related as $\xi_d \sim \xi_s^x$, then the following relation between $\tau_\alpha$ and $\tau_\beta$ can be obtained.

$$\tau_\alpha = \tau_0 \exp \left( \frac{\Omega \xi_s^x}{K_B T} \right), \quad (8)$$

where exponent $\gamma = \psi/zX$. Thus if $\tau_\alpha$ is plotted as a function of $\tau_\beta^3/T$, then one would expect to have a master curve if exponent $\gamma$ is universal for all model systems. In general, there is no reasons to expect $\gamma$ to be universal as exponent $\psi$ and $X$ are somewhat different amongst different model systems (see SI for the values of $\psi$ for different models). As shown in Fig.7 exponent $X$ is indeed different for different models in both two and three dimensions. The exponent $X = 2.7, 3.5, 3.5, 2.8$ and 1.0 for 3dKA, 3dHP, 3dR10, 3dIPL and 3dBMLJ_82 respectively. As the range of power law is somewhat restricted, reliable estimate of $X$ is not easy. The values of $X$ for different two dimensional models are $X = 1.30(2dmKA), 1.23(2dR10), 1.00(2dKA$ and 2dPoly). Note that, there are model systems which show presence of prominent medium range crystalline order at lower temperature or higher density and for them it has been shown that static and dynamic length scales are same [42]. 3dBMLJ_82 in three dimensions and 2dKA, 2dPoly in two dimensions are examples of such models. The data for $\xi_d$ and $\xi_s$ are
taken from [42, 43].

We then test the universality of Eq. [8] for different two and three dimensional model systems. In top panels of Fig. [8] we have plotted \( \tau_\alpha \) as a function of \( \tau_\beta / T \) with \( \gamma = 0.60 \) for three dimensions (top left panel) and \( \gamma = 0.45 \) for two dimensions (top right panel). In these plots we have adjusted the values of non-universal parameters like \( \tau_0 \) and \( \Omega \) in Eq. [8] to collapse all the data on a master curve. The quality of collapse clearly suggest that indeed \( \tau_\alpha \) is universally related to \( \tau_\beta \) via Eq. [8] with a universal exponent \( \gamma \).

Now one might suspect the reliability of the above relation as \( \tau_0 \) and \( \Omega \) are varied freely, to eliminate dependence on \( \tau_0 \), we take a reference temperature \( T_0 \) (highest temperature studied for each model) and divide Eq. [8] from both sides to write,

\[
\log \frac{\tau_\alpha (T)}{\tau_\alpha (T_0)} = \Omega \left[ \left( \frac{T_0}{T} \right)^{\beta} \left( \frac{\tau_\beta (T)}{\tau_\beta (T_0)} \right)^{\gamma} - 1 \right] \tag{9}
\]

Eq. [9] keeps only one parameter free and that is \( \Omega \). In bottom panels of Fig. [8] we have plotted \( \log \left( \frac{\tau_\alpha (T)}{\tau_\alpha (T_0)} \right) \) vs \( \frac{T_0}{T} \left( \frac{\tau_\beta (T)}{\tau_\beta (T_0)} \right)^{\gamma} - 1 \) for both two and three dimensions. The observed data collapse again reconfirms the proposed universal relation between \( \tau_\alpha \) and \( \tau_\beta \). Using reported values of \( \psi \) for the different models (see SI for the details), one obtains the values of \( \gamma \) to be close to 0.51, 0.79, 0.66 and 0.54 for 3dKA, 3dHP, 3dR10 and 3dIPL models. These numbers are in agreement with the chosen value of \( \gamma = 0.6 \) for three dimensional models. However, the exponent turns out to be somewhat different for 3dBMLJ [8] model (\( \gamma \approx 1.125 \)). We believe that this discrepancy is probably due to uncertainties in the values of \( \psi, z \) as well as \( X \). For two dimensional models, the numbers are 0.43, 0.58, 0.57 and 0.54 for 2dmKA, 2dR10, 2dPoly, and 2dKA respectively. Thus the numbers are in agreement with the universal number \( \gamma = 0.45 \) for two dimensions.

If the proposed universal relation between \( \tau_\alpha \) and \( \tau_\beta \) is shown to be valid for experimentally relevant glass forming liquids, then one might be able to understand the vitrification in liquids by probably understanding the relaxation processes only. This might lead us to identify the relevant elementary relaxation process responsible for both \( \beta \) and \( \alpha \) relaxation in glassy systems. On a slightly different note, it was shown in \[44\] that the collapsing dynamics of a polymer chain in a supercooled liquid is controlled by both \( \alpha \) and \( \beta \) relaxation processes with possible implications in bio-preservation. It is suggested that not only \( \alpha \)-relaxation but also \( \beta \)-relaxation should be taken into account in order to understand the degradation process of biomolecules \[45, 46\]. Thus our proposed universal relation between these two relaxation processes might help us design the appropriate glassy matrix in future to preserve bio-macromolecules more efficiently.

To conclude, we have shown that there is a universal relation between \( \alpha \) and \( \beta \) relaxation times of glass forming liquids. The proposed relation is different from the one predicted by Coupling Model \[10\]. The new relation can be rationalized within the framework of Random First Order Transition Theory. In two dimensions, due to long wavelength density fluctuations, different transport quantities show logarithmic system size dependence and disentangling this effect from the effect emanating from glass transition is often difficult. We show how \( \tau_\beta \) can be calculated in two dimensions by appropriately modifying the correlation functions to remove the effect of long wavelength phonon without affecting the cooperative motions at the relevant dynamical heterogeneity length scale. We then shown that finite size scaling of \( \tau_\beta \) is controlled by the dynamic heterogeneity length scale as in the three dimensional models. Finally, the obtained universal relationship between \( \tau_\alpha \) and \( \tau_\beta \) in both the dimensions suggests that the physics of glass transition may be very similar in both two and three dimensions. This observation is in agreement with recent findings \[28, 30\]. As \( \beta \)-relaxation plays in important role below the glass transition, in future it will be very interesting and important for industrial applications to study possible aging behavior of \( \tau_\beta \) and its correlation with the aging behavior of \( \tau_\alpha \).

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We have arranged the supplementary information as follows. First we will give the details of the models that we have studied and discuss about the methods that we have followed in the Sec. I. Next we will discuss how we have calculated $\tau_\beta$ from the log derivative of MSD in Sec. III. In Sec. IV we will discuss about the cage-relative tendency towards local crystalline order at lower temperatures in 2dmKA. These authors contributed equally.

\section{Models and Methods}

We have studied the following model glass-forming liquids in both two and three dimensions.

\subsection*{3dKA}

The well-known Kob-Andersen model is a 80 : 20 binary mixture of two type of particles where the interacting potential is,

$$V_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta} \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{6}.$$  \hfill (1)

Here the parameters are, $\alpha, \beta \in \{A, B\}$ and $\epsilon_{AA} = 1.0, \epsilon_{AB} = 1.5, \epsilon_{BB} = 0.5, \sigma_{AA} = 1.0, \sigma_{AB} = 0.8, \sigma_{BB} = 0.88$, number density $\rho = 1.20$. We have chosen the cut off of the interaction potential as 2.5$\sigma_{\alpha\beta}$. We have used a quadratic polynomial in such a way that the potential and its first two derivatives are continuous at the cutoff radius. Our studied temperature range is $T \in \{0.45, 3.00\}$. We have done simulations in three dimensions with $N = 108000$ number of particles.

\subsection*{2dKA}

This is same model as 3dKA but in two spatial dimensions. The temperature range is $T \in \{0.930, 2.000\}$. The system size is $N = 10000$. This model is known to show prominent medium range crystalline order (mrco) at lower temperatures.

\subsection*{2dmKA}

This is a slightly modified version of 2dKA model. The parameters are same as 2dKA but bigger to smaller particle number ratio is 65 : 35. Temperature range is $T \in \{0.45, 2.0\}$. This model is shown to have less tendency towards local crystalline order at lower temperatures.

\subsection*{3dR10}

This is a 50 : 50 binary mixture of harmonic spheres where the diameter ratio of the two type of particles is 1.4. This model which has been studied extensively in the context of jamming in granular medium. In this model, particles interacts via,

$$V_{\alpha\beta}(r) = \epsilon \left[ 1 - \left( \frac{r}{\sigma_{\alpha\beta}} \right)^2 \right]^2$$  \hfill (4)

if $r < \sigma_{\alpha\beta}$ and 0 otherwise. $\sigma_{\alpha\beta} = (\sigma_{\alpha} + \sigma_{\beta})/2$. Number density $\rho = 0.82$ and $\epsilon = 1.0$. The temperature range studied is $T \in \{0.0045, 0.009\}$.

\subsection*{3dBMLJ82}

This model system consists of equimolar additive mixtures. The system consists of $N = 108000$ particles interact via the Lenard-Jones potential. Particles of species 2 have a smaller diameter than those of species 1 ($\sigma_{22} < \sigma_{11}$). The masses of the two species are equal $m_1 = 1.0$ and $m_2 = 1.0$ and the size ratio $\lambda = 0.82$, keeping $\sigma_{11} = 1.0$. We studied the system in the temperature range $T \in \{0.670, 2.000\}$.

\subsection*{2dPoly}

This is a poly-disperse mixture of particles where the diameter $\sigma_i$ of the particle $i$ is chosen from a Gaussian distribution. The polydispersity parameter is then defined as

$$\Delta = \sqrt{\langle \sigma^2 \rangle} / <\sigma>. \hfill (5)$$
where $\delta\sigma = \sigma - \langle \sigma \rangle$. We have chosen $\Delta = 11\%$. The interaction potential between a pair of particles is defined as,

$$V_{ij}(r) = 4\epsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r} \right)^{12} - \left( \frac{\sigma_{ij}}{r} \right)^6 + \frac{1}{4} \right]$$

(6)

if $r < 2^{1/6}\sigma_{ij}$, else 0. Here $\sigma_{ij} = (\sigma_i + \sigma_j)/2$. The temperature range studied is $T \in \{0.450, 0.900\}$. We simulated the system at packing fraction $\eta = 0.76$.

We have performed NVT molecular dynamic simulations for all the model system studied. We have studied different system sizes. For two dimensional systems we have gone up to $N = 10000$. For three dimensional systems we have gone up to $N = 108000$. For simulation we have made sure that the systems are equilibrated before storing data by equilibrating the systems for at least $100\tau_\alpha$. For better statistical average we have performed 32 statistically independent simulations for each temperature.

II. CORRELATION FUNCTIONS

Overlap Correlation Function : The point density correlation function or the overlap correlation function is defined as

$$Q(t) = \frac{1}{N} \left\langle \sum_{i=1}^{N} w(|\vec{r}_i(t) - \vec{r}_i(0)|) \right\rangle.$$  

(7)

The window function $w(x) = 1.0$ if $x \leq 0.3$ and 0 otherwise. This window function is used to remove possible decorrelation that might happen due to vibrational motion inside the cage formed around a particle by their neighbors. $\langle \ldots \rangle$ represents ensemble average. The $\alpha$ relaxation time is defined as $Q(\alpha) = 1/e$.

Four-point Susceptibility : The four-point susceptibility is defined as the fluctuations of the overlap correlation function as

$$\chi_4(t) = N \left[ \langle Q^2(t) \rangle - \langle Q(t) \rangle^2 \right],$$

(8)

and the peak of $\chi_4(t)$ appears at timescale close to $\alpha$-relaxation time. The peak, $\chi_4^P$, is defined as

$$\chi_4^P \equiv \chi_4(t = \alpha).$$

(9)

Cage relative Mean Squared Displacement (cr-MDS) : Mean squared displacement (MSD) is defined as

$$\langle |\Delta r(t)|^2 \rangle = \left\langle \frac{1}{N} \sum_{i=1}^{N} |\vec{r}_i(t) - \vec{r}_i(0)|^2 \right\rangle.$$  

(10)

In two dimensions, MSD will be affected by the long wavelength density fluctuations and to remove this effect, a relative MSD with respect to cage is calculated. This is termed as cage relative MSD (crMSD). It is defined as follows. First we define the cage related displacement of particle $i$ as

$$\Delta r_{i,CR}(t) = \Delta r_i(t) - \frac{1}{N_{nn}} \sum_{j \neq \text{n.n}} \Delta r_j(t),$$

(11)

where $N_{nn}$ are the number of nearest neighbor of $i^{th}$ particle, and $\Delta r_j(t) = r_j(t) - r_j(0)$. Particles are defined as the neighbors of $i^{th}$ particle if they satisfy $r_{ij} = |r_j(t) - r_j(0)| < A\sigma_{\alpha\beta}$. $A$ is the cutoff value used to define the neighbors. Often one uses the value at which first minimum of the radial distribution function $g(r)$ appears. Now the crMSD is defined as,

$$\langle |\Delta r_{i,CR}(t)|^2 \rangle = \left\langle \frac{1}{N} \sum_{i=1}^{N} |\Delta r_{i,CR}(t)|^2 \right\rangle$$

(12)

We have found that the value of $\tau_\beta$ and particularly the system size dependence of it depends crucially on the choice of the cutoff. Below we have done a systematic analysis to understand this and to choose an appropriate cutoff for the subsequent analysis.

III. CALCULATION OF $\tau_\beta$

As discussed in the main text we have calculated the $\beta$-time scale $\tau_\beta$ from the minimum of the $\log < |\Delta r(t)|^2 > / \log(t)$ vs $\log(t)$ plot. To calculate numerical derivative as we need very closely spaced data, we have used a splined data. We used cubic spline to get the splined data. This splined data was again smoothen before calculating the derivative as a minute fluctuation in

FIG. 1: Showing original and smoothed MSD data to obtain derivative for 2dR10 model for $T = 0.520$. Inset shows the zoomed version of the plot to show how small fluctuation in data is avoided.
the data will cause a huge fluctuation in derivative, which will make it difficult to find the minimum unambiguously. In fig. 1, we have shown the original and smoothed MSD for 2dR10 model for $T = 0.520$. The inset shows the plateau region zoomed to show the quality of the spline interpolation. Similar method is also used for extracting the value of $\tau_3$ for two dimensional models from crMSD.

IV. CUTOFF DEPENDENCE OF CRMSD

We have discussed in the main text that the system size dependence of $\tau_3$ depends on the choice of the cutoff we while calculating crMSD. If we choose 2nd neighbor distance as cutoff instead of 1st neighbor distance, then the system size dependence of $\tau_3$ changes. In Fig. 2, we show the system size dependence of $\tau_3$ for three different cutoff distances. One can clearly see how system size dependence changes on the choice of cutoff for 2dmKA model. As this becomes somewhat ambiguous, we choose dynamic heterogeneity length scale as the cutoff. With this choice, the values of $\tau_3$ seem to show behaviour very similar to the results obtained for three dimensional models. In the main articles, all reported numbers of $\tau_3$ for two dimensional models are obtained with dynamic heterogeneity length scale as cutoff for neighbor calculation.

V. EFFECT OF SYSTEM SIZE ON THE RELATION BETWEEN $\tau_3$ AND $\tau_\alpha$:

In Fig. 3, we have plotted $\tau_\alpha$ as a function of $\tau_3$ for all the model systems in three dimensions using data from different system sizes. The deviation observed from a
The details of the exponents ($\psi$, $X$, $z$ and $\gamma$) quoted in the main articles are given below:

| Three Dimensional Models | Two Dimensional Models |
|--------------------------|------------------------|
| $\psi$ | $z$ | $X$ | $\gamma = \psi/zX$ | $\psi$ | $z$ | $X$ | $\gamma = \psi/zX$ |
| 3dKA | 1.10 | 0.80 | 2.70 | 0.51 | 2dKA | 0.68 | 1.25 | 1.00 | 0.54 |
| 3dHP | 2.22 | 0.80 | 3.50 | 0.79 | 2dmKA | 0.70 | 1.25 | 1.30 | 0.43 |
| 3dR10 | 1.86 | 0.80 | 3.50 | 0.66 | 2dR10 | 0.89 | 1.25 | 1.23 | 0.58 |
| 3dIPL | 1.21 | 0.80 | 2.80 | 0.54 | 2dPoly | 0.71 | 1.25 | 1.00 | 0.57 |
| 3dBMLJ_82 | 0.90 | 0.80 | 1.00 | 1.125 |  |  |  |  |

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