Block Analysis for the Calculation of Dynamic and Static Length Scales in Glass-Forming Liquids

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We present block analysis, an efficient method to perform finite-size scaling for obtaining the length scale of dynamic heterogeneity and the point-to-set length scale for generic glass-forming liquids. This method involves considering blocks of varying sizes embedded in a system of a fixed (large) size. The length scale associated with dynamic heterogeneity is obtained from a finite-size scaling analysis of the dependence of the four-point dynamic susceptibility on the block size. The block size dependence of the variance of the α-relaxation time yields the static point-to-set length scale. The values of the obtained length scales agree quantitatively with those obtained from other conventional methods. This method provides an efficient experimental tool for studying the growth of length scales in systems such as colloidal glasses for which performing finite-size scaling by carrying out experiments for varying system sizes may not be feasible.

The role of growing length scales in the rapid growth of the structural relaxation time of glass-forming liquids near the glass transition has received a lot of attention in recent years. Several length scales, both static and dynamic, have been proposed and their behavior near the glass transition and relevance to the growth of the structural relaxation time have been studied in a large number of theoretical, numerical and experimental investigations. However, there is still a lot of controversy about the behavior of these length scales as the glass transition is approached. Therefore, it is important to develop methods that can be used in simulations and experiments to accurately measure these length scales.

The existence of a growing dynamic length scale ξ_D that describes spatial correlations of the inhomogeneous local dynamics of glass-forming liquids (known as dynamic heterogeneity) is now well-established. This length scale has been calculated from finite-size scaling of a four-point susceptibility χ_4 and its associated Binder cumulant and the wavenumber-dependence of the corresponding structure factor. A similar length scale has also been obtained from the dependence of the local dynamics on the distance from an amorphous wall in which particles are fixed at their positions in an equilibrium configuration. However, the relation of this length scale with the length scale ξ_D of dynamic heterogeneity is controversial. Inhomogeneous mode-coupling theory provides a theoretical description of the growth of ξ_D and χ_4 as the glass transition is approached, but the quantitative predictions of this theory are somewhat different from the results obtained from numerical studies.

Another length scale that has received a lot of attention is the static “mosaic scale” (ξ_s) of the Random First-Order Transition (RFOT) theory of the glass transition. This length scale can be obtained from a “point-to-set” (PTS) construction in which particles outside a spherical cavity are fixed at their positions in an equilibrium configuration, the remaining particles inside the cavity are allowed to equilibrate, and the average overlap of the positions of these particles with their positions in the original equilibrium configuration is studied as a function of the radius of the cavity. The PTS method has been used in several studies to obtain the dependence of ξ_s on the temperature and the density. This length scale has also been calculated from FSS of the α-relaxation time and the minimum eigenvalue of the Hessian matrix that describes vibrations near a local minimum of the potential energy. In the temperature and density range accessible in simulations, ξ_s is found to be smaller than ξ_D and the growth of ξ_s with decreasing temperature or increasing density does not follow that of ξ_D, suggesting that these two length scales are distinct from each other.

Experimental studies of length scales in glass-forming liquids have been limited because quantities such as χ_4 that are required for calculating these length scales are not readily accessible in experiments. A calculation of χ_4 requires information about the trajectories of individual particles, which can be obtained in experiments on colloidal systems, but not in experiments on molecular liquids. Three-point and five-point susceptibilities that are closely related to χ_4 have been measured in experiments on molecular liquids. Three experiments and experiments on colloidal systems provide clear evidence for the growth of spatial correlations as the glass transition is approached. However, it
is difficult to extract values of relevant length scales from these measurements because the exact relation between these susceptibilities and the length scales is not known. A calculation of $\xi_s$ using the PTS method requires detailed information about the equilibrium properties of particles confined in small cavities of varying sizes. Such information is difficult to obtain from experiments, although a recent experiment \[27\] suggests that this may be possible in the near future. The only experiment in which values of both dynamic and static length scales have been obtained is Ref. \[28\] in which the method of Ref. \[14\] was implemented for a two-dimensional colloidal system. However, as mentioned earlier, the physical interpretation of the length scales obtained from this procedure is controversial.

As discussed above, FSS has played an important role in the calculation of both $\xi_D$ \[10\] and $\xi_s$ \[10, 13, 19\]. The conventional FSS method involves studies of the equilibrium behavior of systems with periodic boundary conditions and different sizes that are comparable to the (often rather small) values of the relevant length scales. This method can not be implemented in experiments because experimental studies of such systems are very difficult. It is, therefore, important to develop alternative FSS methods that can be implemented in experiments. Also, the conventional FSS method suffers from a few problems such as the necessity of carrying out long simulations and extensive averaging for obtaining reliable results for small systems and artifacts \[14\] arising from the suppression of density and composition fluctuations in small samples with periodic boundary conditions. FSS methods in which these problems are not present would be highly desirable.

In this Letter, we present a method of performing FSS in which the problems of conventional FSS analysis are avoided, leading to excellent scaling behavior. In this method, which we call “block analysis”, we perform equilibrium molecular dynamics (MD) simulations for a single large system. We then consider blocks of varying sizes embedded in the large system \[31\] and measure various quantities of interest, such as $\chi_4$ and the $\alpha$-relaxation time $\tau_\alpha$, for individual blocks. The length scale $\xi_D$ associated with dynamic heterogeneity is obtained from a FSS analysis of the dependence of $\chi_4$ and the associated Binder cumulant on the block size. The block-size dependence of the variance of the $\alpha$-relaxation time of individual blocks yields the static PTS length scale $\xi_s$. We show that the values of the obtained length scales agree quantitatively with those obtained from other methods for three different glass-forming liquids. Since this method involves observation of the trajectories of particles in a single large system, it can be readily implemented in experiments on colloidal systems.

We study three model glass-formers in three spatial dimensions. The first is the well-known Kob-Andersen binary mixture interacting via Lennard-Jones potentials \[32\]. We call this system the 3dKA model. Second, we study a 50 : 50 binary mixture interacting via purely repulsive interactions falling off of as $1/r^{10}$ \[33\]. We call this the 3dR10 model. Lastly, we study a variant of 3dKA system with only repulsive power law interactions \[34\]. We refer to this as the 3dIPPL model. Further details of the models and simulations can be found in the supplementary information (SI).

MD simulations are carried out for a single, moderately large system size, $N = \rho L_0^3$, where $\rho$ is the number density and $L_0$ is the length of the system. We then construct blocks of size $L_B = L_0/n$, where $n \in \{2, 3, 4, \ldots \}$ and calculate various dynamic quantities using the particles which are present inside one such box at a chosen time origin.

**The dynamic susceptibility $\chi_4$:** The self overlap correlation for a particular block size is defined as,

$$Q(L_B, t) = \frac{1}{N_B} \sum_{i=1}^{N_B} \sum_{j=1}^{n_i} w(|\vec{r}_j(t) - \vec{r}_j(0)|),$$  \(1\)

where $N_B$ is the number of blocks with size $L_B$, $n_i$ is the number of particles in the $i$-th block at time $t = 0$, and the window function $w(x) = \Theta(a - x)$ where $\Theta$ is the Heaviside step function and the value of the parameter $a$ is chosen to remove the decorrelation arising from vibrations of particles inside the cages formed by their neighbours. We take $a$ to be $0.3\sigma_{AA}$ for the 3dKA model where $\sigma_{AA}$ is the Lennard-Jones length parameter for the larger particles. The dynamical susceptibility associated with blocks of size $L_B$ is defined as follows.

$$\chi_4(L_B, t) = \frac{N L_B^3}{L_0^3} \langle [Q(L_B, t) - \langle Q(L_B, t) \rangle ]^2 \rangle$$  \(2\)

We consider the dependence of $\chi_4^\alpha(L_B, T)$, the peak value of $\chi_4(L_B, t)$ at temperature $T$, on the block size $L_B$ for a fixed value of $N = \rho L_0^3$. This dependence is shown in Fig. \[1\]. The left panel of the figure shows the data for $\chi_4^\alpha(L_B, T)$ as a function of the block length $L_B$ for different temperatures. The peak value of the dynamical susceptibility at a given temperature grows with $L_B$ and saturates at a temperature-dependent value $\chi_4^\alpha(\infty, T)$. The dependence of $\chi_4^\alpha(L_B, T)$ on $L_B$ is expected to exhibit the following FSS form:

$$\chi_4^\alpha(L_B, T) = \chi_4^\alpha(\infty, T) f(L_B/\xi(T)),$$  \(3\)

with $\xi(T) = \xi_D(T)$, the dynamic length scale. The data for all temperatures can be collapsed to a master curve using the two parameters, $\chi_4^\alpha(\infty, T)$ and $\xi(T)$, for each temperature, as shown in the right panel of Fig. \[1\]. The quality of the data collapse is very good and the length scale obtained this way is in complete agreement with that obtained using conventional FSS, as shown in the inset of the same figure. The legend “$\chi_4^\alpha$ FSS” refers to...
conventional FSS and the data are taken from Ref. [10]. We have also shown the comparison of \( \chi_4^B(\infty, T) \) with the conventional FSS values. One can see that at low temperatures, \( \chi_4^B(\infty, T) \) obtained from the block analysis is systematically larger than the conventional FSS result. This is due to the fact that particles can move in and out of individual blocks, so that the constraint of the total number of particles of each type being constant in simulations with periodic boundary conditions is not present for the blocks. This enhances fluctuations, leading to an increase in the peak height of \( \chi_4(t) \) for the blocks.

To ascertain whether the above analysis generically gives correct results for any model system, we have performed similar analysis for the 3dIPL and 3dR10 model systems. For both these models, the scaling collapses observed are quite good and the extracted dynamic length scales are also in good agreement with those obtained from conventional methods (see the SI for details).

**Distribution of \( Q(\tau_\alpha) \) and the Binder Cumulant:**

The FSS of \( \chi_4^B(N, T) \) requires two unknown parameters, \( \chi_4^B(\infty, T) \) and \( \xi(T) \). A better way of extracting the length scale is the FSS of the Binder cumulant obtained from the distribution of \( Q(\tau_\alpha) \) where \( \tau_\alpha \) is the long time \( \alpha \)-relaxation time defined as \( \langle Q(L_0, t = \tau_\alpha) \rangle = 1/e \). At a fixed temperature, the distribution of \( Q(\tau_\alpha) \) becomes flatter and more skewed as \( L_B \) is decreased, becoming nearly bimodal for small \( L_B \). A similar effect is seen if the temperature is decreased for fixed \( L_B \). This is because lowering the temperature is another way of lowering the block size measured in units of the dynamic correlation length. This is clearly seen in the top panels of Fig. 2. An earlier study [10] reported that the distribution of \( Q(\tau_\alpha) \) becomes bimodal for small systems with periodic boundary conditions. Such bimodality is not observed in our analysis using the block construction, presumably because of enhanced fluctuations of the density and the composition in the blocks for which the number of particles of each type is not conserved.

The skewness of the distribution of \( Q(\tau_\alpha) \) is quantitatively captured by the Binder cumulant which measures the deviation of the distribution from the Gaussian form. It is defined as

\[
B(L_B, T) = \frac{\langle (Q(L_B, \tau_\alpha) - \langle Q(L_B, \tau_\alpha) \rangle)^4 \rangle}{3\langle (Q(L_B, \tau_\alpha) - \langle Q(L_B, \tau_\alpha) \rangle)^2 \rangle^2} - 1. \tag{4}
\]

This quantity approaches zero at high temperatures and for large block sizes where the correlation length in the system is much smaller than the block size. The Binder cumulant is an ideal quantity to measure in a scaling analysis because it is known to be a scaling function of only the underlying correlation length:

\[
B(L_B, T) = F \left( \frac{L_B}{\xi_D} \right). \tag{5}
\]

The estimation of the length scale from FSS of \( B(L_B, T) \) is more reliable as it involves only one parameter for each temperature. The bottom left panel of Fig. 2 shows the Binder cumulant calculated from the distribution of \( Q(L_B, \tau_\alpha) \), plotted versus \( L_B \) for the 3dKA model. The middle panel shows the corresponding data collapse obtained using the dynamic length scale shown in the bottom right panel of the same figure. We also show a comparison of the length scales obtained using FSS of \( \chi_4^B \) and the Binder cumulant. The length scales obtained in these two calculations are in reasonably good agreement with each other.

**The statistics of \( \tau_\alpha \) - Calculation of the static
the static length scale obtained from this method and from the 3dKA model data has also been included for comparison.

FIG. 2. (For the 3dKA model) Top panel: Histograms of $Q(L_B, \tau)$ for blocks of size $L_B = 3.764, 6.274$ and $9.410$, going from left to right. Bottom left panel: Binder cumulant versus block size at different temperatures. Bottom middle panel: Binder cumulant versus block size scaled using the lengths shown in the bottom right panel where the length scale used for the collapse of $\chi_i^\tau$ data has also been included for comparison.

**length-scale:** Earlier studies [10, 19] have shown that

the $\alpha$-relaxation time of small systems with periodic boundary conditions increases as the system size is decreased and its system-size dependence is described by the static length scale $\xi$. We find that the $\tau$ for individual blocks, obtained from the self overlap correlation function $Q(t)$, does not show appreciable block-size dependence - the movement of particles in and out of blocks makes the system-size dependence of $\tau$ for blocks much weaker than that for systems with periodic boundary conditions. We then look at the dependence of the statistics of $\tau$ on the block size by calculating the distribution of $\tau$ as a function of block size. For each block, we first calculate $\tau^{(i)}_{\alpha}(L_B)$ by measuring the time at which the corresponding $Q^{(i)}(L_B, t)$ for a fixed time origin attains a value of $1/e$ (the superscript $i$ signifies that this is a quantity for a single block $i$ before any averaging is done). We then calculate the mean and the variance of this quantity and finally define $\chi_{\tau}(L_B, T)$ as,

$$\chi_{\tau}(L_B, T) = L_B^3 \left\langle \frac{1}{n_B} \sum_{i=1}^{n_B} \Delta \tau^{(i)}_{\alpha}(L_B) \right\rangle,$$

(6)

where $\Delta \tau^{(i)}_{\alpha}(L_B) = \frac{1}{n_B} \sum_{i=1}^{n_B} \tau^{(i)}_{\alpha}(L_B)$, $\Delta \tau^{(i)}_{\alpha}(L_B) = \tau^{(i)}_{\alpha}(L_B) - \tau^{(i)}_{\alpha}(L_B)$, and the outermost angular brackets stand for time-origin averaging. This quantity measures the spatial fluctuations in $\tau$. The dependence of $\chi_{\tau}$ on $L_B$ and $T$ (see the SI) clearly shows the presence of a length scale that grows at $T$ is decreased. Since the system-size dependence of $\tau$ itself (for systems with periodic boundary conditions) is governed by the static length scale $\xi$, one can expect the system-size dependence of $\chi_{\tau}$ also to be controlled by the same length scale. To check whether this is true, we performed a scaling analysis to find the length scale $\xi$ that leads to a scaling collapse of the data for $\chi_{\tau}(L_B, T)$. We find good data collapse, as shown in Fig. 3. The temperature de-
dependence of the length scale obtained from the scaling collapse is found to be very similar to that of the static length scale $\xi_s$ obtained in earlier work (see the inset of Fig. 3). This result, which shows that the block-size dependence of $\chi_r(L_B, T)$ is indeed governed by $\xi_s$, is very useful as it shows that the static length scale can be extracted from experimental or simulation data obtained for a single system of moderately large size. Similar analysis done for the 3dR10 model system are shown in SI. For this model also the results are in good agreement with those obtained from conventional methods.

To summarize, in this work, we present an efficient method which can be used in simulations as well as in colloidal experiments on glass forming liquids to obtain both static and dynamic length scales. Our results are validated from comparisons with those of conventional methods. This method has the advantage of capturing all the important fluctuations in the system which is not possible in simulations in the canonical ensemble for varying system sizes. Block analysis also provides extremely well-averaged results without any additional computational overhead in simulations and it can be implemented without much difficulty in colloidal glass experiments. We hope that this work will motivate experiments on colloidal glasses to measure these length scales.

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Block Analysis for the Calculation of Dynamic and Static Length Scales in Glass-Forming Liquids: Supplementary Information

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I. MODELS AND SIMULATION DETAILS

We have studied three different model glass forming liquids in three dimensions. The model details are given below:

3dKA Model:
The KABLJ [1] liquid is an 80:20 mixture of two LJ particles, A and B. This model was first introduced by Kob-Anderson to simulate \( N_{iso} P 20 \). This liquid has become the well known model system for studying viscous liquid dynamics, like other binary mixtures it’s size ratio is 6.50:50. This liquid has be~en studied for many years. The interaction range of the 3dKA model is much larger than the 3dIPL model.

3dR10 Model:
The interaction potential is cut-off at 1.38\( \sigma_{AA} \). These units corresponds to a length of 3.4\( \AA \). The interaction potential is given by

\[
V_{\alpha\beta}(r) = 4.0\epsilon_{\alpha\beta}[\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12} - 2\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{6}]
\]

where \( \alpha, \beta \in \{A, B\} \) and \( \epsilon_{AA} = 1.0, \epsilon_{AB} = 1.5, \epsilon_{BB} = 0.5; \sigma_{AA} = 1.0, \sigma_{AB} = 0.80, \sigma_{BB} = 0.88 \). The interaction potential was cut off at 2.50\( \sigma_{\alpha\beta} \) and the number density is \( \rho = 1.20 \). we use a quadratic polynomial to make the potential and its first two derivatives smooth at the cutoff distance. Length, energy and time scale are measured in units of \( \sigma_{AA}, \epsilon_{AA} \) and \( V_{\sigma_{AA}} \). For Argon these units corresponds to a length of 3.4\( \AA \), an energy of 120\( k_{b} \) and time of 3 \( \times 10^{-13} \)s.

3dIPL Model:
This is a 50:50 binary mixture [3] with the pure repulsive pair wise interaction potential, defined as

\[
V_{\alpha\beta}(r) = \epsilon_{\alpha\beta}[\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12}]
\]

Here \( \epsilon_{AA} = 1.0, \sigma_{AA} = 1.0, \sigma_{AB} = 1.22, \sigma_{BB} = 1.40 \). The interaction potential is cut-off at 1.38\( \sigma_{AA} \). The number density \( \rho = 0.81 \).

II. BLOCK SIZE DEPENDENCE OF \( \chi^L \) FOR THE 3DR10 AND 3DIPL MODELS.

To confirm the results reported for the block size dependence of \( \chi^L \) for 3dKA model, we have done similar analysis for 3dR10 and 3dIPL models. The results are shown in Fig[3]. Top panels of Fig[3] show the system size dependence of \( \chi^L \) for the 3dR10 model and the scaling collapse. Bottom panels of the same figure show the results obtained for the 3dIPL model. The scaling observed for both these model systems are very good and the obtained length scales from the block FSS analysis are in very good agreement with the results obtained by other conventional methods. For the 3dIPL model (shown in

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FIG. 1. Top panels: Block size dependence of $\chi_4^P$ for the 3dR10 model. In the left panel, we plot $\chi_4^P$ versus the block size and in the right panel a collapse is done by rescaling the x-axis using a suitable length-scale, $\xi(T)$ and the y-axis using the saturation value of $\chi_4^P$ for infinite block size. In the inset, the temperature dependences of $\xi(T)$ and $\chi_4^P(\infty, T)$ are compared with the corresponding quantities obtained using conventional finite-size-scaling. We have used $T_K \simeq 0.40$ for this model. Bottom Panels: Similar analysis done for 3dIPL model. Here in addition, the length scale obtained from the collapse of the $\beta$-relaxation time is also compared with the above length scales.

lower panels of the same figure) the length scale obtained from the FSS of short time scale, $\tau_\beta$ (taken from) are also compared with the length scale obtained from the block analysis. In it is shown that the FSS of $\tau_\beta$ are also controlled by the dynamic heterogeneity length scale.

III. BLOCK SIZE DEPENDENCE OF $\chi_\tau$

In the main article, we have discussed about the scaling collapse of $\chi_\tau$ using the static length scale for 3dKA model. Here in Fig. we have shown the $\chi_\tau$ for different block sizes for different temperatures for both 3dKA and 3dR10 model. Similar to 3dKA model, FSS of $\chi_\tau$ for 3dR10 model is also controlled by the corresponding static length scale of 3dR10 model. This establishes the usefulness of block analysis for extracting both static and dynamic length scales in a system with relatively less computational or experimental efforts. “FSS+PTS” refers to the static length scale obtained using FSS of $\alpha$-relaxation time and Point-to-Set (PTS) method in cavity geometry. The data is taken from Ref. [8].
FIG. 2. Top panels: Block size dependence of $\chi_\tau$ for the 3dKA model. In the left panel, we plot $\chi_\tau$ versus the block size and in the right panel a collapse is done by rescaling the $x$-axis using a suitable length-scale, $\xi(T)$ and the $y$-axis using the saturation value of $\chi_\tau$ for infinite block size. In the inset, the temperature dependence of $\xi(T)$ is compared with the static length scale obtained using conventional finite-size-scaling and PTS method. Bottom panels: Similar analysis done for 3dR10 model.

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