Four-point susceptibility of a glass-forming binary mixture: Brownian dynamics

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We study the four-point dynamic susceptibility $\chi_4(t)$ obtained from Brownian dynamics computer simulations of the Kob-Andersen Lennard-Jones mixture. We compare the results of the simulations with qualitative predictions of the mode-coupling theory. In addition, we test an estimate of the four-point susceptibility recently proposed by Berthier et al. [Science, 310, 1797 (2005)].

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I. INTRODUCTION

The origin of the extreme slowing down of liquids’ dynamics upon approaching the glass transition and the very nature of this transition are still hotly debated despite many experimental, simulational, and theoretical studies performed in the last two decades. One of the fundamental difficulties is that the slowing down appeared to be a local, small scale phenomenon which is not accompanied by a growing correlation length. No long-range correlations have ever been found in any static quantity upon approaching the glass transition. The attention has recently shifted to dynamic correlations, and there is evidence that there is a dynamic correlation length that slows down upon approaching the glass transition. Unfortunately this dynamic correlation length cannot be easily obtained from experimental data.

The dynamic correlation length is often obtained from the four-point dynamic susceptibility $\chi_4(t)$. This susceptibility is related to a space integral of a four-point density correlation function that quantifies correlations between relaxation processes at different points in space. It is assumed that the increase of the dynamic susceptibility signals the growth of the range of the correlations between relaxation processes. Furthermore, the four-point dynamic susceptibility can be used to discriminate between different theoretical approaches to glassy dynamics. For example, in a recent paper Toninelli et al. discussed predictions for $\chi_4(t)$ obtained from various theoretical approaches to glassy dynamics and compared these predictions with two different simulations of atomistic models of glass forming liquids.

A problem with the four-point susceptibility is that it is difficult to extract from experiments. This problem has been addressed in another recent paper Berthier et al. argued that the four-point susceptibility can be estimated using derivatives of an intermediate scattering function with respect to thermodynamic parameters such as temperature or density. Since the intermediate scattering function can be easily obtained from experiments, Ref. introduced a way to experimentally investigate the existence of a growing correlation length.

The goal of this contribution is twofold. First, we analyze the four-point susceptibility of the Kob-Andersen Lennard-Jones binary mixture undergoing Brownian dynamics. Second, we test the approximate estimate for the four point susceptibility proposed by Berthier et al.

The paper is organized as follows: in Sec. II we briefly review the details of the simulation; in Sec. III we define the four-point susceptibility and analyze the simulation results for this quantity; in Sec. IV we compare the four-point susceptibility obtained directly from computer simulations to the approximate expression derived by Berthier et al.; and in Sec. V we discuss our findings.

II. SIMULATION DETAILS

We simulated a binary mixture that was introduced by Kob and Andersen which consists of $N_A = 800$ particles of type A and $N_B = 200$ particles of type B. The interaction potential is $V_{\alpha\beta}(r) = 4\epsilon_{\alpha\beta}(\sigma_{\alpha\beta}/r)^12 - (\sigma_{\alpha\beta}/r)^6$, where $\alpha, \beta \in \{A, B\}$, $\epsilon_{AA} = 1.0$, $\sigma_{AA} = 1.0$, $\epsilon_{AB} = 1.5$, $\sigma_{AB} = 0.8$, $\epsilon_{BB} = 0.5$, and $\sigma_{BB} = 0.88$. The interaction potential was cut off at 2.5 $\sigma_{\alpha\beta}$. We used a cubic simulation cell with the box length of 9.4 $\sigma_{AA}$ with periodic boundary conditions.

We performed Brownian dynamics simulations. The equation of motion for the position of the $i$th particle of type $\alpha$, $\vec{r}_i^\alpha$, is

$$\dot{\vec{r}}_i^\alpha = \frac{1}{\xi_0} \vec{F}_i^\alpha + \vec{\eta}_i(t),$$

(1)

where $\xi_0$ is the friction coefficient of an isolated particle, $\xi_0 = 0.1$, and $\vec{F}_i^0$ is the force acting on the $i$th particle of type $\alpha$.

$$\vec{F}_i^\alpha = -\nabla_\alpha \sum_{j=1}^{2} V_{\alpha\beta} \left( |\vec{r}_i^\alpha - \vec{r}_j^\beta| \right)$$

(2)

with $\nabla_\alpha$ being the gradient operator with respect to $\vec{r}_i^\alpha$ (note that the term with $\beta = \alpha$ and $i = j$ has to be excluded from the double sum in Eq. (2)). In Eq. (1) the random noise $\vec{\eta}_i$ satisfies the fluctuation-dissipation theorem,

$$\langle \vec{\eta}_i(t) \vec{\eta}_j(t') \rangle = 2D_0 \delta(t - t') \delta_{ij} \mathbf{1}.$$  

(3)

Here $D_0$ is the diffusion coefficient of an isolated particle, $D_0 = k_B T/\xi_0$, where $k_B$ is Boltzmann’s constant. Furthermore, in Eq. (3) $\mathbf{1}$ is the unit tensor. The equations
of motion [13] allow for diffusive motion of the center of mass, thus all the results will be presented relative to the center of mass (i.e., momentary positions of all the particles are always relative to the momentary position of the center of mass [13]). The results are presented in terms of the reduced units with $\sigma_{AA}, \epsilon_{AA}, \epsilon_{AA}/k_B$, and $\sigma_{AA}^2\sigma_0/\epsilon_{AA}$ being the units of length, energy, temperature, and time, respectively. In these units the short-time self-diffusion coefficient is proportional to the temperature, thus the time is rescaled to a reduced time equal to $tD_0/\sigma_{AA}^2$ to facilitate comparisons with theoretical approaches that often assume temperature-independent short-time dynamics.

The equations of motion were solved using a Heun algorithm with a small time step of $5 \times 10^{-5}$. We simulated a broad range of temperatures $0.44 \leq T \leq 5.0$. Here we present results for the following temperatures: $T = 0.45, 0.47, 0.50, 0.55, 0.60, 0.65, 0.80$, and $1.0$. We ran equilibration runs and 4-6 production runs. The equilibration runs were typically twice to four times shorter than the production runs, and the latter were up to $1.2 \times 10^9$ time steps long for the lowest temperature discussed here, $T = 0.45$. The results presented are averages over the production runs.

### III. FOUR-POINT DYNAMIC SUSCEPTIBILITY

In the context of the dynamics of supercooled liquids, the four-point susceptibility has been introduced by Glotzer and collaborators [3]. The four-point susceptibility that we discuss in this paper is slightly different from that defined in Refs. [3]. We consider here the susceptibility which was extensively analyzed by Toninelli et al. [3], which is defined as the variance of the fluctuations of the self-intermediate scattering function.

We start with the definition of the self-intermediate scattering function, $F_s(k; t)$,

$$F_s(k; t) = \left\langle \frac{1}{N} \sum_i \cos \vec{k} \cdot \left[ \vec{r}_i(t) - \vec{r}_i(0) \right] \right\rangle. \quad (4)$$

Next, we define the fluctuation of the instantaneous value of the scattering function, $\delta F_s(\vec{k}; t)$,

$$\delta F_s(\vec{k}; t) = \frac{1}{N} \sum_i \cos \vec{k} \cdot \left[ \vec{r}_i(t) - \vec{r}_i(0) \right] - F_s(k; t). \quad (5)$$

The four-point susceptibility, $\chi_4(t)$, is then defined as

$$\chi_4(t) = N \left\langle \delta F_s(\vec{k}; t)\delta F_s(\vec{k}; t) \right\rangle. \quad (6)$$

The four-point susceptibility depends on the wave vector $\vec{k}$ (and it is sometimes denoted by $\chi_4(t)$). This wave vector is customarily fixed at the position of the maximum of the static structure factor [3].

The system considered in this paper is a two-component mixture, thus instead of one self-intermediate scattering function $F_s(k; t)$ we could introduce two different scattering functions involving particles $A$ and $B$. All the results presented in this paper concern the $A$ particles only, thus, e.g., sums in Eqs. (4-5) run only over the $A$ particles. Since we are not presenting any results for the $B$ particles we do not introduce additional sub-scripts or super-scripts indicating particle labels. The magnitude of the wave vector $\vec{k}$ is fixed at the position of the maximum of the partial structure factor of the $A$ particles, $|\vec{k}| = 7.25$.

The mode coupling theory of the glass transition was formulated by Götte and collaborators for Newtonian systems [3], and was later extended by Szamel and Löwen to Brownian systems [10]. The theory makes predictions for the self- and collective intermediate scattering functions. Biroli and Bouchaud [11] recently argued that the mode coupling theory could be understood as a dynamic mean-field theory, and that the usual mode coupling equations are saddle point equations obtained from an action functional. This new interpretation made it possible to calculate fluctuations of the order parameter (i.e., fluctuations of a two-point dynamic correlation function) from the inverse of the second derivative of the action functional. The details of the calculation have not been published, but the main predictions have already been discussed and compared with Newtonian dynamics simulations [3]. According to Biroli and Bouchaud, on the $\beta$ relaxation time scale the four-point susceptibility grows with time as a power law, $\chi_4(t) \propto t^{\alpha}$, with exponents equal to the standard mode coupling exponents $a$ and $b$ in the early and late $\beta$ regime, respectively. Furthermore, $\chi_4(t)$ reaches its maximum value, $\chi_4(t^*)$, on the time scale of the $\alpha$ relaxation time, $t^* \sim \tau_\alpha$, and the maximum value diverges upon approaching the mode coupling temperature, $\chi_4(t^*) \propto (T - T_\ast)^{-\gamma_1}$ with $\gamma_1 = 1$.

The authors of Refs. [3, 4] emphasized that these predictions are valid for Newtonian systems in the microcanonical (NVE) ensemble and hinted that they may be different in the canonical (NVT) ensemble. We present results obtained from Brownian dynamics simulations. In such simulations constant temperature is maintained automatically by the equations of motion. Thus, in principle it is not clear whether the predictions of Biroli and Bouchaud [11] are relevant for our simulation results. However, there are subtle theoretical arguments that suggest that the reverse is true [12]. In addition, at the level of the mode coupling equations, there is no difference between Newtonian systems in the NVE or NVT ensemble, and Brownian systems (beyond the microscopic time scale, i.e., on the time scales of the $\beta$ relaxation and longer) [10]. These reasons encouraged us to compare our results obtained from Brownian dynamics simulations to the predictions of Biroli and Bouchaud [11].

In Fig. [1] we show the general shape of the four-point susceptibility, $\chi_4(t)$, for several temperatures. The time dependence of $\chi_4(t)$ is similar to that obtained for Newto-
nian systems, see Figs. 4 and 5 of Ref. 3. In particular, it can be argued that there is power law like growth in time of the four-point susceptibility as it approaches its maximum value. This power law-like dependence will be further analyzed at the end of this section.

In Fig. 2 we present the results of some quantitative analysis of \( \chi_4(t) \). First, in Fig. 2a we show the temperature dependence of the its maximum value, \( \chi_4(t^*) \). We plot the maximum value vs. \( \epsilon = (T-T_c)/T_c \) where \( T_c = 0.435 \) is the standard value of the mode coupling temperature for the Kob-Andersen Lennard-Jones binary mixture 3. In an intermediate range of temperatures, \( \chi_4(t^*) \) grows as a power law with decreasing \( (T-T_c)/T_c \), \( \chi_4(t^*) \propto \epsilon^{-\gamma_1} \). The exponent obtained from the fit, \( \gamma_1 = 0.995 \pm 0.05 \), is very close to the theoretical prediction of Biroli and Bouchaud, \( \gamma_1^{th} = 1 \). It should be noted that the range of reduced temperatures for which the power law dependence of \( \chi_4(t^*) \) is observed coincides with the range of reduced temperatures for which mode coupling theory was found to correctly describe the time evolution of the self-intermediate scattering function and the mean square displacement 13. In Fig. 2b, we compare the temperature dependence of the time at which \( \chi_4(t) \) reaches its maximum value, \( t^* \), with that of the \( \alpha \) relaxation time, \( \tau_\alpha \). The latter time is defined as the time at which the self-intermediate scattering function decays to \( e^{-1} \) of its initial value, \( F^s(q, \tau_\alpha) = e^{-1} \). We find that \( \tau_\alpha \) is very close and have the same temperature dependence. In particular, in the same intermediate range of temperatures these times grow according to power laws with decreasing \( (T-T_c)/T_c \), \( \tau_\alpha \propto \epsilon^{-\gamma_\alpha} \), \( \tau_\alpha \propto \epsilon^{-\gamma} \). The exponents obtained from the fits, \( \gamma_\alpha = 2.27 \pm 0.04 \) and \( \gamma = 2.31 \pm 0.02 \) are very close. The numerical solution of the mode coupling equations predicts a slightly higher value of the scaling exponent for \( \tau_\alpha \), \( \gamma_\alpha^{th} = 2.46 \) 13. Finally, in Fig. 2c we plot \( \chi_4(t^*) \) vs. \( t^* \). We find that slight deviations from power laws that are visible in Figs. 2a and 2b are magnified in Fig. 2c. Nevertheless we can still fit a power law \( \chi_4(t^*) \propto (t^*)^{\gamma_2} \) with the exponent \( \gamma_2 = 2.31 \pm 0.21 \).

In Fig. 3 we address the question of the power law dependence of the four-point susceptibility on time. Instead of trying to fit power laws to \( \chi_4(t) \) over some specific time intervals, we have numerically calculated the derivative of \( \ln \chi_4(t) \) with respect to \( \ln t \), \( d \ln \chi_4(t)/d \ln t = (t/\chi_4(t))d\chi_4(t)/dt \). For any time interval over which

\[ \gamma = \ln(\ln t) \]

FIG. 1: Time dependence of the four-point susceptibility \( \chi_4(t) \) plotted vs. \( tD_0/\sigma_{AA}^2 \) for \( T = 1.0, 0.80, 0.60, 0.55, 0.50, 0.47, \) and 0.45 (left to right).

FIG. 2: (a) Temperature dependence of the maximum value of the four-point susceptibility: \( \chi_4(t^*) \) plotted vs. \( \epsilon = (T-T_c)/T_c \). Solid line is the power law fit \( \chi_4(t^*) \propto \epsilon^{-\gamma_1} \) with \( \gamma_1 = 0.995 \). (b) Temperature dependence of the time at which \( \chi_4(t) \) reaches its maximum value, \( t^* \) (circles), and the \( \alpha \) relaxation time, \( \tau_\alpha \) (squares). Both times are plotted vs. \( \epsilon = (T-T_c)/T_c \). The solid line and the dashed line are power law fits \( t^* \propto \epsilon^{-\gamma} \) with \( \gamma = 2.27 \) and \( \tau_\alpha \propto \epsilon^{-\gamma} \) with \( \gamma = 2.31 \), respectively. (c) The maximum value of the four-point susceptibility, \( \chi_4(t^*) \), plotted vs. \( t^* \). The solid line is the power law fit \( \chi_4(t^*) \propto (t^*)^{\gamma_2} \) with \( \gamma_2 = 2.31 \).
\( \chi_4(t) \) depends on \( t \) in a power law fashion, \( d \ln \chi_4(t) / d \ln t \) should have a constant value (a plateau). On the basis of the predictions of Ref. [11] we expect two different plateaus in our data. A plateau in the early-\( \beta \) regime and another one in the late-\( \beta \) regime, i.e. on approaching the maximum of \( \chi_4(t) \) (additional plateaus are predicted for shorter times, and we do not analyze them here). However, the simulations agree with mode coupling predictions only over a restricted range of temperatures. It is not clear whether these two time scales are well separated over this temperature range, thus it is not clear if we could see two different plateaus.

Indeed, Fig. 3 does not show well developed plateaus. However, we notice that \( \chi_4(t) \) grows with \( t \) with an exponent of approximately \( b = 0.8 \) upon approaching its maximum for all temperatures. This exponent is comparable to the exponents obtained from Newtonian dynamics simulations [2]. It is quite a bit higher than the exponent obtained from the mode coupling theory \( b^{MC} = 0.62 \) [14].

IV. TEST OF THE ESTIMATE OF THE FOUR-POINT SUSCEPTIBILITY PROPOSED BY BERTHIER ET AL.

The four-point susceptibility can be easily obtained from computer simulations, but it is not readily determined from experiments. To address this problem Berthier et al. [4] proposed an approximate estimate for the four-point susceptibility in terms of the derivative of the self-intermediate scattering function with respect to temperature, \( \chi_T(t) \),

\[
\chi_T(t) = \frac{\partial F_4(k; t)}{\partial T}.
\]

The starting point of their argument was a fluctuation-dissipation relation (Eq. (2) of Ref. [2]). If we naively adopted this relation to our system (i.e. Brownian dynamics, NVT ensemble) we would get

\[
k_B T^2 \chi_T(t) = \left\langle \delta F_4(k; t) \delta V(0) \right\rangle.
\]

Here \( \delta V(t) \) denotes the instantaneous fluctuation of the total potential energy,

\[
V(t) = \sum_{i,j} \sum_{\alpha\beta} V_{\alpha\beta} \left( |\mathbf{r}_i^\alpha(t) - \mathbf{r}_j^\beta(t)| \right),
\]

\[
\delta V(t) = V(t) - \left\langle V(t) \right\rangle.
\]

The derivative with respect to the temperature in Eq. (11), Eq. (8) and in the remainder of this section have to be calculated while keeping the short-time diffusion coefficient \( D_0 \) constant since our short-time diffusion coefficient is proportional to the temperature.

The main analytical result obtained by Berthier et al. was an exact lower bound for \( \chi_4(t) \) in terms of \( \chi_T(t) \) (Eq. (5) of Ref. [4]). Naively applying this result to our Brownian system we would get

\[
\chi_4(t) \geq \frac{k_B}{c_V^{pot}} T^2 \chi_T^2(t).
\]

Here \( c_V^{pot} \) is the potential contribution to the constant volume specific heat per particle.

Berthier et al. found that the difference between the right and left sides of relation (11) diminishes with decreasing temperature. On this basis, they proposed using the right-hand-side of Eq. (11) as an approximate estimate for the four-point susceptibility. Furthermore, they showed that this estimate can be easily calculated using either experimental or simulational results for the self-intermediate scattering function.

Both the fluctuation-dissipation relation and the bound derived by Berthier et al. are valid only if the equations of motion do not involve the temperature. The only place where the temperature can enter is the initial condition which is given by the canonical distribution. This is true neither for the usual NVT computer simulations, where the temperature enters into the equations of motion via a thermostat, nor for our Brownian dynamics simulations, where the temperature enters via the noise strength. Thus we have no arguments in favor of either the fluctuation-dissipation relation (8) or the bound (11) for our system. In fact, we show explicitly in Fig. 4 that the relation (8) is violated in Brownian systems. On the other hand we do not have strong numerical evidence for the violation of the inequality (11).

Figure 5 suggests that this inequality is violated, but the extent of the violation is smaller than the error bars.

We should mention that there is a related, exact bound for the four-point susceptibility that follows from the Cauchy-Schwarz inequality (see footnote 22 of Ref. [2]):

\[
\chi_4(t) \geq \frac{\left\langle \delta F_4(k; t) \delta V(0) \right\rangle^2}{k_B T^2 c_V^{pot}}.
\]
However, as we show in Fig. 5 this bound does not lead to a useful estimate for the four-point susceptibility.

Berthier et al. also gave a different, general argument leading to an estimate for the four-point susceptibility in terms of the right-hand-side of inequality (11). This argument can be easily adopted for our system. If we assume that the main source of fluctuations of the instantaneous expression for the scattering function is the potential energy, we get

\[
\delta F_s(\vec{k}; t) \approx \frac{\partial F_s(k; t) \delta V(0)}{\partial T} \frac{\delta V(0)}{N c_V^\text{pot}}.
\]  

(13)

Eq. (13) leads immediately to

\[
\chi_4(t) \approx \frac{k_B T^2 \chi_T^2(t)}{c_V^\text{pot}}.
\]

(14)

In Fig. 5 we show the time dependence of both sides of the relation (14) at three representative temperatures. While the approximation (14) is inaccurate, it becomes better, especially near the maximum value of \(\chi_4(t)\), when the temperature approaches the mode coupling temperature.
**V. CONCLUSIONS**

We performed a quantitative analysis of the four-point susceptibility $\chi_4(t)$ of the Kob-Andersen Lennard-Jones binary mixture. We compared the results of Brownian dynamics computer simulations to predictions obtained from a recent re-formulation of the mode coupling theory. We did not compare computer simulation results to other approaches to glassy dynamics (e.g., to the predictions obtained using facilitated kinetic Ising models) because these other approaches are more concerned with a temperature range lower than the one accessible in our simulations.

We found that some of the mode coupling predictions agree with our simulation results. Most notably, the height of the maximum of the four-point susceptibility grows upon approaching the mode coupling temperature from above in the way predicted by the theory. Moreover, the time at which $\chi_4(t)$ reaches its maximum value has the same temperature dependence as the $\alpha$ relaxation time. For each of these two quantities, which are derived from a four-point function, the power law dependence on $(T-T_c)/(T_c)$ is obeyed over the same temperature range as for quantities derived from two-point functions, e.g., the $\alpha$ relaxation time and the self-diffusion coefficient.

On the other hand, we found some disagreement with the theory with regard to the power law dependence of $\chi_4(t)$ on time. We found that upon approaching its maximum value $\chi_4(t)$ grows with time with an effective exponent of about 0.8. This exponent is quite a bit larger than the exponent predicted by the theory. It should be recalled that the exponent predicted by the theory is in turn larger than the one obtained by fitting a formula inspired by the mode coupling theory to the (two-point) self-intermediate scattering function. Hence, our understanding of the connection between the time dependence of the four-point susceptibility and the time dependence of two-point functions seems incomplete.

Finally, we tested the approximate estimate of the four-point susceptibility in terms of the temperature dependence of the mode coupling temperature.

**FIG. 6:** Temperature dependence of the maximum value of $k_B T^2 \chi_T^2(t)/c_V^{pot}$ (closed circles) compared to that of the maximum value of $\chi_4(t)$ (open circles).

**FIG. 7:** Comparison of the derivative $d\ln \chi_4(t)/d\ln t = (t/\chi_4(t))d\chi_4(t)/dt$ (open circles) and the derivative $d\ln k_B T^2 \chi_T^2(t)/c_V^{pot}/d\ln t = 2(t/\chi_T(t))d\chi_T(t)/dt$ (closed circles) at two temperatures: (a) $T=0.55$, and (b) $T=0.45$. 

![Temperature dependence of the maximum value of $k_B T^2 \chi_T^2(t)/c_V^{pot}$](image-url)
derivative of the self-intermediate scattering function. We found that the estimate becomes accurate around the peak of $\chi_4(t)$ upon approaching the mode coupling temperature. However, the temperature dependence of the maximum value of $\chi_4(t)$ is weaker than that of the approximate estimate. Moreover, the time dependence of $\chi_4(t)$ differs from that of the estimate even near the mode coupling temperature.

We should emphasize that all our computer simulation results were obtained using Brownian dynamics (and, therefore, NVT ensemble). It is possible that results obtained from Newtonian dynamics computer simulations would agree better (or worse) with theoretical predictions.

Acknowledgments

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