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

Grzegorz Szamel and Elijah Flenner

Department of Chemistry, Colorado State University, Fort Collins, Colorado 80523, USA

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)].

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 [1]. 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 [2], and there is evidence that there is a dynamic correlation length that slowly grows 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 [3], 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 [4]. 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. [4] 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 [5] Lennard-Jones binary mixture undergoing Brownian dynamics. Second, we test the approximate estimate for the four-point susceptibility proposed by Berthier *et al.* [4].

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 ith particle of type α, $\vec{r}_i^\alpha$, is

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

where ξ_0 is the friction coefficient of an isolated particle, $\xi_0 = 1.0$, and $\vec{F}_i^\alpha$ is the force acting on the ith particle of type α,

$$\vec{F}_i^\alpha = -\nabla_i^\alpha \sum_j \sum_{\beta=1}^2 V_{\alpha\beta}(|\vec{r}_i^\alpha - \vec{r}_j^\beta|) \tag{2}$$

with ∇_i^α 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

$$\langle \vec{\eta}_i(t)\vec{\eta}_j(t') \rangle = 2D_0 \delta(t - t')\delta_{ij}\mathbf{1}. \tag{3}$$

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 (1)–(3) 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 [6]). The results are presented in terms of the reduced units with σ_{AA}, ϵ_{AA}, ϵ_{AA}/k_B, and $\sigma_{AA}^2 \xi_0/\epsilon_{AA}$ 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/σ_{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 5×10^{-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 1.2×10^9 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 [7]. The four-point susceptibility that we discuss in this paper is slightly different from that defined in Refs. [7]. We consider here the susceptibility that 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, $F_s(k;t)$,

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

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

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

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

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

The four-point susceptibility depends on the wave vector $\vec{k}$ [and it is sometimes denoted by $\chi_{\vec{k}}(t)$]. This wave vector is customarily fixed at the position of the maximum of the static structure factor [8].

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) and (5) run only over the A particles. Since we are not presenting any results for the B particles, we do not introduce additional subscripts or superscripts 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 Götze and collaborators for Newtonian systems [9], and was later extended by Szamel and Löwen to Brownian systems [10]. The theory makes predictions for the self-intermediate and collective intermediate scattering functions.
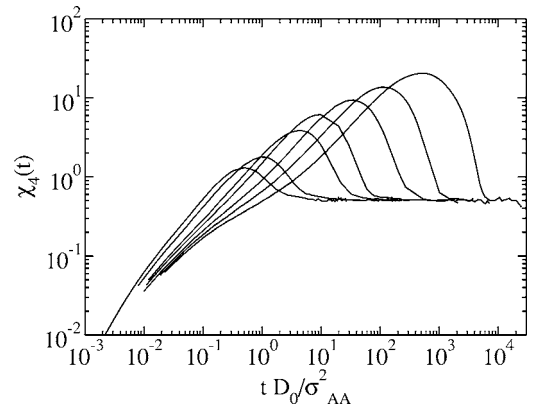

FIG. 1. Time dependence of the four-point susceptibility $\chi_4(t)$ plotted vs tD_0/σ^2 for $T = 1.0$, 0.80, 0.60, 0.55, 0.50, 0.47, and 0.45 (left to right).

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 β relaxation time scale the four-point susceptibility grows with time as a power law, $\chi_4(t) \propto t^\mu$, with exponents equal to the standard mode coupling exponents a and b in the early and late β regime, respectively. Furthermore, $\chi_4(t)$ reaches its maximum value $\chi_4(t^*)$ on the time scale of the α relaxation time $t^* \sim \tau_\alpha$, and the maximum value diverges upon approaching the mode coupling temperature $\chi_4(t^*) \propto (T - T_c)^{-\gamma_1}$ 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.[1] 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 β 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 depen-

---

[1]Ensemble dependence of the four-point susceptibility has also been discussed in the appendix of the cond-mat report [8].
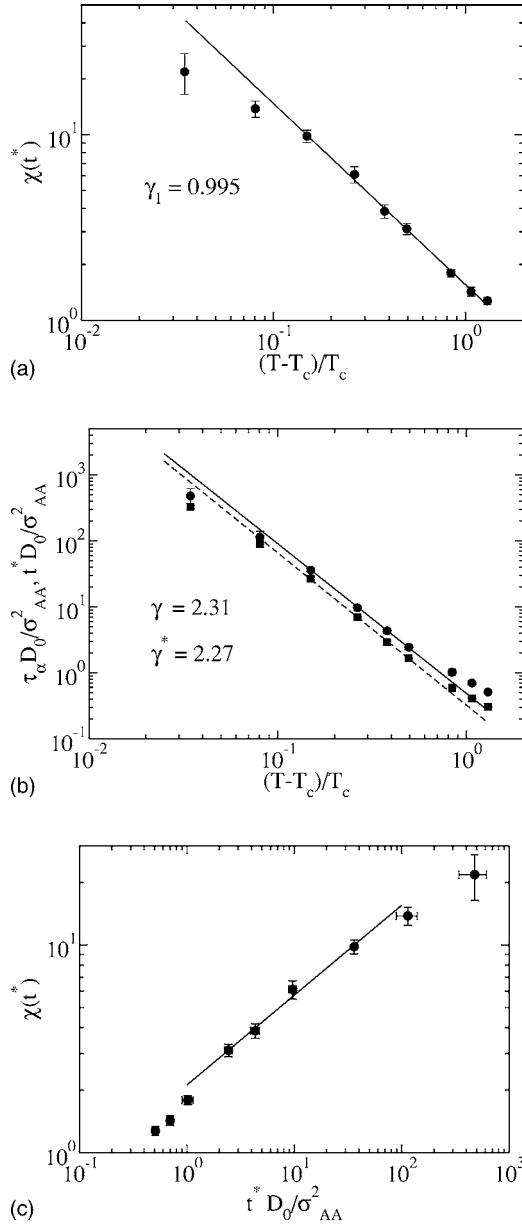
(a)

(b)

(c)

FIG. 2. (a) Temperature dependence of the maximum value of the four-point susceptibility: $\chi_4(t^*)$ plotted vs $\epsilon=(T-T_c)/T_c$. The 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 α relaxation time, τ_α (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 $\chi_4(t^*) \propto (t^*)^{1/\gamma_2}$ with $\gamma_2=2.31$.

dence of $\chi_4(t)$ is similar to that obtained for Newtonian 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. 2(a) we show the temperature
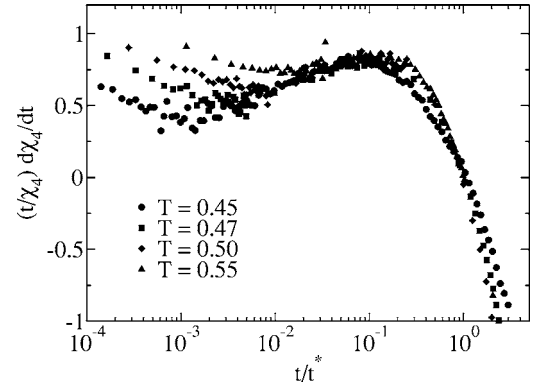

FIG. 3. Derivative of $\ln \chi_4(t)$ with respect to $\ln t$, $d \ln \chi_4(t)/d \ln t = [t/\chi_4(t)]d\chi_4(t)/dt$, plotted vs t/t^* for $T=0.45$, 0.47, 0.50, and 0.55 (left to right).

dependence of its maximum value $\chi_4(t^*)$. We plot the maximum value versus $\epsilon=(T-T_c)/T_c$ where $T_c=0.435$ is the standard value of the mode coupling temperature for the Kob-Andersen Lennard-Jones binary mixture [5]. 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, $\gamma_1=0.995\pm0.05$, 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. 2(b) we compare the temperature dependence of the time at which $\chi_4(t)$ reaches its maximum value t^* with that of the α relaxation time τ_α. 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 t^* and τ_α are 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 $(T-T_c)/T_c$, $t^* \propto \epsilon^{-\gamma^*}$, $\tau_\alpha \propto \epsilon^{-\gamma}$. The exponents obtained from the fits, $\gamma^*=2.27\pm0.04$ and $\gamma=2.31\pm0.02$ are very close. The numerical solution of the mode-coupling equations predicts a slightly higher value of the scaling exponent for τ_α, $\gamma^{th}=2.46$ [13]. Finally, in Fig. 2(c) we plot $\chi_4(t^*)$ vs t^*. We find that slight deviations from power laws that are visible in Figs. 2(a) and 2(b) are magnified in Fig. 2(c). Nevertheless, we can still fit a power law $\chi_4(t^*) \propto (t^*)^{1/\gamma_2}$ with the exponent $\gamma_2=2.31\pm0.21$.

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 $\chi_4(t)$ 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-β regime and another one in the

late-β 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 [3]. It is quite a bit higher than the exponent obtained from the mode coupling theory $b^{th}=0.62$ [14]. Additionally, it seems that the time scale on which the exponent $b=0.8$ is observed, scales with the time at which $\chi_4(t)$ reaches its maximum value t^* rather than with the characteristic time of the β relaxation τ_β [note that as we show in Fig. 2(b), t^* has the same temperature dependence as the α relaxation time τ_α].

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 $\chi_T(t)$,

$$\chi_T(t) = \frac{\partial F_s(k;t)}{\partial T}. \tag{7}$$

The starting point of their argument was a fluctuation-dissipation relation [Eq. (2) of Ref. [4]]. If we naively adopted this relation to our system (i.e., Brownian dynamics, *NVT* ensemble) we would get

$$k_B T^2 \chi_T(t) = \langle \delta F_s(\vec{k};t) \delta V(0) \rangle. \tag{8}$$

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

$$V(t) = \sum_{i,j}{}' \sum_{\alpha\beta} V_{\alpha\beta}(|\vec{r}_i^\alpha(t) - \vec{r}_j^\beta(t)|), \tag{9}$$

$$\delta V(t) = V(t) - \langle V(t) \rangle. \tag{10}$$

The derivative with respect to the temperature in Eqs. (7) and (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

$$\chi_4(t) \geq \frac{k_B}{c_V^{pot}} T^2 \chi_T^2(t). \tag{11}$$

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(c) 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. [4]):

$$\chi_4(t) \geq \frac{\langle \delta F_s(\vec{k};t) \delta V(0) \rangle^2}{k_B T^2 c_V^{pot}}. \tag{12}$$

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

$$\delta F_s(\vec{k};t) \approx \frac{\partial F_s(k;t)}{\partial T} \frac{\delta V(0)}{N c_V^{pot}}. \tag{13}$$

Equation (13) leads immediately to

$$\chi_4(t) \approx \frac{k_B}{c_V^{pot}} T^2 \chi_T^2(t). \tag{14}$$

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.

Figure 6 compares the temperature dependence of the maximum value of the right-hand side of the relation (14) to the temperature dependence of the maximum value of the four-point susceptibility. The difference between the two diminishes with decreasing temperature. However, it is clear that the temperature dependence of these quantities is different. In particular, one should be cautious when using the temperature dependence of the estimate (14) to obtain the
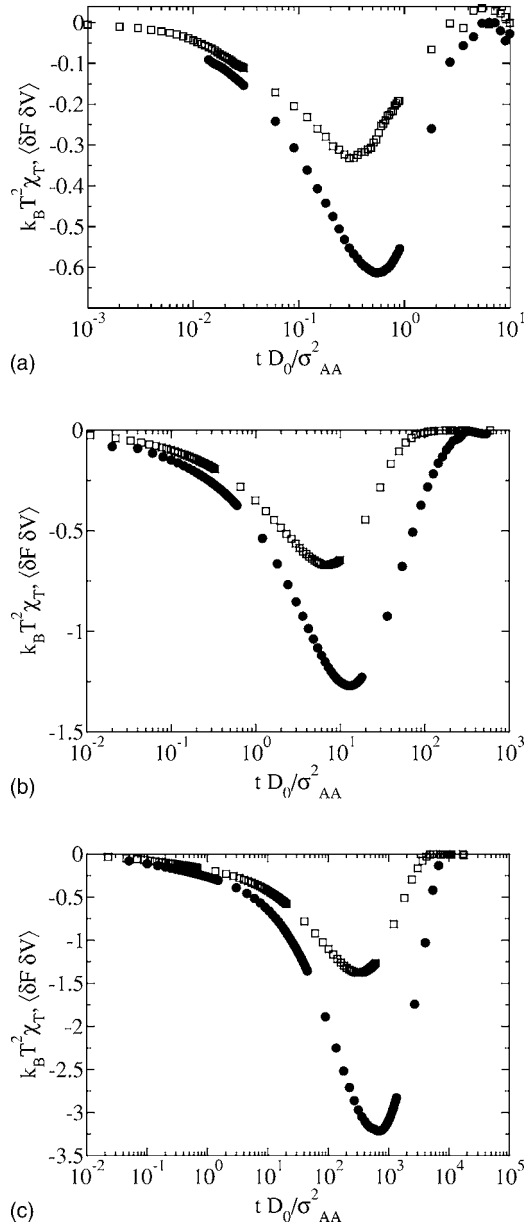
FIG. 4. Comparison of $k_B T^2 \chi_T(t) = k_B T^2 \partial F(k;t)/\partial T$ (closed circles) with $\langle \delta F_s(\vec{k};t) \delta V(0) \rangle$ (open squares) at three representative temperatures: (a) $T=1.00$, (b) $T=0.55$, and (c) $T=0.45$.

temperature dependence of the dynamic correlation length, at least for temperatures above the mode-coupling temperature.

Finally, Fig. 7 compares the derivative of the logarithm of the four-point susceptibility, $\ln \chi_4(t)$, with respect to the logarithm of time, $\ln t$ and the derivative of the logarithm of the estimate (14), $\ln k_B T^2 \chi_T^2(t)/c_V^{pot} = 2 \ln \chi_T(t) + \text{const.}$, with respect to $\ln t$. We find that the estimate (14) approaches its maximum value with a power-law exponent that is approximately equal to one and thus somewhat higher than the power-law exponent describing $\chi_4(t)$'s approach to its maximum value. Moreover, comparing Figs. 7(a) and 7(b) we find that the difference between exponents obtained from the estimate (14) and from the four-point susceptibility does not seem to diminish with decreasing temperature.
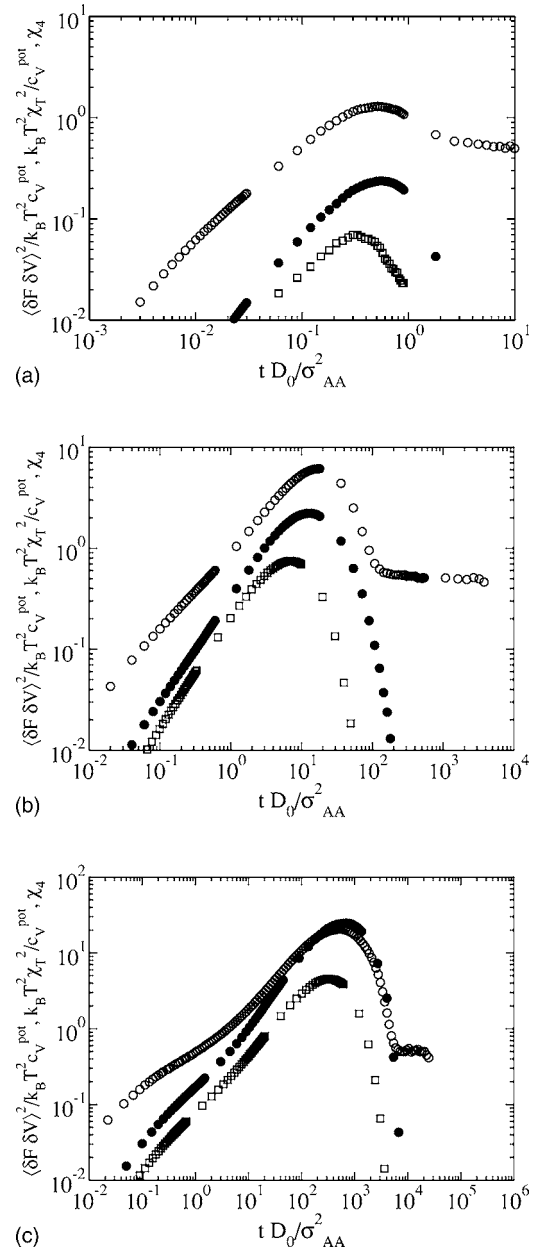

FIG. 5. Comparison of $\chi_4(t)$ (open circles) with $k_B T^2 \chi_T^2(t)/c_V^{pot}$ (closed circles) and $\langle \delta F_s(\vec{k};t) \delta V(0) \rangle^2/(k_B T^2 c_V^{pot})$ (open squares) at three representative temperatures: (a) $T=1.00$, (b) $T=0.55$, and (c) $T=0.45$.

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 reformulation 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 [15,16]) 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
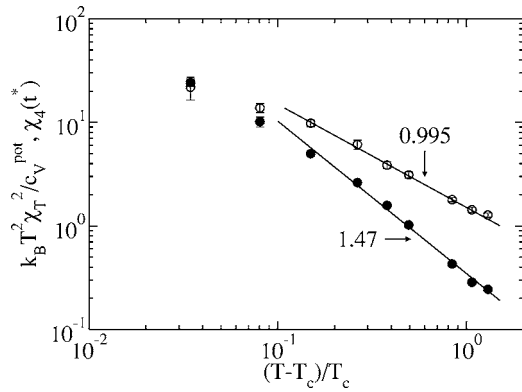
FIG. 6. Temperature dependence of the maximum value of $k_B T^2 \chi_T^2(t)/c_V^{pot}$ (closed circles) compared to that of the maximum value of $\chi_4(t)$ (open circles). Solid lines are power law fits; note that the exponent describing the temperature dependence of $k_B T^2 \chi_T^2(t)/c_V^{pot}$ is significantly larger than that describing the temperature dependence of χ_4.

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 α 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 α 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 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
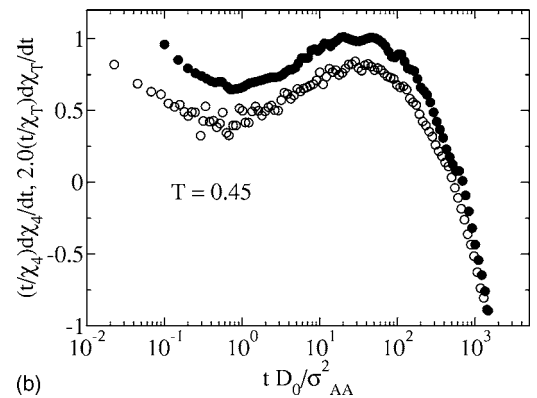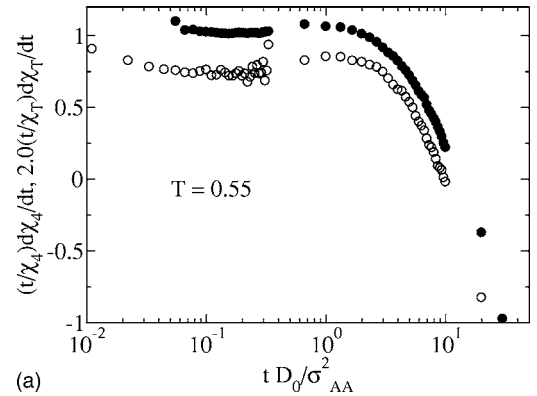

FIG. 7. Comparison of the derivative $d \ln \chi_4(t)/d \ln t = [t/\chi_4(t)] d\chi_4(t)/dt$ (open circles) and the derivative $d \ln k_B T^2 \chi_T^2(t)/c_V^{pot}/d \ln t = 2[t/\chi_T(t)] d\chi_T(t)/dt$ (closed circles) at two temperatures: (a) $T=0.55$ and (b) $T=0.45$.

temperature dependence of the maximum value of $\chi_4(t)$ 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.

[1] See, e.g., P. G. Debenedetti and F. H. Stillinger, Nature (London) **410**, 259 (2001).

[2] J.-P. Bouchaud and G. Biroli, Phys. Rev. B **72**, 064204 (2005).

[3] C. Toninelli, M. Wyart, L. Berthier, G. Biroli, and J.-P. Bouchaud, Phys. Rev. E **71**, 041505 (2005).

[4] L. Berthier, G. Biroli, J.-P. Bouchaud, L. Cipelletti, D. El Masri, D. L'Hote, F. Ladieu, and M. Pierno, Science **310**, 1797 (2005).

[5] W. Kob and H. C. Andersen, Phys. Rev. Lett. **73**, 1376 (1994); Phys. Rev. E **51**, 4626 (1995); **52**, 4134 (1995).

[6] The alternative used in other stochastic dynamics simulations is to explicitly modify the noise in such a way that the center of mass does not move [W. Kob (private communication)].

[7] S. Franz, C. Donati, G. Parisi, and S. C. Glotzer, Philos. Mag.

B **79**, 1827 (1999); C. Donati, S. Franz, S. C. Glotzer, and G. Parisi, J. Non-Cryst. Solids **307**, 215 (2002); S. C. Glotzer, V. N. Novikov, and T. B. Schrøder, J. Chem. Phys. **112**, 509 (2000).

[8] Recently a cond-mat report was posted (D. Chandler, J. P. Garrahan, R. L. Jack, L. Maibaum, A. C. Pan, eprint cond-mat/0605084) which discusses the dependence of the four-point susceptibility $\chi_{\vec{k}}(t)$ on the magnitude of the wave vector $\vec{k}$.

[9] W. Götze, in *Liquids, Freezing and Glass Transition*, edited by J. P. Hansen, D. Levesque, and J. Zinn-Justin (North-Holland, Amsterdam, 1991).

[10] G. Szamel and H. Löwen, Phys. Rev. A **44**, 8215 (1991).

[11] G. Biroli and J.-P. Bouchaud, Europhys. Lett. **67**, 21 (2004).

[12] L. Berthier and G. Biroli (private communication).

[13] E. Flenner and G. Szamel, Phys. Rev. E **72**, 031508 (2005).

[14] We obtained $b^{th}=0.62$ by fitting the asymptotic functional form predicted by the mode-coupling theory to the solution of the full mode-coupling equations [13]. This value is very close to $b^{th}=0.63$ quoted in Ref. [3].

[15] J. P. Garrahan and D. Chandler, Phys. Rev. Lett. **89**, 035704 (2002); Proc. Natl. Acad. Sci. U.S.A. **100**, 9710 (2003).

[16] L. Berthier and J. P. Garrahan, J. Chem. Phys. **119**, 4367 (2003); Phys. Rev. E **68**, 041201 (2003).