Time-dependent, four-point density correlation function description of dynamical heterogeneity and decoupling in supercooled liquids

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Dynamical heterogeneity and the decoupling of diffusion and relaxation in a supercooled liquid is investigated via a time-dependent, four-point density correlation function. We show that the main contribution to the corresponding generalized susceptibility $\chi_4(t)$ in a molecular dynamics simulation of a Lennard-Jones liquid arises from spatial correlations between temporarily localized (''caged'') particles. By comparing $\chi_4(t)$ with a generalized susceptibility $\chi_M(t)$ related to a correlation function for the squared particle displacements, we demonstrate a connection between dynamical heterogeneity and the decoupling of relaxation and diffusion. © *2000 American Institute of Physics.* [S0021-9606(00)52402-7]

Spatially heterogeneous dynamics (''dynamical heterogeneity'') in otherwise homogeneous supercooled, glass-forming liquids is now well established in experiments[1–6] close to the glass transition temperature T_g, and this heterogeneity is even apparent at higher temperatures above the mode coupling[7] temperature T_c in simulations.[8–12] For example, recent studies[9–12] of the dynamics of supercooled, glass-forming polymeric and binary simple liquids in terms of the correlations of monomer or particle displacements revealed the dynamical heterogeneity of these liquids and a rapidly growing range of correlated motion on cooling towards T_c. At the same time, the decoupling of translational diffusion and relaxation as well as translational and rotational diffusion in these fluids is also well-known, and simulations show[12–15] that this decoupling begins well above T_c where dynamical heterogeneity first appears. Several authors have argued that the decoupling of diffusion and relaxation is a direct result of dynamical heterogeneity, with the slowest particles dominating structural relaxation and the fastest particles dominating diffusion.[1,4,5,11,12,16,17]

In this letter we use a four-point time correlation function of the density to probe dynamical heterogeneity in a glass-forming liquid, and elucidate the connection between this heterogeneity and the decoupling of bulk transport processes. This four-point function was first investigated in a supercooled liquid by Dasgupta *et al.*,[18] and recently Donati *et al.*[19] have demonstrated analytically and computationally the interesting behavior of the related generalized four-point susceptibility $\chi_4(t)$ (defined below). As shown in Ref. 19, $\chi_4(t)$ can be represented in terms of the fluctuations of an ''order parameter'' that is a bilinear, time-dependent product of densities. Here we show that the self-part of $\chi_4(t)$ is directly related to spatial correlations between temporarily localized particles, while the distinct-part is related to the correlated motion of particles into positions previously occupied

by neighboring particles. We evaluate these quantities for a cold Lennard-Jones (LJ) liquid, and show that in this system $\chi_4(t)$ is dominated by growing spatial correlations between localized particles. We then compare the behavior of $\chi_4(t)$ with a generalized time-dependent susceptibility related to a correlation function of squared particle displacements. From these two quantities we find two *different* characteristic time scales: the time scale on which temporarily localized particles are most spatially correlated scales with temperature like the structural relaxation time, while the time scale on which the correlation between squared particle displacements is strongest scales like the inverse diffusion coefficient. In this way, we demonstrate that the decoupling of diffusion and relaxation in this model liquid arises from dynamical heterogeneity.

Consider a liquid of N particles in a volume V, with density $\rho(\mathbf{r},t) = \sum_{i=1}^{N} \delta(\mathbf{r} - \mathbf{r}_i(t))$. The simplest density correlation function that contains information on correlated particle motion is fourth-order. We write this function in terms of the deviations of $\rho(\mathbf{r},t)$ from its average value, $\Delta\rho(\mathbf{r},t) = \rho(\mathbf{r},t) - \rho_0$, where $\rho_0 = \langle\rho\rangle = N/V$, and $\langle\cdots\rangle$ denotes an ensemble average:

$$
\begin{aligned}
\mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t) = &\langle \Delta\rho(\mathbf{r}_1,0)\Delta\rho(\mathbf{r}_1,t)\Delta\rho(\mathbf{r}_2,0)\Delta\rho(\mathbf{r}_2,t)\rangle \\
&- \langle \Delta\rho(\mathbf{r}_1,0)\Delta\rho(\mathbf{r}_1,t)\rangle \\
&\times \langle \Delta\rho(\mathbf{r}_2,0)\Delta\rho(\mathbf{r}_2,t)\rangle.
\end{aligned} \tag{1}
$$

Terms involving one position only are subtracted in Eq. (1) since they contain no information on spatial correlations of particle motions. $\mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t)$ can be written,

$$
\mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t) = \mathcal{G}_4(\mathbf{r}_1,\mathbf{r}_2,t) + \Delta\mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t),
$$

where the two-point, two-time, fourth-order correlation function of densities $\mathcal{G}_4$ is defined as[18,19]

$$
\begin{aligned}
\mathcal{G}_4(\mathbf{r}_1,\mathbf{r}_2,t) \equiv &\langle \rho(\mathbf{r}_1,0)\rho(\mathbf{r}_1,t)\rho(\mathbf{r}_2,0)\rho(\mathbf{r}_2,t)\rangle \\
&- \langle \rho(\mathbf{r}_1,0)\rho(\mathbf{r}_1,t)\rangle\langle \rho(\mathbf{r}_2,0)\rho(\mathbf{r}_2,t)\rangle.
\end{aligned}
$$

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$\Delta \mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t)$ consists of second- and third-order correlation functions of density. A straightforward calculation shows that $\int\int d\mathbf{r}_1\, d\mathbf{r}_2 \Delta \mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t)$ vanishes by symmetry, and as a result, the volume integrals of $\mathcal{F}_4(\mathbf{r}_1,\mathbf{r}_2,t)$ and $\mathcal{G}_4(\mathbf{r}_1,\mathbf{r}_2,t)$ are equal to each other and correspond to the *same* generalized susceptibility $\chi_4^0(t)$.

$$\chi_4^0(t)=\frac{\beta V}{N^2}\int\int d\mathbf{r}_1\, d\mathbf{r}_2 \mathcal{G}_4(\mathbf{r}_1,\mathbf{r}_2,t).$$

It is straightforward to show that $\chi_4^0(t)$ can be written as

$$\chi_4^0(t)=\frac{\beta V}{N^2}[\langle Q_0^2(t)\rangle - \langle Q_0(t)\rangle^2], \tag{2}$$

where $\beta = 1/k_B T$, and the time-dependent ''order parameter'' $Q_0(t)$ equals

$$Q_0(t)=\int d\mathbf{r}\, \rho(\mathbf{r},0)\rho(\mathbf{r},t)=\sum_{i=1}^N \sum_{j=1}^N \delta(\mathbf{r}_i(0)-\mathbf{r}_j(t)). \tag{3}$$

In a simulation, $Q_0(t)$ is numerically ill-defined (for a finite system) since the probability that particle j exactly replaces particle i is infinitely small. Following Parisi,[20] we therefore modify $Q_0(t)$ by an ''overlap'' function $w(r)$ that is unity inside a region of size a and zero otherwise, where a is taken on the order of a particle diameter.[19,21] This leads to an a-dependent counterpart to $Q_0(t)$,

$$Q(t)=\int d\mathbf{r}_1\, d\mathbf{r}_2 \rho(\mathbf{r}_1,0)\rho(\mathbf{r}_2,t)w(|\mathbf{r}_1-\mathbf{r}_2|)$$

$$=\sum_{i=1}^N \sum_{j=1}^N \int d\mathbf{r} w(|\mathbf{r}|)\delta(\mathbf{r}+\mathbf{r}_i(0)-\mathbf{r}_j(t))$$

$$=\sum_{i=1}^N \sum_{j=1}^N w(|\mathbf{r}_{ij}-\vec{\mu}_j|), \tag{4}$$

where $\mathbf{r}_{ij}\equiv\mathbf{r}_i(0)-\mathbf{r}_j(0)$ and $\vec{\mu}_i\equiv\mathbf{r}_i(t)-\mathbf{r}_i(0)$ is the displacement of particle i during the time interval from zero to t. We choose $a=0.3\sigma_{AA}$ as in Ref. 19.

Replacing $Q_0(t)$ in Eq. (2) by $Q(t)$ yields

$$\chi_4(t)=\frac{\beta V}{N^2}[\langle Q^2(t)\rangle - \langle Q(t)\rangle^2], \tag{5}$$

which gives the following expression[19] for $\chi_4(t)$ in terms of the four-point correlation function $G_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4,t)$:

$$\chi_4(t)=\frac{\beta V}{N^2}\int d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3\, d\mathbf{r}_4 w(|\mathbf{r}_1-\mathbf{r}_2|)w(|\mathbf{r}_3-\mathbf{r}_4|)$$

$$\times G_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4,t), \tag{6}$$

where

$$G_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4,t)=\langle\rho(\mathbf{r}_1,0)\rho(\mathbf{r}_2,t)\rho(\mathbf{r}_3,0)\rho(\mathbf{r}_4,t)\rangle$$

$$-\langle\rho(\mathbf{r}_1,0)\rho(\mathbf{r}_2,t)\rangle$$

$$\times\langle\rho(\mathbf{r}_3,0)\rho(\mathbf{r}_4,t)\rangle. \tag{7}$$

We can write Q in terms of its self and distinct parts, $Q=Q_S+Q_D$. The self part Q_S corresponds to terms with $i=j$ in Eq. (4):

$$Q_S(t)=\sum_i \int d\mathbf{r}w(r)\delta(\mathbf{r}+\mathbf{r}_i(0)-\mathbf{r}_i(t))=\sum_i w(\mu_i), \tag{8}$$

where μ_i is the magnitude of $\vec{\mu}_i$. The distinct part Q_D is equal to

$$Q_D(t)=\sum_{i=1}^N \sum_{\substack{j=1\\ j\neq i}}^N w(|\mathbf{r}_{ij}-\vec{\mu}_j|). \tag{9}$$

Then $\chi_4(t)$ can be decomposed into self- (χ_{SS}), distinct- (χ_{DD}), and interference (χ_{SD}) parts: $\chi=\chi_{SS}+\chi_{DD}+\chi_{SD}$. From Eq. (5), χ_{SS} and χ_{DD} describe the fluctuations of Q_S and Q_D, respectively, and χ_{SD} describes the cross fluctuations: $\chi_{SS}\propto\langle Q_S^2\rangle-\langle Q_S\rangle^2$, $\chi_{DD}\propto\langle Q_D^2\rangle-\langle Q_D\rangle^2$, and $\chi_{SD}\propto\langle Q_S Q_D\rangle-\langle Q_S\rangle\langle Q_D\rangle$. According to Eq. (8), $Q_S(t)$ contains only contributions from small displacements, $\mu_i<a$, since $w(\mu_i)=0$ for $\mu_i>a$, and thus $\chi_{SS}(t)$ is the susceptibility of localized particles,[22] those which during a time interval $[0,t]$ move less than a distance a. In contrast, $Q_D(t)$ contains contributions from particles for which $|\mathbf{r}_{ij}-\vec{\mu}_j|<a$; that is, particles that are replaced by a neighboring particle.

In Ref. 8, a different generalized susceptibility $\chi_U(t)$ was defined in terms of the fluctuations in an ''order parameter'' given by the total particle displacement $U(t)$ in a time interval t: $U(t)=\Sigma_{i=1}^N \mu_i(t)=\int d\mathbf{r}\, u(\mathbf{r},t)$, where the displacement density field $u(\mathbf{r},t)=\Sigma_{i=1}^N \mu_i(t)\delta(\mathbf{r}-\mathbf{r}_i(0))$. Here we compare $\chi_4(t)$ with $\chi_M(t)$, defined as

$$\chi_M(t)=\frac{\beta V}{\langle M(t)\rangle^2}[\langle M^2(t)\rangle - \langle M(t)\rangle^2], \tag{10}$$

where $M(t)\equiv\Sigma_{i=1}^N \mu_i^2(t)$ (i.e., $M(t)$ is the sum of the *squared* displacements for one system in a time interval $[0,t]$). Like $\chi_U(t)$, $\chi_M(t)$ is proportional to the volume integral of a correlation function of (in this case squared) particle displacements.[9,23] Both the displacement density field $u(\mathbf{r},t)$ and squared-displacement density field $m(\mathbf{r},t)$ are dominated by particles with large displacements.

To evaluate these quantities we use data obtained from a molecular dynamics simulation of a model LJ glass-former. The system is a three-dimensional binary mixture (50:50) of 500 particles interacting via LJ interaction parameters.[24] We analyze data from state points at seven different temperatures T approaching $T_c\approx0.592$ from above[25] at a constant density $\rho\approx1.3$. (In the remainder of this letter all values are quoted in reduced units.)[24] All quantities presented here are evaluated in the *NVE* ensemble following equilibration of the system at each state point. Further simulation details may be found in Refs. 25, 26.

In Fig. 1(a) the susceptibility $\chi_4(t)$ calculated via Eq. (5) is shown as a function of time for different values of T. As found for a different LJ mixture in Ref. 19, for all T, $\chi_4(t)$ is zero at short time and attains a small constant value at large
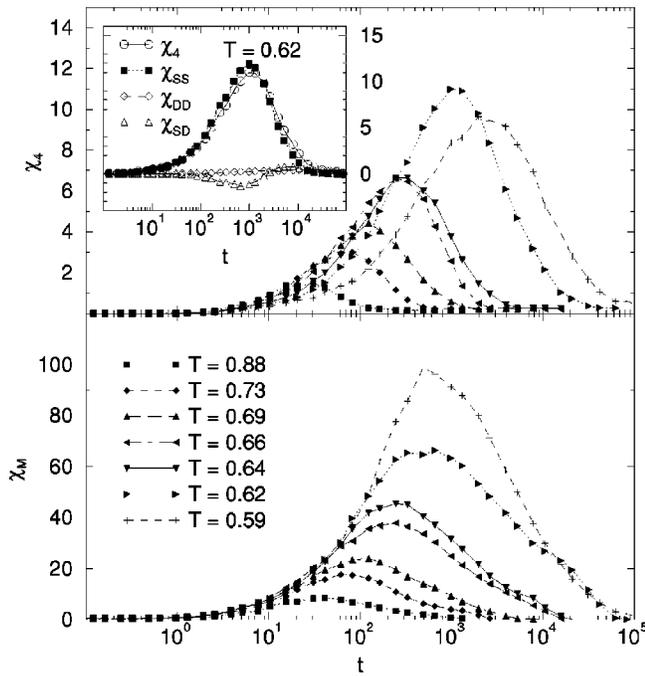
FIG. 1. (a) Time dependence of the susceptibility $\chi_4(t)$ at various temperatures as indicated in (b). Inset: Self; distinct, and cross-terms of $\chi_4(t)$ at $T=0.62$. (b) Time-dependence of the ''squared-displacement'' susceptibility $\chi_M(t)$ at the same values of T as in (a).

FIG. 2. Temperature dependence of $\chi_4(t_4^*)$ and $\chi_M(t_M^*)$. The solid and dashed lines are power law fits to the data as indicated (excluding the lowest temperature). The error bars are estimated from deviations between three independent samples, where for each sample, $\chi_4(t)$ and $\chi_M(t)$ are calculated by averaging over 128 different time origins.

time, and has a maximum at some intermediate time t_4^*. Both t_4^* and the amplitude of the peak, $\chi_4(t_4^*)$, increase strongly with decreasing T. At the lowest value of T, the amplitude of $\chi_4(t)$ decreases, possibly due to finite size effects or to the change in dynamics[25] near T_c. The inset shows the self-, distinct-, and cross-terms of $\chi_4(t)$ for one value of T, and we see that χ_{SS} is indeed the dominant term. Thus, $\chi_4(t)$ is dominated by the growing range of spatial correlations between *localized* particles in this fluid, and t_4^* is the time when this correlation is strongest.[22] In fact, several authors have reported evidence of a growing length scale associated with solid-like behavior in dense fluids.[27]

Figure 1(b) shows $\chi_M(t)$ calculated from Eq. (10) as a function of time for different values of T. We find that $\chi_M(t)$ becomes negligable at small and large times and has a maximum at some intermediate time t_M^* where the spatial correlation of squared particle displacements is strongest. This behavior is similar to that exhibited by $\chi_U(t)$ calculated in Ref. 9.

As shown in Fig. 2, both $\chi_4(t_4^*)$ and $\chi_M(t_M^*)$ increase strongly with decreasing T [with the exception of $\chi_4(t_4^*)$ at the lowest temperature]. Over the limited temperature range of our simulations, both functions may be reasonably fitted by power-law functions $(T-T_c)^{-\gamma}$ with $T_c=0.592$, with the apparent exponents $\gamma_4=0.80\pm0.07$ and $\gamma_M=0.87\pm0.05$, as shown in the figure. (In fitting the power law, T_c is held fixed to the value $T_c=0.592$ determined in previous work.)[25] Of course, precise determination of the functional form requires simulations at lower temperatures and larger simulations to reduce any possible finite size effects expected due to the growing range of correlated particle motion and localization driving the growth of $\chi_M(t)$ and $\chi_4(t)$.

Figures 3(a) and (b) show the T-dependence of t_4^* and t_M^*, respectively, and compare them with both the inverse self-diffusion coefficient D^{-1} and the structural relaxation time τ_α. [Here D is calculated from the mean-square displacement for the B (small) particles, and τ_α is calculated by fitting the α-relaxation part of the self-intermediate scattering function at the wave vector corresponding to the first peak in
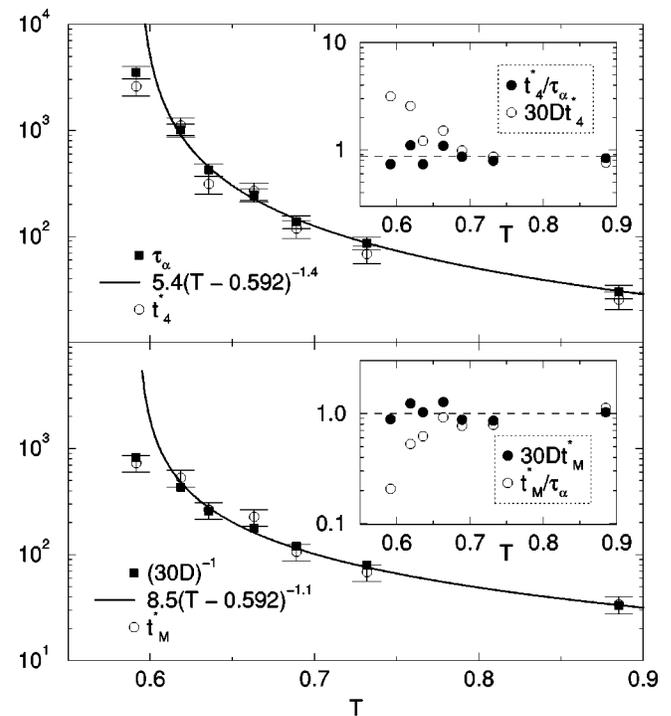
FIG. 3. (a) Temperature dependence of τ_α and the time t_4^* at which $\chi_4(t)$ exhibits a maximum. (b) Temperature dependence of the inverse self-diffusion coefficient D and the time t_M^* at which $\chi_M(t)$ exhibits a maximum. The solid lines are power law fits to τ_α and D^{-1}, respectively (excluding the lowest temperature), with T_c fixed. Insets: Comparison of t_4^* and t_M^* with both D^{-1} and τ_α. As plotted, a line of zero slope (dashed line) indicates proportionality.

the static structure factor for the A (large) particles (not shown, see Ref. 25) by a stretched exponential function.] Also shown are power law fits to D^{-1} and τ_α excluding the lowest temperature (see Ref. 25 for details). Diffusion and relaxation are found to be ''decoupled'' in this cold liquid, as observed in many other real and simulated cold liquids.[4,5,12,13,28] In the present system, we find that with $T_c = 0.592$, $\gamma_D = 1.11 \pm 0.03$, and $\gamma_\tau = 1.41 \pm 0.07$. Remarkably, we find[29] that the T-dependence of t_4^* coincides within our numerical error with that of τ_α, while t_M^* behaves like D^{-1}. That is, the time scale on which the localized particles are most spatially correlated[22] scales with temperature like the structural relaxation time, and the time scale on which the correlation between squared particle displacements is strongest scales like the inverse diffusion coefficient. Thus, our data demonstrates that the ''decoupling'' of diffusion and relaxation (or viscosity) may be directly attributed to the emergence of dynamical heterogeneity, as argued by, e.g., Sillescu and co-workers,[4] Ediger and co-workers,[5] Stillinger,[16] and Douglas.[17]

Our results demonstrate the importance of time-dependent higher-order density correlation functions in the characterization of dynamical heterogeneity in supercooled liquids, and the ramifications of this heterogeneity for the bulk dynamics. In particular, the increasing amplitude of the generalized time-dependent susceptibility $\chi_4(t)$ with decreasing T, as shown also in Ref. 19, demonstrates an essential difference between two- and four-point density correlation functions in these fluids. For a glass-forming LJ liquid, we have shown that $\chi_4(t)$ is dominated by growing spatial correlations between temporarily localized particles.[22] Finally, we have demonstrated that the decoupling of diffusion and structural relaxation observed in supercooled liquids follows naturally from dynamical heterogeneity, as discussed by many authors: the time scale for spatial correlations of localized particles to develop governs structural relaxation, while the (different) time scale for the development of spatial correlations of squared particle displacements governs diffusion. We note that it should be possible to determine the four-point functions studied here in colloidal suspensions using particle tracking methods.

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