Transport coefficients at Metastable Densities from models of Generalized Hydrodynamics

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ABSTRACT

In the present work we compute the enhancement in the long time transport coefficients due to correlated motion of fluid particles at high density. The fully wave vector dependent extended mode coupling model is studied with the inclusion of an additional slow variable of the defect density for the amorphous system. We use the extremely slow relaxation of the density correlation function observed in the light scattering experiments on colloids to estimate the input parameters for the model. The ratio of long time to short time diffusion coefficient is studied around the peak of the structure factor.

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1 Introduction

Liquids undercooled below the freezing temperature without crystallization shows a sharp increase in their transport properties like viscosity and acquire solid like properties e.g. has a finite shear modulus. The dynamics of the dense liquids are studied in terms of generalized transport coefficients, expressed in the form of memory functions which account for the correlated motion of the fluid particles at high density. Frequency and wave vector dependence of these quantities, relevant for behavior over different time and length scales are studied through models of the liquid obtained from the statistical mechanics of a many particle system. Much progress has been made in this field through a self-consistent approximation of the memory function in terms of the slowly decaying modes that exist as a consequence of microscopic conservation laws. This has been termed in the literature as the mode coupling theory (MCT) [1, 2]. A key result of the simple mode coupling model [3, 4] for the memory function, is a two step relaxation process involving the power law decay of correlation over intermediate time scales crossing over to the stretched exponential behavior in the long time, termed as the $\alpha$-relaxation. With increase of density, the liquid become very viscous and relaxation slows down strongly. The basic driving mechanism for the enhancement of the viscosity comes from a feedback to the transport coefficient from the coupling of the slowly decaying density correlation functions. In the simple model, long time correlation of density fluctuations freeze completely beyond a critical density and the fluid undergoes a dynamic transition to a nonergodic phase. An extended version [5, 6, 7] of the mode coupling theory with proper coupling of the density and current modes demonstrated that there is a final exponential relaxation of the density correlation function over asymptotic time scales. The final decay process which shifts to longer time scales with density can be interpreted as a process similar to the diffusion of free volumes in the amorphous system whereby ergodicity is maintained.

In the theoretical formulation of the dynamics of the supercooled liquids often extra slow
modes \[8, 9, 10\] in addition to the usual ones that follow from simple hydrodynamics, has been considered. Extension of the simple MCT has been done \[11, 12\] along these lines to include the extra slow mode of defect density that develop in the amorphous solid like state. In Ref. \[13\] and \[14\] the displacement field \(\vec{u}\) was used to describe the extra hydrodynamic mode due to the solid like nature of the system. Ref. \[14\] demonstrated that due to the nonlinear couplings of the density fluctuations and the displacement field \(\vec{u}\) in a solid like state, the renormalized expression for transport coefficients include terms linear in density correlation function. Such terms in the memory function appears over time scales in which the defect correlation is taken to be a constant. Due to this phenomenological inclusion of the extra slow mode of the defect density, a line of transition is obtained instead of a single critical density. In this formulation of the extended MCT, temperature or density dependent exponents of power law relaxation \[12\] are obtained. Over the longest time scale the defect correlation as well as the density correlation function do however decay to zero and thus restores the ergodic behavior. In a recent work \[11\] we have studied the coupled dynamics over longest time scale, through a schematic model dropping all wave vector dependencies. The asymptotic behavior for both these correlation functions have been considered there to study the relaxation over various time regimes. However, in order to study the transport behavior with respect to change in the thermodynamic parameters like density, the wave vector dependence of the model has to be taken into account. In the present work we consider the wave vector dependent model for the dense supercooled liquid and the coupling of the density fluctuations to the extra slow mode of defect density are included. The dynamics of the density correlation function is considered up to the time scales where the final ergodicity restoring mechanism present. It was demonstrated \[5\] from a non perturbative approach that the self consistent feedback mechanism allows a non-zero value of the cutoff function. However, explicit evaluation of the strength of the function is extremely difficult beyond the one loop order. On the other hand in certain simple systems like colloids very slow
relaxation of the structure function has indeed been observed. This indicates that the cutoff function is effective over very long time scales. In the present calculation we model this through a self-consistent picture where the defect density and the density fluctuations over longest time scales behave in a similar way. We use here this criteria to close the dynamical equation for the density correlation function - instead of actually evaluating cutoff function self consistently. The cutoff function is estimated to give decay of the structure factor that agrees with experimental results on hard sphere like systems like Colloids. We then compute the mode coupling integral to obtain the renormalized transport coefficient as function of density. The coupling of the slowly decaying density fluctuations to the defect density in the super cooled state is a key aspect of this model and allows a physical picture for the ergodicity restoring mechanism.

The plan of the paper is as follows. In the section II we give a brief description of the model studied and the approximations involved while in section III we describe our results for the transport coefficients. We end the paper with a small discussion.

2 Transport Coefficients at High Density

2.1 Longitudinal Viscosity

In this section we describe briefly the model used in computing the longitudinal viscosity or the sound attenuation constant. The key quantity of interest is the dynamic structure factor or the density auto correlation function (DACF). The Fourier-Laplace transform of the DACF \( \psi_q(t) \) normalized with respect to its equal time value \(^1\), defined as,

\[
\psi_q(z) = -i \int_0^\infty e^{izt} \psi_q(t)dt \quad \text{Im}(z) > 0
\]

In the basic formulation of the MCT, this quantity is expressed in terms of the generalized
memory function $\Gamma_q(z)$,

$$\psi_q(z) = \frac{z + i\Gamma_q(z)}{z^2 - \Omega_q^2 + i\Gamma_q(z)[z + i\gamma_q(z)]}. \tag{2}$$

$\Omega_q$ corresponds to a characteristic microscopic frequency for the liquid state dynamics. The memory function which is the generalized longitudinal viscosity, consists of the bare and mode coupling part, $\Gamma_q(z) = \Gamma_0^q + \Gamma_{mc}^q(z)$. $\Gamma_0^q$ is related to bare or short time dynamics with uncorrelated collisions. The mode coupling contribution $\Gamma_{mc}^q(z)$ signify the correlated motion in the dense liquid and can be expressed in terms of the coupling of density fluctuations dominant at supercooled densities. Several models has been used to compute the Generalized memory function for the supercooled state. The simplest model \cite{4, 15}, express the memory function in terms of a bilinear product of the density correlation functions i.e. $\Gamma_{mc}(t) = c_2 \psi^2 (t)$ . The model that contains a linear term of the density correlation function in addition to the quadratic one, i.e. $\Gamma_{mc}(t) = c_1 \psi(t) + c_2 \psi^2 (t)$, has been referred to in the literature \cite{3, 16} $\phi_{12}$ model. In the wave vector independent form this model gave rise to the stretched exponential relaxation. In later works it was demonstrated that by considering the solid like nature of the amorphous state at high density such a linear term can be obtained in the renormalization of the transport coefficients. The set of hydrodynamic variables were now extended to include the slowly relaxing defect density. The resulting non linearities in the equations of Generalized Hydrodynamics gives rise to the above memory function in terms the DACF. In order to consider the very dense system approaching the glass transition we will use here this model for the dynamical behavior. Following Ref. \cite{17, 18}, the mode coupling part of the generalized transport coefficient is given by,

$$\Gamma_{mc}^q(t) = \int \frac{d\vec{k}}{n_0(2\pi)^3} S(q)[\tilde{V}^{(1)}(q, k)\psi_{k_1}(t)\phi_{k_1}(t) + \tilde{V}^{(2)}(q, k)\psi_k(t)\psi_{k_1}(t)] \tag{3}$$

with $\vec{k}_1 = \vec{q} - \vec{k}$. Here $\phi_{k_1}(t)$ is the normalized defect density auto correlation function and gives the contribution to the transport coefficient coming from the coupling of the defect density with the density fluctuations. The mode coupling vertices with full wave vector
dependence are given by

$$\tilde{V}^{(1)}(q,k) = 2\tilde{U}(q,k)S(k)S(k_1) + \kappa n_0 c(k)S(k)$$  \hspace{1cm} (4)$$

and

$$\tilde{V}^{(2)}(q,k) = \frac{1}{2}\tilde{U}^2(q,k) - y\tilde{U}(q,k)S(k)S(k_1)$$  \hspace{1cm} (5)$$

The vertex function $\tilde{U}$ in (4) is given by,

$$\tilde{U}(q,k) = \frac{n_0}{q}[\hat{q}.kc(k) + \hat{q}.(k_1)c(k_1)]$$  \hspace{1cm} (6)$$

where $n_0$ is the particle number density. It is assumed that the defects are moving in a metastable double well potential and this well depth is characterized by $y$. The defect density is assumed here to be a variable similar to mass density and is weakly interacting with the latter. $\kappa$ is a dimensionless parameter characterizing the coupling of the defect density with the particle density in the Free energy functional corresponding to the stationary state around which fluctuations are considered. For $\kappa$ and $y$ both equal to zero, (3) reduces to the simple MCT result, [15]. If the defect correlation is assumed to be very long lived compared to the time scales over which initial power law relaxation persists, $\phi_{k_1}(t)$ in equation (3) can be taken as a constant and the so called $\phi_{12}$ model results on ignoring all wave vector dependencies. The density dependence of the power law exponents and the resulting dynamic instability in the model has been studied in Ref. [12]. Here we will consider the time scale over which both density correlation and the defect correlation decays. The quantity $\gamma_q(z)$ in the R.H.S of eqn. (2) has crucial implications for the asymptotic dynamics [5]. If $\gamma$ is ignored then a sharp transition in the supercooled liquid to an ideal glassy phase results beyond a critical density. The Laplace transform of the density correlation function develops a $1/z$ pole. This is a widely studied [2] model for the dynamics of supercooled liquids. However, with the presence of $\gamma_q$ at high density when $\Gamma$ gets large, the pole shifts to $1/(z + \gamma_q)$. It has been demonstrated [4, 7] that in the small $q$ and $\omega$ limit, the quantity $\gamma_q \sim q^2 \gamma$, where
\( \gamma \) remains finite, instead of self consistently going to zero. This implies a diffusive decay of the density correlation restoring ergodicity in the longtime limit. The quantity \( \gamma \) that provides a mechanism that cuts off the sharp transition of the fluid to an ideal glassy phase is \( O(k_B T) \) to leading order in the perturbation theory. It is a consequence of the coupling of the density and current correlation in the compressible fluid. Formal expression for \( \gamma \) was obtained in self consistent form in ref. \cite{5} in terms of current correlation function. However beyond the one loop order explicit evaluation of the strength of the cutoff function becomes extremely complicated. In the present work, we estimate this by interpreting it as similar to slow diffusion of free volumes in the dense liquid. We use the simple model of defect correlation as \( \phi_q(t) = e^{-\delta(q)t} \) and replace the \( \gamma(q) \) in equation (2) by \( \delta(q) \) to have a closed equation for the density correlation function. The density correlation function is obtained by solving equation (2) in the time space \cite{19}. The quantity \( \delta \) is treated as a parameter and is adjusted to obtain agreement in the asymptotic behavior of the dynamic structure factor as seen in experiments. For this purpose the data from light scattering on colloids by Pusey et al. is \cite{20} is considered. The solution of the MCT equations provide the result for the relaxation of the density correlation function with time scaled w.r.t the microscopic time scale for the Hard Sphere system - namely the Enskog time. The present analogy of the Hard Sphere system with the colloids is used only with regard to the nature of the cooperativity. The unit of time \( \tau \) in terms of which the experimental results are presented, then needs to be related to the characteristics microscopic time scale of the Hard Sphere model i.e. \( \sqrt{\beta m \sigma} \). \( m \) is the mass of the liquid particle and \( \beta \) is the Boltzmann factor. The latter is related to the Enskog time \cite{21} for the system. The MCT models for the dynamics we consider here does not describe the short time dynamics or the bare transport coefficient for Colloids - the relevant comparison is over the long time scales or the nature of the correlated motion of individual units. These correlated motions are quantified in terms of the mode coupling terms. Thus the comparison of the theoretical data with the results
of the experiment is done upto an undetermined scaling factor of the short time scales in the two systems. The decay of density correlation as a function of time is evaluated over a suitable wave number grid. The generalized transport coefficients expressed in terms of the coupling of density correlations from the mode coupling contribution is then evaluated. The longitudinal viscosity is computed relative to its bare or short time contribution. The dependence of the Generalized Viscosity can be probed in the present model from various approach - namely the time dependence as well as the wave vector dependence of the long time limit. The detailed results from the numerical solutions are presented in the next section.

2.2 Long time Diffusion

The density fluctuation in a dense fluid at finite wave number close to the peak of the static structure factor follows a diffusive behavior. Once again similar to the viscosity the diffusion constant has short time value $D_s$ and the long time $D_L$ behaviors. If all kinds of correlated motions are completely ignored then the diffusion coefficient is related to the bare transport coefficient. At high densities the cooperative nature of the dynamics is essential in computing the diffusion coefficient. This is obtained by including in the memory function the mode coupling contribution. The short time dynamics depends on the microscopic nature of the system. We use this diffusion constant to define the unit of time namely $\tau = \sigma^2/D_s$. The mode coupling part is computed through the self-consistent expression in terms of the density correlation function. For the DACF, the model described in the first part of this section is used. Once again instead of focussing on the microscopic model we compute the ratio of the longtime to short time diffusion coefficient. This ratio of diffusion coefficients for the density fluctuations at finite wave number is expressed in terms of the memory function...
through the formula

\[
\frac{D_L(q)}{D_s} \frac{\Gamma_q}{s(q)\bar{\Gamma}_q} = \frac{1}{s(q)\bar{\Gamma}_q}
\]

where \(\bar{\Gamma}_q = \int_0^\infty d\tau \Gamma_q^\text{mc}(\tau)\), is the long time limit of the memory function given by equation (3) and \(s(q)\) is the static structure factor. The colloidal time \(\tau\) is related to the Enskog time \(t_E\) through a constant \(\Delta\), i.e. \(\tau = \Delta t_E\). The numerical results for the transport coefficients obtained from these formulations w.r.t to density and wave number variations are described in the next section.

3 Results for the transport coefficients

The dynamic density-density correlation function \(\psi_q(t)\) is obtained as a solution of the non-linear equation obtained by inverse transforming equation (2). The generalized transport coefficient \(\Gamma_q(t)\) has two parts namely the bare and the mode coupling part. The bare quantities in the Hard Sphere system for which the dynamical equations are solved are obtained in terms of standard models of kinetic theory and the unit of time is expressed in terms of the Enskog time \(t_E\). The integro-differential equation obtained from equation (4) is solved for \(\psi_q(t)\) self-consistently over a wave vector grid of \(N = 200\) and an upper cutoff of \(\Lambda \sigma = 30\) using numerical integration. Here we have used the Percus-Yevick \(\text{[22]}\) solution with Verlet-Wiess \(\text{[23]}\) correction for the hard sphere static structure factor. The constant \(\Delta\) relating the two time scales, colloidal and the microscopic time, is estimated from the dynamic structure factor obtained from colloidal experiments. The colloidal time \(\tau\) is estimated as \(2.9 \times 10^5\) times Enskog time \(t_E\). As discussed in the previous section the cutoff function \(\gamma\) in equation (2) is estimated through a parameter \(\delta\) which is inversely related to the relaxation time for the defects. The parameter \(\delta\) is adjusted to get an agreement with the light scattering data, reported in Fig. 5(a) of Ref. \(\text{[20]}\) for the dynamic structure factor, in the asymptotic limit. The light scattering data supplied in Ref. \(\text{[20]}\) was thus used to obtain the corresponding
value of the parameter $\delta$ at that density. In Fig. 1 the $\delta$ values thus obtained in units of $\sigma^2/\tau_H$ are plotted as a function of packing fraction for the dense fluid. Here $\tau_H = 10t_E$ is the hard sphere unit of time with respect to which the time is scaled. For intermediate densities, the $\delta$ value follows a power law behavior approaching divergence at $\eta \approx 0.536$ as shown by dotted line. With increase of density the viscosity smoothes off to a slower enhancement. The parameter $\delta$ as estimated from the asymptotic behavior of the dynamic structure factor of the colloids, is then used to compute the density correlation function from the model equations self consistently. This is also used to compute the memory function over very long time scales and the transport coefficients in the supercooled state is thereby computed in terms of the Mode Coupling contributions accounting for the correlated motions at higher densities. The long wavelength limit of the memory function $\Gamma_q(t)$, reduces to the form,

$$\lim_{q \to 0} \Gamma_q(t) = \frac{1}{\beta m} \int \frac{d\vec{k}}{(2\pi)^2} \left\{ \tilde{V}^{(1)}(k)\phi_{k_1}(t) + \tilde{V}^{(2)}(k)\phi_k(t)\phi_{k_1}(t) \right\}$$

with the vertex functions given by,

$$\tilde{V}^{(1)}(q \to 0, k) \equiv \tilde{V}^{(1)}(k) = 2c(k)\{2yS(k) + \kappa\}S(k) + \frac{2}{3}c_1(k)S^2(k)$$

and in a similar way,

$$\tilde{V}^{(2)}(k) = [c^2(k) + \frac{1}{5}c_1^2(k) + \frac{2}{3}c(k)c_1(k) - 2yc(k) - \frac{2y}{3}c_1(k)]n_0S^2(k)$$

where $c_1(k) = kc'(k)$. In Fig. 2 the generalized memory function $\Gamma(t)$ in the long wavelength limit is plotted as a function of time for the packing fraction $\eta = 0.542$. Here the time $(\tau_H)$ is expressed in the units of the Enskog time, $t_E$. The corresponding value of $\delta$ is $1.0 \times 10^{-7}$ in units of $(\sigma^2/\tau_H)$ and the metastability parameters in the MCT equation are $\kappa = -0.01, y = 0.12$ respectively. The metastability parameter $y$ determines the potential well used for the defect density. In figure 3 the form of the well used in the calculations for figure 2 is shown. We have expressed the result for $\Gamma(t)$ in a dimensionless quantity in terms of the unit $(\beta m)^{-1}$. $\Gamma(t)$ also follows the two step relaxation process. At intermediate
time scales long compared to the microscopic time scales, it follows a power law decay finally crossing over to the stretched exponential decay at longer times. The variation of the stretching exponent $\beta$ as a function of packing fraction is shown in Fig. 4. With increase in density $\beta$ decreases implying increase or stretching of relaxation of the DACF at higher densities. This critical slowing down of the system is estimated through the zero frequency limit of the longitudinal viscosity. In Fig. 5 longitudinal viscosity scaled with respect to the corresponding bare transport coefficient, $\Gamma(z = 0)$ is plotted as a function of packing fraction. Over the low density region the viscosity shows a power law behavior approaching a divergence around $\eta = 0.538$. The corresponding exponent is 1.1. With increase of density an abrupt increase in the viscosity by approximately 4 orders of magnitude is observed. Thus at the onset of the glassy regime, a relatively strong enhancement in viscosity is obtained from the self-consistent mode coupling models. We also present the wave number dependence of the viscosity to illustrate the nature of transport over different length scales. Indeed an analysis of the self consistent expression for the density correlation function indicates that the relevant quantity for long time decay is inverse of $S(q)\Gamma_q$. This is plotted in Fig. 6 scaled with respect to the bare time $v_o \sigma$, where $v_o$ is the thermal velocity of the particles. Here it is shown as a function of the wave number $q \sigma$ for $\eta = 0.542$. The values for the metastability parameters $\kappa$ and $y$ are same as stated for figure 2. The result shows that the time scale indicating decay rates for different wave numbers is slowest near the peak of the structure factor and for small wave number it approaches a constant value. It should be noted that this is the final decay process in the case when one completely ignores the cutoff mechanism. Thus with increase of density this will go into a delta function type peak. However the existence of the cutoff mechanism ensures the proper ergodic behavior through the diffusive process over longest time scales. At very small wavelength i.e. large wave numbers the transport coefficients are same as calculated from the short time properties of the liquid, i.e., $\Gamma_q \to 1$. This indicate that over small length scales the effect
of correlated motion is insignificant even at very high density and the mode coupling has negligible implications on the transport properties. With increasing wavelength the viscosity increases and is maximum at $\lambda \to \infty$ or $q\sigma \to 0$. The signature of the structure of the liquid are seen in terms of very small maximas and minimas (see Fig. 6) over the whole wave vector range. Fig 7 shows the ratio of long time to short time diffusion coefficient at the peak of the structure factor as a function of density of the supercooled system. It shows a power law decay of diffusion at intermediate densities which shows a divergence around $\eta = 0.538$ with an exponent close to unity. As the density increases further the decrease in the diffusion coefficient slows down due to the presence of cutoff effect in the present model. The diffusion coefficients are calculated in colloidal units with $\Delta = 2.9 \times 10^5$. To study the dynamics at the microscopic time scales we have calculated the ratio of the long time to short time diffusion coefficient as a function of wave vector. Fig 8, presents the behavior of the Diffusion Coefficient over a wave vector range around the diffraction peak. The results shown correspond to the packing fraction $\eta = .542$. We observe that the diffusion coefficient is minimum at the peak of the structure factor where maximum number of nearest neighbours are present. We observe qualitatively similar behavior as observed by Pusey et al. [25]. The values of the metastability parameter used are same as mentioned above for figure 2. It should be noted that it is only over the finite wave vector range one can describe the density fluctuations following a diffusive mechanism even in the models where there is no cutoff mechanism of hydrodynamic diffusion is considered. Of course in this case it will only be vaild upto densities below the ideal transition point beyond which transport coefficient at all wave numbers would diverge in the ideal nonergodic state. However, with cutoff mechanism taken into account in the present work, as a motion of defect density - one is not restricted to densities below the ideal glass instability only. Fig. 8 considers the effect on the diffusive process at finite length scale within the self consistent formulation of the problem where the cutoff mechanism is also being taken into account.
4 Discussion

The standard mode coupling theory of glass transition predicts a sharp dynamic transition with diverging transport coefficients. However in realistic systems such a freezing does not occur through substantial slowing down of the relaxation as seen in the light scattering data of colloids.

In the present work we considered a phenomenological model for the dynamics of the supercooled system where structure factor for the hard sphere system was used as an input for thermodynamic properties. The density correlation function is the key quantity in the mode coupling model. In this formulation the self-consistent form of the cutoff function used is obtained in terms of the coupling to current correlation functions to lowest order in perturbation theory. The present model includes the phenomenological description of slowly decaying defects in the amorphous structures. We interpret the cutoff mechanism of final decay of density correlation function as being related to the diffusion of free volume or defect in the amorphous system. The strength of the cutoff function is estimated by seeking an agreement with the relaxation of dynamic structure factor seen in experimental studies on colloidal systems. The correlation of the density fluctuations in the presence of very slowly decaying defect correlations contribute to the transport coefficients calculated using the feedback mechanism. The present theory of course does not compute the bare transport coefficient which is related to the short time scale dynamical behavior. The mode coupling contributions are computed relative to the short time or the bare properties. The longitudinal viscosity in the long time limit is computed and shows an enhancement by four orders of magnitude over small density range with an approach to power law divergence around a critical range of density $\rho = 1.028$ which finally smooths off to a slower decay. The wave vector dependence of the transport coefficient is also considered. The decay rate over finite wave vector shows a minimum at the peak of the structure factor.

The relaxation of density fluctuations over the intermediate length scales follows a dif-
fusive behavior [26, 27]. Due to effects of correlated motions in the fluid particles at high
density the diffusion coefficient is changed over long times. We compute this by taking into
account the Mode Coupling effects due to slow decay of density correlations. Thus the ratio
of the long time to short time diffusion constant is calculated showing that the diffusion
process slows down. However the diffusion process is faster than the final diffusive decay
of defect density fluctuations given by $\delta$ in the $q \to 0$ limit. The latter is related to the
ergodicity restoring mechanism in the fluid and correspond to the largest time scale in the
dynamics.

The present work demonstrates that the very slow decay of DACF seen in colloids can
be understood in terms of the extended MCT and the corresponding values of the transport
coefficients like longitudinal viscosity or long time diffusion constant is calculated in terms
of the short time properties of the system.

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**Figure Captions**

**Figure 1**

Plot of $\delta$ in units $\sigma^2/\tau_H$ Vs packing fraction.

**Figure 2**

The normalized generalized viscosity $\Gamma(t)$ in units of $(\beta m)^{-1}$ as a function of time at $q\sigma = 0.0$, $\eta = 0.542$, $y = 0.15$, $\kappa = -0.01$ and $\delta = 1.0 \times 10^{-7}$.

**Figure 3**

Variation of depth of the potential with $n/n^*$ for $\eta_c$ 0.542. In the figure $h^*(n)$ represent the dimensionless quantity $h(n)\beta\epsilon n\sigma^3$. The metastability parameters used are $\kappa = -0.01$ and $y = 0.12$.

**Figure 4**

Variation of $\alpha$-relaxation stretching exponent $\beta$ with packing fraction.

**Figure 5**

Plot of the longitudinal viscosity $\Gamma$ scaled w.r.t to the bare transport coefficient as a function of packing fraction $\eta$.

**Figure 6**

Variation of $\Gamma_q^{-1}$ in units of $v_0\sigma$ with wave number $q\sigma$ at $\eta = 0.542$.

**Figure 7**

Variation the longtime to short time diffusion constant ($D_L/D_s$) as a function of packing fraction.
Figure 8

Wave vector dependence of the ratio of longtime to short time diffusion constant, \( D_L(q^*)/D_s \times 10^5 \) near the peak of the structure factor.
\[ \frac{\Gamma(qt)/q^2}{t^*} \]
