

Mode-coupling theory of the glass transition in colloidal systems

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Within a mode-coupling approximation we investigate the influence of the short-time dynamics of colloidal fluids on the glass-transition scenario. We assume Brownian dynamics, neglect hydrodynamic interactions, and follow approximations used to construct a mode-coupling theory (MCT) for structural relaxation in dense atomic fluids with underlying Newtonian dynamics. This approach predicts the same scenario for the kinetic glass transition in colloidal systems as the MCT for atomic systems. We compare this result with recent computer-simulation studies of the glass transition for atomic and colloidal liquids.

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I. INTRODUCTION

Recently, dynamical properties of concentrated colloidal suspensions have gained increasing interest. Extensive light-scattering studies on sterically stabilized colloidal liquids [1] have revealed that with increasing concentration the liquid undergoes a kinetic transition to a glassy state. The structural relaxation then becomes frozen over experimentally accessible time scales. As far as static properties are concerned one can view a colloidal suspension as an atomic liquid [2,3] and use usual liquid state theory [4] to predict the structure. This is not true, in general, for the dynamics: due to the complete separation of time scales between the colloidal and the solvent particles [2] the dynamics is Brownian for colloidal liquids, whereas it is Newtonian for atomic ones. Since the kinetic glass transition is connected with dynamics, it is *a priori* unclear whether the relaxation scenario near the transition of a colloidal fluid is similar to that of an atomic liquid where, by now, experiments [5], mode-coupling theories [6] and computer simulations [7–9] have provided a lot of detailed insights. It has been argued that on long-time scales the motions both in a colloidal liquid and in an atomic fluid can be considered as “interactive diffusion” [10], which makes it plausible that the relaxation scenarios are similar, too. In order to clarify the dependence of the long-time relaxation on the short-time dynamics, a computer simulation was performed [11] for a simple model of a charge-stabilized colloidal suspension. To facilitate the comparison, both Brownian dynamics (BD) and molecular (or Newtonian) dynamics (MD) have been used for the same model system. It was shown that upon cooling the glass transition occurs at the same temperature for both BD and MD. The nonergodicity (Edwards-Anderson [12]) parameter turns out to be very similar, too, as well as the final α -relaxation scenario, whereas for short and intermediate times the relaxation has qualitative differences.

The main objective of the present paper is to investi-

gate within a mode-coupling approximation the influence of the short-time dynamics of colloidal systems on the glass-transition scenario. For atomic systems a mode-coupling theory (MCT) has been constructed during the past decade [6]. This theory gives detailed predictions on the asymptotic relaxation scenario of density fluctuations near the kinetic glass transition. It was originally developed for simple one-component systems with spherically symmetric interactions and then extended to two-component mixtures [13,14]. Recently, it was shown that some of its predictions are experimentally encountered in much more complex systems like sterically stabilized colloidal fluids [15], polymers [16], molten salts [17], and other complex liquids [18]. It has been suggested [6] that the predictions of the MCT are universal, i.e., they do not depend on the form of the short-time dynamics. In the present paper, we prove this statement explicitly for the case of Brownian dynamics, valid for colloidal systems, where the equations of motion are governed by the Smoluchowski rather than by the Liouville operator [2]. We start with the memory function representation of the density autocorrelation function derived by Cichocki and Hess [19] and follow a mode-coupling approach, very similar in spirit to that used for Newtonian dynamics [6]. In this way we construct a MCT [20] which leads to the same asymptotic form of the time dependence of the density autocorrelation function as the MCT derived for atomic systems. This means that all the analytical conclusions [6] of the original MCT remain true for the glass-transition scenario in colloidal systems.

II. MEMORY FUNCTION REPRESENTATION

We consider a suspension of identical colloidal particles interacting via a pairwise additive potential $V(r)$. Familiar choices for $V(r)$ are a hard-sphere potential for sterically stabilized and a Derjaguin-Landau-Verwey-Overbeek-type Yukawa potential for charge-stabilized colloidal suspensions. The basic quantity that is studied

within the mode-coupling approach is the intermediate scattering function, i.e., the Fourier transform of the density autocorrelation function

$$F(k; t) = \frac{1}{N} \langle n(\mathbf{k}) e^{\hat{\Omega}_S t} n(-\mathbf{k}) \rangle. \quad (1)$$

Here, $n(\mathbf{k})$ denotes a Fourier component of the density of suspended particles

$$n(\mathbf{k}) = \sum_{i=1}^N \exp(-i\mathbf{k} \cdot \mathbf{r}_i), \quad (2)$$

and the brackets $\langle \dots \rangle$ denote the equilibrium average. We take the convention that the equilibrium distribution $P_{\text{eq}} \sim \exp[-\sum_{i \neq j} V(r_{ij})/k_B T]$ is formally represented by the ket vector \dots .

In Eq. (1), $\hat{\Omega}_S$ denotes the N -particle Smoluchowski operator that in absence of hydrodynamic interactions is given by the formula [2]

$$\hat{\Omega}_S = D_0 \sum_{i=1}^N \frac{\partial}{\partial \mathbf{r}_i} \cdot \left[\frac{\partial}{\partial \mathbf{r}_i} - \beta \mathbf{F}_i \right]. \quad (3)$$

Here, D_0 is the Stokes-Einstein diffusion coefficient of a single particle, $\beta = 1/k_B T$, and $\mathbf{F}_i = -\sum_{j \neq i} \partial V(r_{ij})/\partial \mathbf{r}_i$ is the force on particle i . Note that in Eq. (1) the evolution operator $\hat{\Omega}_S$ acts on everything to its right, also on the equilibrium distribution P_{eq} .

Starting from the formal expression (1), Cichocki and Hess [19] derived a very useful memory function representation of the intermediate scattering function:

$$F(k; z) = \frac{S(k)}{z + D(k; z)k^2}, \quad (4)$$

$$D(k; z) = \frac{D_0/S(k)}{1 + M(k; z)}. \quad (5)$$

Here, $F(k; z)$ is the Laplace transform of the intermediate scattering function

$$F(k; z) = \int_0^\infty dt e^{-zt} F(k; t) \quad (6)$$

and $S(k)$ is the static structure factor, $S(k) = F(k; t=0)$.

In Eq. (5), $M(k; z)$ is the Laplace transform of the memory function $M(k; t)$:

$$M(k; t) = D_0 k^2 \beta^2 \frac{1}{N} \langle \sigma(\mathbf{k}) e^{\hat{\Omega}_S^{\text{irr}} t} \sigma(-\mathbf{k}) \rangle, \quad (7)$$

where $\sigma(\mathbf{k})$ is the microscopic expression for the momentum-averaged longitudinal stress fluctuations

$$\sigma(\mathbf{k}) = -\hat{Q}_c \sum_{i=1}^N (i\mathbf{k} \cdot \mathbf{F}_i / k^2 + k_B T) \exp(-i\mathbf{k} \cdot \mathbf{r}_i), \quad (8)$$

$\hat{Q}_c = 1 - \hat{P}_c$, and \hat{P}_c is a projection operator on the density subspace

$$\hat{P}_c = \dots n(-\mathbf{k}) \frac{1}{NS(k)} \langle n(\mathbf{k}) \dots \rangle. \quad (9)$$

The evolution operator $\hat{\Omega}_S^{\text{irr}}$ in Eq. (7) is the ‘‘one-particle irreducible Smoluchowski operator’’ [19]

$$\hat{\Omega}_S^{\text{irr}} = \hat{Q}_c D_0 \sum_{i=1}^N \frac{\partial}{\partial \mathbf{r}_i} \hat{Q}_i \cdot \left[\frac{\partial}{\partial \mathbf{r}_i} - \beta \mathbf{F}_i \right] \hat{Q}_c, \quad (10)$$

where $\hat{Q}_i = 1 - \hat{P}_i$, and the projection operator \hat{P}_i is given by the formula

$$\hat{P}_i = \dots \exp(i\mathbf{k} \cdot \mathbf{r}_i) \rangle \langle \exp(-i\mathbf{k} \cdot \mathbf{r}_i) \dots. \quad (11)$$

Cichocki and Hess have shown that $M(k; z)$ is, essentially, the contribution to the generalized friction coefficient due to interactions of the suspended particles. It is also closely related to the longitudinal component of the generalized dynamic viscosity.

III. MODE-COUPLING APPROXIMATION

To construct a MCT of the structural relaxation in colloidal systems we follow the approach of Ref. [6]. We do not try to substantiate the approximations involved [21] as the basic aim of this paper is to investigate the universality of the MCT predictions. We start with the memory function (7) and make the following approximations. *First* we project stress fluctuations σ onto the subspace of density products:

$$\begin{aligned} \sigma(-\mathbf{k}) &\approx \sum_{(\mathbf{k}_1, \mathbf{k}_2)} \sum_{(\mathbf{k}_3, \mathbf{k}_4)} n(-\mathbf{k}_1) n(-\mathbf{k}_2) \\ &\quad \times g(\mathbf{k}_1 \mathbf{k}_2; \mathbf{k}_3 \mathbf{k}_4) \\ &\quad \times \langle n(\mathbf{k}_3) n(\mathbf{k}_4) \sigma(-\mathbf{k}) \rangle. \end{aligned} \quad (12)$$

Here the sums over $\mathbf{k}_i < \mathbf{k}_{i+1}$, $i=1, 3$, are understood, and g denotes the normalization matrix

$$\begin{aligned} \sum_{(\mathbf{k}_3, \mathbf{k}_4)} g(\mathbf{k}_1 \mathbf{k}_2; \mathbf{k}_3 \mathbf{k}_4) \langle n(\mathbf{k}_3) n(\mathbf{k}_4) n(-\mathbf{k}_5) n(-\mathbf{k}_6) \rangle \\ = \delta_{\mathbf{k}_1 \mathbf{k}_5} \delta_{\mathbf{k}_2 \mathbf{k}_6}. \end{aligned} \quad (13)$$

The *second approximation* consists of factorizing averages of products that result from substituting projection (12) into (7), and replacing the irreducible operator $\hat{\Omega}_S^{\text{irr}}$ with the operator $\hat{\Omega}_S$:

$$\begin{aligned} \langle n(\mathbf{k}_1) n(\mathbf{k}_2) e^{\hat{\Omega}_S^{\text{irr}} t} n(-\mathbf{k}_3) n(-\mathbf{k}_4) \rangle \\ \approx \langle n(\mathbf{k}_1) e^{\hat{\Omega}_S t} n(-\mathbf{k}_3) \rangle \langle n(\mathbf{k}_2) e^{\hat{\Omega}_S t} n(-\mathbf{k}_4) \rangle. \end{aligned} \quad (14)$$

Consistently, for the normalization matrix g we get

$$g(\mathbf{k}_1 \mathbf{k}_2; \mathbf{k}_3 \mathbf{k}_4) = \frac{\delta_{\mathbf{k}_1 \mathbf{k}_3} \delta_{\mathbf{k}_2 \mathbf{k}_4}}{N^2 S(\mathbf{k}_1) S(\mathbf{k}_2)}. \quad (15)$$

Substituting Eqs. (12), (14), and (15) into Eq. (7), we obtain the mode-coupling contribution to the memory function. To get explicit results we first need to calculate $\langle n(\mathbf{k}_1) n(\mathbf{k}_2) \sigma(-\mathbf{k}) \rangle$. There are two terms contributing to this expression. The first one can be easily calculated:

$$\begin{aligned}
\left\langle n(\mathbf{k}_1)n(\mathbf{k}_2) \sum_{i=1}^N (i\mathbf{k} \cdot \mathbf{F}_i / k^2 - k_B T) \exp(i\mathbf{k} \cdot \mathbf{r}_i) \right\rangle &= \frac{ik_B T \mathbf{k}}{k^2} \left\langle n(\mathbf{k}_1)n(\mathbf{k}_2) \sum_{i=1}^N \frac{\partial}{\partial \mathbf{r}_i} \exp(i\mathbf{k} \cdot \mathbf{r}_i) \right\rangle \\
&= -\frac{ik_B T \mathbf{k}}{k^2} \left\langle \sum_{i=1}^N \exp(i\mathbf{k} \cdot \mathbf{r}_i) \left[\frac{\partial}{\partial \mathbf{r}_i} n(\mathbf{k}_1)n(\mathbf{k}_2) \right] \right\rangle \\
&= -\frac{k_B T}{k^2} \delta_{\mathbf{k}_1+\mathbf{k}_2, \mathbf{k}} N [\mathbf{k} \cdot \mathbf{k}_1 S(k_2) + \mathbf{k} \cdot \mathbf{k}_2 S(k_1)] . \tag{16}
\end{aligned}$$

The second contribution to $\langle n(\mathbf{k}_1)n(\mathbf{k}_2)\sigma(-\mathbf{k}) \rangle$ involves the projection operator \hat{P}_c . It is given by a three-particle average. As the *third approximation* we calculate it using the convolution approximation [4]:

$$\begin{aligned}
-\left\langle n(\mathbf{k}_1)n(\mathbf{k}_2)\hat{P}_c \sum_{i=1}^N (i\mathbf{k} \cdot \mathbf{F}_i / k^2 - k_B T) \exp(i\mathbf{k} \cdot \mathbf{r}_i) \right\rangle &= -\langle n(\mathbf{k}_1)n(\mathbf{k}_2)n(-\mathbf{k}) \rangle \frac{1}{NS(k)} \\
&\quad \times \left\langle n(\mathbf{k}) \sum_{i=1}^N (i\mathbf{k} \cdot \mathbf{F}_i / k^2 - k_B T) \exp(i\mathbf{k} \cdot \mathbf{r}_i) \right\rangle \\
&\approx N \delta_{\mathbf{k}_1+\mathbf{k}_2, \mathbf{k}} S(k)S(k_1)S(k_2) \frac{1}{NS(k)} \frac{Nk_B T}{k^2} . \tag{17}
\end{aligned}$$

Adding contributions (16) and (17) we get

$$\langle n(\mathbf{k}_1)n(\mathbf{k}_2)\sigma(-\mathbf{k}) \rangle = -\frac{k_B T}{k^2} \delta_{\mathbf{k}_1+\mathbf{k}_2, \mathbf{k}} N n S(k_1)S(k_2) [\mathbf{k} \cdot \mathbf{k}_1 c(k_1) + \mathbf{k} \cdot \mathbf{k}_2 c(k_2)] . \tag{18}$$

Here, $c(k)$ is the Ornstein-Zernike direct correlation function [4] and n denotes the density of suspended particles. Finally, using Eqs. (7), (12), (14), (15), and (18) we obtain the following expressions for the mode-coupling contribution to the memory function:

$$M^{\text{MC}}(k; t) = \frac{nD_0}{2k^2} \int \frac{d\mathbf{k}_1}{(2\pi)^3} [\mathbf{k} \cdot \mathbf{k}_1 c(k_1) + \mathbf{k} \cdot (\mathbf{k} - \mathbf{k}_1) c(|\mathbf{k} - \mathbf{k}_1|)]^2 F(k_1; t) F(|\mathbf{k} - \mathbf{k}_1|; t) . \tag{19}$$

The *fourth approximation* consists of replacing all the other contributions to $M(k; t)$ by a white-noise term $\nu(k)$. In this way we obtain the final expression for the memory function:

$$M(k; t) = \nu(k) + M^{\text{MC}}(k; t) . \tag{20}$$

For further analysis it is convenient to introduce the *density correlator*

$$\phi_k(t) = F(k; t) / S(k) \tag{21}$$

and to write down the equation of motion for the correlator in the following form:

$$\frac{\phi_k(z)}{1 - z\phi_k(z)} = \frac{1 + \nu(k) + \omega_k m(k, z)}{\omega_k} . \tag{22}$$

Here, $\phi_k(z)$ is the Laplace transform of the correlator, $\omega_k = D_0 k^2 / S(k)$ is the characteristic frequency for BD, and $m(k, z)$ is the Laplace transform of the reduced memory function $m(k, t) = M^{\text{MC}}(k; t) / \omega_k$. One should note that the reduced memory function derived here has precisely the same form as that derived for MD in Ref. [6]. Let us now assume that Eq. (22) leads to a glass transition, i.e., that at a certain density and temperature the long-time limit of the correlator $\phi_k(t)$ [and, via Eq. (19), the long-time limit of the memory function] is greater than 0. Then, near the glass transition, for small z the memory function $m(k, z)$ is very large. This suggests that for small z (large times) we can approximate Eq. (22) by

the following one:

$$\phi_k(z) / [1 - z\phi_k(z)] = m(k, z) . \tag{23}$$

In this way we obtain the same equation of motion for the asymptotic long-time dynamics near the glass transition as that derived for MD [6] (the minus sign in the denominator on the left-hand side of (23) is connected to a difference in the definition of the Laplace transform used here and in Ref. [6]). So whereas at short times BD and MD give different results (compare the short-time expansions of the intermediate scattering functions for BD and MD given in Ref. [2]), the asymptotic form of $\phi(k, t)$ for long times is the same. This implies that all analytical results based on Eq. (23) remain the same for BD [22].

IV. DISCUSSION

We have presented a simple mode-coupling theory for the kinetic glass transition in colloidal liquids, where the short-time dynamics is Brownian. The theory leads to the same asymptotic decay of the density fluctuations as its atomic counterpart. An *ideal* glass transition is predicted, i.e., the structure remains frozen for infinite times at the glass-transition point, or equivalently, the ergodicity is broken. In reality, activated hopping processes may restore ergodicity on a very-long-time scale. The exact nature of these jump processes is complicated, but can probably be understood as a collective exchange of particle positions of a few particles or whole particle clusters.

The actual presence of thermally activated jumps seems to depend sensitively on the microscopic interactions; for example, they can be suppressed by local charge ordering in molten salts. In order to incorporate activated hopping processes, an extension of simple MCT was recently proposed [23] for atomic systems. Within the extended theory the ideal glass transition changes to a smooth crossover. Hence it is clear that a direct comparison of the asymptotic predictions of our simple MCT is only possible if activated processes are strongly suppressed so that one is in the asymptotic regime of an ideal glass transition. A good candidate for comparison is a sterically stabilized colloidal suspension. The experimental data [1] were compared with simple MCT for an atomic hard-sphere system and remarkable agreement was found [15], although the real colloidal short-time dynamics is completely different. This agrees with our result that the MCT predictions are robust with respect to different short-time dynamics (BD or MD). At least for the asymptotic decay of density fluctuations, the MCT predictions seem to be compatible with data of the experimental system whose true short-time dynamics is even more complicated than BD due to solvent-mediated hydrodynamic interactions.

As regards the recent BD-MD simulation of a strongly charge-polydisperse charge-stabilized colloidal suspension [11], the thermally activated hopping events are more pronounced in this system. As was shown by studying the behavior of high-charge and low-charge particles separately [8], this can be attributed to the relative high mobility of low-charge particles. In the simulation the glass transition point turned out to be independent of short-time dynamics (MD or BD), and the wave-vector dependence of the Edwards-Anderson parameter, defined as $\phi_k(t_0)$ where t_0 is a time typically lying between β -relaxation and activated hopping processes, was found to be very similar for MD and BD. Although the presence of hopping processes make a direct comparison with the asymptotic predictions of MCT more difficult, this is in agreement with our result that asymptotic features near the glass transition do not depend on the short-time dynamics. On the other hand, in the simulation, the relaxational behavior was qualitatively different for intermediate times that are larger than short-time dynamics but

still smaller compared with a time for thermally activated hopping events. Since MCT only makes asymptotic predictions for long times, this different dynamical onset of β relaxation, induced by the different short-time dynamics (MD or BD), is not captured in our approach. The behavior of β relaxation for longer times, however, can be well fitted by MCT to experimental data [15].

We end with three comments. First, one should think of hydrodynamic interactions between the colloidal particles. This type of interaction can be incorporated in the theory by replacing the short-time diffusion constant D_0 by a $3N \times 3N$ diffusion tensor that depends explicitly on the positions of N colloidal particles [2]. The exact expression for this diffusion matrix is not known. It has been found [2] that the very simple way of incorporating the hydrodynamic interactions [24], i.e., replacing D_0 by the actual, density-dependent short-time self-diffusion constant, leads to very good predictions for the so-called long-time self-diffusion coefficient. If the same idea could be used to describe time-dependent processes hindered by the hydrodynamic interactions, the main result of this paper, i.e., Eq. (23), would remain valid. A second remark concerns Fokker-Planck dynamics, which can be viewed as interpolating between Newtonian and Brownian dynamics. This kind of dynamics is relevant, e.g., for ionic solutions [25]. The asymptotic form of the corresponding equation for the density correlator does not change within MCT and consequently the same conclusions apply to this case. Finally, as already mentioned, it has been found for atomic systems with Newtonian dynamics that an extension of the simple MCT changes the ideal glass transition to a continuous crossover [23]. It would be interesting to investigate whether the glass transition predicted by the MCT for colloidal systems is stable against extensions of a simple approach presented here.

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