Glassy dynamics of partially pinned fluids: An alternative
mode-coupling approach

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Abstract – We propose a simple mode-coupling theory for glassy dynamics of partially pinned fluids. Our approach is different from the mode-coupling theory developed by Krakoviack (Phys. Rev. Lett., 94 (2005) 065703; Phys. Rev. E, 84 (2011) 050501(R)). In contrast to Krakoviack’s theory, our approach predicts a random pinning glass transition scenario that is qualitatively the same as the scenario obtained using a mean-field analysis of the spherical \( p \)-spin model and a mean-field version of the random first-order transition theory. We use our approach to calculate quantities which are considered to be indicators of growing dynamic correlations and static point-to-set correlations. We find that the so-called static overlap is dominated by the simple, low pinning fraction contribution. Thus, at least for randomly pinned fluid systems, only a careful quantitative analysis of simulation results can reveal genuine, many-body point-to-set correlations.

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Introduction. – Recently there have been several theoretical and simulational studies of glassy dynamics of fluid systems in which some particles, randomly selected out of an equilibrium configuration, have been frozen or pinned [1–9]. Originally these so-called partially pinned systems were considered to be just one special example of a broad class of model porous systems known as quenched-annealed mixtures [10–12]. However, it has now been realized that glassy partially pinned systems can be used to reveal still unresolved aspects of the glass transition.

First, it was proposed that one can study a growing “amorphous order” that is supposed to develop in glassy fluids by analyzing systems in which some particles, which are taken out of an equilibrium configuration, are frozen [13]. In early studies this idea was implemented using the so-called cavity geometry: all particles except those within a spherical cavity were frozen and the local overlap of the original configuration with configurations equilibrated in the presence of frozen particles was monitored [14]. It was argued that the dependence of this overlap on the cavity diameter reveals a length characterizing the so-called static point-to-set correlations, i.e., correlations between the density at the center of the cavity (point) and the positions of the frozen particles (set). It was shown [15] that, at least in simple models, these point-to-set correlations grow with increasing relaxation time. Subsequently, other geometries were introduced: one in which all particles except those in a layer are frozen (the sandwich geometry), one in which all particles in a semi-infinite space are frozen (the wall geometry) [16] or one in which a randomly chosen subset of particles, distributed uniformly throughout the system, is frozen. It has been argued [5] that the last geometry, i.e., the partially pinned system, is the best candidate to study growing static correlations.

The second motivation for the recent interest in partially pinned systems comes from the realization that pinned particles, while maintaining the equilibrium structure of the fluid [11,17], may induce an ideal glass transition at temperatures or densities that are more accessible to computer simulation studies [3,8]. Thus, the analysis of this so-called random pinning glass transition [9] could both shed light on the glass transition itself and provide a new way to test diverse theoretical descriptions used to describe it. In particular, Krakoviack [4] recently argued that two different approaches, the mode-coupling theory [18] and the random first-order theory (RFOT) [19] make strikingly different predictions for the glassy behavior of partially pinned systems. Thus, a simulational study of glassy behavior of a partially pinned system could easily disprove one of these approaches.

The conclusion reached in ref. [4] was surprising for two reasons. First, it is usually assumed that mode-coupling theory is, albeit in some not well understood...
way, included in the RFOT approach. Specifically, the mode-coupling theory is applicable to and describes rather well the onset of glassy behavior. It predicts a sharp ergodicity breaking transition referred to as the mode-coupling transition. The RFOT approach replaces this transition with a crossover and provides an alternative description of the properties of deeply super-cooled fluids.

It has to be admitted that recent theoretical analyses of these theories in higher dimensions revealed some rather disturbing discrepancies [20–22]. In spite of this fact, qualitative disagreement between them in three dimensions was not expected. Second, Cammarota and Biroli [9] showed that a mean-field analysis of the p-spin model with partially pinned spins predicts qualitatively the same random pinning glass transition scenario as the mean-field version of the RFOT. Thus, we are now faced with a rather unpleasant qualitative disagreement between two mean-field-like calculations, mode-coupling theory and the mean-field p-spin model. This disagreement is even more striking if we recall that in the case of an un-pinned system the so-called schematic model of mode-coupling theory is identical to the p-spin model (for \( p = 3 \) used in ref. [9]).

Faced with the above-described conundrum, we shall recall that there is some freedom in the formulation of the mode-coupling approach, especially for more complex systems like mixtures and partially pinned systems. Our goal in this letter is to propose a simple, alternative mode-coupling approach that predicts the random pinning glass transition scenario which is qualitatively consistent with the mean-field analysis of both the p-spin model [9] and the RFOT theory [3]. In addition, we will use our approach to calculate quantities that have been used in earlier simulational studies to monitor the growth of dynamic correlations and static point-to-point correlations. We will show that a careful analysis of these quantities is required to reveal genuine many-body effects.

We should note that our approach is applicable to “standard” partially pinned systems, i.e., systems in which the particles to be pinned are selected from the equilibrium configuration corresponding to the state point of which the behavior of the remaining particles is studied. Our approach would have to be modified for systems in which a more general pinning procedure is used, i.e., in which the particles to be pinned are selected from the equilibrium configuration corresponding to a state point different from the state point at which the behavior of the remaining particles is studied (e.g., a higher-temperature state point [23,24]). Cammarota and Biroli’s [3,9] and Krakoviack’s [4] calculations lead to qualitatively different results in the former case and seem to agree in the latter one.

Derivation of an alternative mode-coupling approach. – We will outline a projection operator derivation of our approach, which is easily compared with Krakoviack’s theory [4,25]. Following refs. [4,25] we will refer to the mobile (un-pinned) particles as the fluid particles and to the pinned ones as the matrix particles. The fundamental dynamical variable used in our theory is the Fourier transform of the microscopic fluid density, \( n_f(q) = \sum_{j=1}^{N_f} e^{-iqr_j} \), where \( r_j \) denotes the position of the \( j \)-th fluid particle and \( N_f \) is the number of fluid particles. To describe the dynamics of the system, we use the fluid intermediate scattering function,

\[
F(q;t) = \frac{1}{N_f} \langle n_f(q) \exp(i\Omega t) n_f(-q) \rangle,
\]

with \( \Omega \) being the system’s evolution operator and \( \langle \ldots \rangle \) denoting the average over all (fluid and matrix) particles of the system. For simplicity, we assume here that the microscopic dynamics is Brownian. Thus, \( \Omega \) is the many-body Smoluchowski operator, \( \Omega = D \sum_{j=1}^{N_f} \partial_{r_j} \cdot (\partial_{r_j} - \beta F_j) \), with \( D \) being the diffusion coefficient of an isolated fluid particle, \( \beta = 1/(k_BT) \) and \( F_j \) denoting a force acting on fluid particle \( j \) (note that this force is due to both fluid and matrix particles). To derive an equation of motion for \( F(q;t) \) we follow the standard procedure [18,26] and arrive at an exact but formal equation involving the so-called irreducible memory function [27],

\[
\int_0^t du \left( \delta(t-u) + M^{irr}(q;t-u) \right) \partial_u F(q;u) = \sum_{j=1}^{N_f} D_q^2 n_f m(c) h(q) - D_q^2 (1-n_f c(q)) F(q;t).
\]

In eq. (2) \( n_f \) and \( n_m \) are the densities of the fluid and matrix particles, respectively, and \( h(q) \) and \( c(q) \) are the Fourier transforms of the complete system’s correlation function and direct correlation function, respectively. Finally, \( M^{irr}(q;t) \) is the irreducible memory function,

\[
M^{irr}(q;t) = \frac{1}{DN_f} Q \cdot \langle j(q) \exp(i\Omega^{irr} t) j(-q) \rangle \cdot \dot{q}.
\]

In eq. (3) \( j(q) \) is the projected current density,

\[
j(q) = Q D \sum_{j=1}^{N_f} (-i q + \beta F_j) e^{-iqr_j}
\]

with \( Q = I - \mathcal{P} \) and \( \mathcal{P} \) is the projection operator on the space spanned by the fluid density, \( n_f(q) \), and the matrix density, \( n_m(q) \), where \( n_m(q) = \sum_{j=1}^{N_m} e^{-iqr_j} \) with \( r_j \) being the position of the \( j \)-th matrix particle, and \( N_m \) being the number of the matrix particles. Finally, in eq. (3) \( \Omega^{irr} \) is the one-particle irreducible Smoluchowski operator [27],

\[
\Omega^{irr} = \sum_{j=1}^{N_f} \partial_{r_j} Q \cdot (\partial_{r_j} - \beta F_j) Q,
\]

where \( Q_j = I - P_j \) and \( P_j \) is the projection operator on the Fourier transform of a single-particle density, \( P_j = \ldots e^{-iqr_j} \langle e^{-iqr_j} \rangle \ldots \). Next, we project the current densities \( j(q) \) on the space spanned by the products of the fluid densities, \( n_f(k) n_f(q-k) \), and the products of the fluid and matrix densities, \( n_f(k) n_m(q-k) \). More precisely, we use only the parts of these products that are orthogonal to the single-particle densities, i.e., we project on \( Q n_f(k) n_f(q-k) \)

\[\text{[1] In the derivation of eqs. (2), (6)–(9) we used simplifications concerning static correlation functions of a partially pinned system [11].} \]
and \( Qn^f(k)n^\alpha(q-k) \) (see footnote 2). For a Brownian system this step is exact; in contrast, for a system evolving with Newtonian dynamics an analogous step involves an approximation [18].

As a result of the projection described in the previous paragraph, we obtain an expression for the irreducible memory function in terms of four-point correlation functions. We factorize [18,26] the four-point functions and at the same time replace the irreducible Smoluchowski operator \( \Omega^\text{irr} \) by the un-projected Smoluchowski operator, \( \Omega \),

\[
\langle n^a(q_1) n^b(q_2) \exp(\Omega^\text{irr} t) \rangle \approx \langle n^a(q_1) \exp(\Omega t) n^b(-q_3) \rangle \langle n^b(q_2) \exp(\Omega t) n^d(-q_4) \rangle + \langle n^a(q_1) \exp(\Omega t) n^b(-q_3) \rangle \langle n^b(q_2) \exp(\Omega t) n^d(-q_4) \rangle, \tag{5}
\]

where pairs \( a, b \) and \( c, d \) denote \( f, f \) or \( f, m \). In addition, we also factorize analogous time-independent four-point functions that enter in the definition of the projection operator on density products. Next, we use a mixture version of the convolution approximation [18]. Finally, we use the time independence of the matrix density.

In this way, we obtain the following approximate expression for the irreducible memory function:

\[
M^\text{irr}(q; t) = \int \frac{dk}{(2\pi)^3} \left[ V^2(q, k) F(k; t) F(|q-k|; t) \right. \\
\left. + V^1(q, k) F(k; t) + V^0(q, k) \right], \tag{6}
\]

where

\[
V^2(q, k) = \frac{n_f D}{2} |\textbf{q} \cdot \textbf{k} c(k) + \textbf{q} \cdot (\textbf{q} - \textbf{k}) c(|\textbf{q} - \textbf{k}|)|^2, \tag{7}
\]

\[
V^1(q, k) = n_m D |\textbf{q} \cdot (\textbf{q} - \textbf{k}) c(|\textbf{q} - \textbf{k}|)|^2 \\
\times [1 + n_m h(|\textbf{q} - \textbf{k}|) + 2n_m n_m h(|\textbf{q} - \textbf{k}|) \cdot (\textbf{q} - \textbf{k}) c(|\textbf{q} - \textbf{k}|) \\
\times |\textbf{q} \cdot \textbf{k} c(k) + \textbf{q} \cdot (\textbf{q} - \textbf{k}) c(|\textbf{q} - \textbf{k}|)| h(\textbf{q} - \textbf{k}), \tag{8}
\]

\[
V^0(q, k) = D n_f n^m \textbf{q} \cdot \textbf{k} c(k) \textbf{q} \cdot (\textbf{q} - \textbf{k}) c(|\textbf{q} - \textbf{k}|) \\
\times h(\textbf{q} - \textbf{k}) h(|\textbf{q} - \textbf{k}|). \tag{9}
\]

Comparison with an earlier mode-coupling theory. — Before turning to the results obtained from eqs. (2), (6)–(9) we compare our present approach to Krakoviack’s [4,25]. He also follows the projection operator procedure detailed in ref. [18]. However, he takes the fundamental dynamical variable the so-called relaxing part of the fluid density, \( \delta n^f(k) = n^f(q) - \langle n(q) \rangle \) where \( \langle \ldots \rangle \) denotes the equilibrium average over the positions of the fluid particles only, with the matrix particles treated as a set of fixed obstacles. Furthermore, he projects the fluctuating force on the space spanned by the products of the relaxing fluid densities, \( \delta n^f(k) \delta n^f(q-k) \), the products of the relaxing fluid density and the matrix density, \( \delta n^f(k) n^\alpha(q-k) \), and the products of the relaxing fluid density and the average fluid density, \( \delta n^f(k) \langle n(q-k) \rangle \)\(^f \). We note that the variables that we use are either one-particle or pairwise additive, which is natural for systems with pairwise additive interactions. In contrast, the average fluid density \( \langle n(q) \rangle \)\(^f \) used in ref. [25] is not one-particle or pairwise additive in terms of the matrix particles.

The main difference between our and Krakoviack’s approaches seems to be that we use the mode-coupling approach to find both the time evolution of the relaxing density fluctuations and the average non-relaxing density fluctuations. In our language the latter quantity is the long-time limit of the fluid intermediate scattering function, \( F(q) = \lim_{t \to \infty} F(q; t) \). In contrast, Krakoviack uses the mode-coupling approach only to find the time evolution of the relaxing density fluctuations and resorts to a separate static approach, replica OZ integral equation theory [10,11], to analyze the non-relaxing density fluctuations. In his language the latter fluctuations are characterized by the so-called blocking (disconnected) fluid structure factor, \( S^b(q) = \frac{1}{17} \langle n(q) \rangle \langle n(-q) \rangle \)\(^f \), where \( \langle \rangle \)\(^f \) denotes the average over the matrix realizations.

We should point out, however, that the self-consistent equation for \( F(q) \) that follows from eqs. (2), (6)–(9) can also be obtained from a static approach. This equation has the exact same structure as the equation for \( S^b(q) \) derived from replica OZ equations used by Krakoviack [11],

\[
F(q) = \frac{n_m n_f c(q) h(q)}{1 - n_f c(q) - n_f \tilde{c}(q)} + \frac{n_f \tilde{c}(q)(1 + n_f h(q))}{1 - n_f c(q) - n_f \tilde{c}(q)}, \tag{10}
\]

where the blocking direct correlation function \( \tilde{c} \) (denoted by \( c^b \) in ref. [11]) is related to the long-time limit of the irreducible memory function,

\[
\tilde{c}(q) = \lim_{t \to \infty} M^\text{irr}(q; t)/(Dq^2 n_f) \tag{11}
\]

(see also eq. (49) of ref. [11]). Equation (11) expresses \( \tilde{c}(q) \) in terms of \( F(q) \) and equilibrium correlation functions, and thus serves as a closure relation for the replica OZ equations. Importantly, this closure can be derived\(^3\) independently of mode-coupling equations (6)–(9), using a generalization of the static approach of ref. [28].

Thus, the main difference between our and Krakoviack’s approaches is that we use approximations for the statics and dynamics which, by construction, are consistent whereas he uses separate approaches for the statics and the dynamics. In particular, our \( \tilde{c} \) is constrained by eq. (11) while Krakoviack can choose any closure for \( \tilde{c} \) (see footnote 4).

\(^2\)The parts of the products proportional to the single-particle densities do not contribute due to the presence of the projection \( Q \) in the definition of the projected current density (4).

\(^3\)The derivation will be presented elsewhere.

\(^4\)Krakoviack uses the Madden-Glandt approximation, \( \tilde{c} = 0 \), which, upon identification of \( F(q) \) with \( S^b(q) \), reduces eq. (10) to eq. (36d) of ref. [11].
We should note that both the static analysis of ref. [11] and our static calculation would have to be modified if particles to be pinned were selected from the equilibrium configuration corresponding to a state point different from the state point at which the behavior of the remaining particles is studied.

Equation (10) implies that for fluid states the small pinning fraction limit of $F(q)$ has the following simple form:

$$F(q) = xn^2h^2(q) + O(x^2). \quad (12)$$

Here $x$ is the fraction of particles that are pinned, $x = n_m/n$, with $n = n_f + n_m$ being the total density of the system (in refs. [3,5,9,24] the pinning fraction is denoted by $\epsilon$; we changed the notation to avoid confusing the pinning fraction and the direct correlation function).

Two arguments were put forward for the approach used in refs. [4,25]. First, since the calculation of the properties of non-relaxing fluid density fluctuations is a static problem, it is appropriate to use the mode-coupling approach for the dynamics of the relaxing part only. Second, in the formulation of refs. [4,25] the so-called connected fluid structure factor, $S^c(q) = \frac{1}{n_f} \langle \delta n_f(q) \delta n_f(-q) \rangle$, appears naturally in the expression for the so-called characteristic frequency [25], which is given by $\omega(q) = Dq^2/S^c(q)$ for a Brownian system.

To answer these arguments we note that, as discussed above, our approach can be considered a combination of a specific static calculation that is consistent with the mode-coupling prediction for the non-decaying part of $F(q;t)$ and the mode-coupling calculation of the decaying part of $F(q;t)$. Moreover, if the equation of motion is re-written in terms of the decaying part only, the characteristic frequency acquires the form $\omega(q) = Dq^2/(S^f(q) - F(q))$, where $S^f(q) = F(q;t = 0) = 1 + n_fh(q)$ is the fluid static structure factor. The functional form of the characteristic frequency is identical to that obtained by Krakoviack [25].

Our mode-coupling equations (2), (6)–(9) can also be derived in other ways. In particular, we can start from a standard mode-coupling theory for a binary mixture in which the only difference between the two components is the diffusion coefficient of the isolated particles, equal to $D$ for the first component and $D_f$ for the second one. It can be showed that in the limit of $D_f/D \to 0$ (and for finite times) we recover eqs. (2), (6)–(9) for the intermediate scattering function of the fast component (see also fig. 2).

Random pinning glass transition scenario. – Equations (2), (6)–(9) allow us to calculate both $F(q)$ and the time dependence of $F(q;t)$. To solve these equations numerically we discretized the wave vector space using 500 wave vectors with the smallest one equal to 0.05 and a spacing of 0.1. The resulting set of coupled integro-differential equations was solved using the procedure outlined in ref. [29]. We used the hard-sphere interaction potential and the Percus-Yevick approximation for the static correlations, $h(q)$ and $c(q)$. The results below are presented using reduced units: distance is measured in terms of the hard-sphere diameter $\sigma$ and time in terms of $\sigma^2/D$.

The random pinning glass transition phase diagram is shown in fig. 1(a). The standard mode-coupling transition present at $x = 0$ extends into the $x > 0$ plane. However, as shown in fig. 1(b), the discontinuity of the long-time limit of the intermediate scattering function at this transition decreases with increasing $x$ and disappears at $x_c \approx 0.1395$. Beyond $x_c$, $F(q)$ changes continuously with the volume fraction $\varphi = n\pi/6$. This scenario is qualitatively consistent with the $p$-spin model results presented in ref. [9] and with the mean-field RFOT analysis presented in ref. [3].

In fig. 1(a) we also indicate the localization transition [30], i.e., the line of the vanishing self-diffusion coefficient of the fluid particles. The localization transition is qualitatively similar to that predicted by Krakoviack’s approach.

The time dependence of the fluid intermediate scattering function is presented in fig. 2. Thick solid lines in this figure illustrate relaxation in the vicinity of the mode-coupling transition line. We see the familiar evolution of the time dependence predicted by the standard mode-coupling calculations: upon approaching a transition the intermediate-time plateau of the scattering function extends to longer and longer times and the relaxation time diverges. At the transition there is a discontinuous change of the long-time limit of the intermediate scattering function. The presence of pinned particles is reflected in the presence of a non-zero long-time plateau of the intermediate scattering function even on the ergodic side of the mode-coupling transition. This plateau is a manifestation of the frozen-in density fluctuations induced by...
the randomly pinned particles. Thin solid lines in fig. 2 illustrate the evolution of the time dependence of the intermediate scattering function for pinning fractions exceeding the critical value $x_c$. One sees that in this case the long-time limit of the scattering function increases continuously, and no divergence of the relaxation time is visible. We checked that a suitably defined characteristic time $\tau$ where $F(r; t)$ is the inverse Fourier transform of $F(q; t)$ and $Q_{\text{rand}} = 4\pi n a^3/3$. In the definition of the single-particle overlap $Q_{\text{self}}(t)$, $F(r; t)$ is replaced by the inverse Fourier transform of the self-intermediate scattering function $F^s(q; t)$ [30] and $Q_{\text{rand}}$ is absent. To make connection with ref. [5] we chose $a = 0.3$.

In fig. 3 we show the time dependence of the collective and single-particle overlaps for $\varphi = 0.51$. For smaller $x$ our predictions are qualitatively similar to computer simulation results showed in fig. 2(c) of ref. [5]. However, upon approaching the mode-coupling transition $Q(t)$ predicted by the theory shows a classic mode-coupling–like two-step decay, whereas the simulation results exhibit a continuous increase of both the intermediate-time plateau and the long-time plateau.

In fig. 4(a) we show the length dependence of the so-called static overlap, i.e., the non-trivial part of the long-time plateau of the collective overlap, $Q_{\infty} - Q_{\text{rand}}$, where $Q_{\infty} = \lim_{t \to \infty} Q(t)$. The length, $l = 0.5 x^{-1/3}$, is the so-called confining length [5]. Perhaps fortuitously, for the range of lengths corresponding to non-glassy states, the values of $Q_{\infty} - Q_{\text{rand}}$ are quite close to those obtained from computer simulations (see fig. 4(a) of ref. [5]). However, according to our mode-coupling approach an increase of $Q_{\infty} - Q_{\text{rand}}$ is followed by a discontinuity at the mode-coupling transition, whereas the values obtained from simulations increase continuously with decreasing $l$. Importantly, growing values of $Q_{\infty} - Q_{\text{rand}}$ are often associated with growing amorphous order and, more specifically, growing static point-to-set correlations. As indicated in fig. 4(a) (and also noted in ref. [5]), for the range of lengths corresponding to non-glassy states, $Q_{\infty} - Q_{\text{rand}}$ is dominated by the small-$x$ contribution that originates from eq. (12). This contribution has a rather trivial origin and it should not be associated with any many-body correlations

\[ F(r; t) = \int dq \int dq' F(q; t) F(q'; t) \theta(r - q) \theta(r - q') \tag{15} \]

A similar conclusion was reached in ref. [7].

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there is a similar simple “baseline” contribution for other geometries discussed in ref. [5].

Finally, in fig. 4(b) we show the length dependence of the relaxation time $\tau$ defined through the single-particle overlap, $Q_{\text{self}}(\tau) = e^{-1}$. We find that our mode-coupling approach overestimates the influence of the pinning on the relaxation time: the values of $\ln(\tau/\tau_{\infty})$, where $\tau_{\infty}$ is the relaxation time of the $l = \infty$ (i.e., unpinned) system, are consistently above those obtained from computer simulations (see fig. 4(b) of ref. [5]).

**Summary.** – We proposed an alternative mode-coupling approach for partially pinned systems. Our approach agrees quantitatively with the mean-field analysis of the p-spin model and with the results obtained from the RFOT theory. We showed that the contribution to the long-time limit of the collective overlap originating from the presence of pinning is dominated by the trivial small-$x$ limit. This small-$x$ contribution is not related to any growing point-to-set correlations.

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