Mode-coupling theory predictions for the dynamical transitions of partly pinned fluid systems

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The predictions of the mode-coupling theory (MCT) for the dynamical arrest scenarios in a partly pinned (PP) fluid system are reported. The corresponding dynamical phase diagram is found to be very similar to that of a related quenched-annealed (QA) system. The only significant qualitative difference lies in the shape of the diffusion-localization lines at high matrix densities, with a reentry phenomenon for the PP system but not for the QA model, in full agreement with recent computer simulation results. This finding clearly lends support to the predictive power of the MCT for fluid-matrix systems. In addition, the predictions of the MCT are shown to be in stark contrast with those of the random first-order transition theory. The PP systems are thus confirmed as very promising models for differentiating tests of theories of the glass transition.

A partly pinned (PP) fluid system is a model of a fluid in contact with a disordered substrate which is obtained by instantaneously arresting the motion of a fraction of the particles in an equilibrium bulk system and letting the remaining mobile fraction evolve under the influence of the static random environment generated by the pinned particles. The pinning process can be homogeneous, if, for instance, the pinned particles are randomly chosen in the whole volume of the bulk system, or heterogeneous, if the arrested particles are those located in a predefined region of space. In the first case, one gets a model of a fluid adsorbed in a statistically homogeneous disordered porous solid; in the second one, a model of a fluid confined by amorphous rough walls.

Because of their peculiar preparation process from an equilibrium bulk fluid in which the future mobile and pinned components influence each other, these systems display a number of interesting features. For instance, one can show that the realization-averaged configurational properties of a PP system exactly match those of the bulk fluid from which it is prepared [1,2]. Stated otherwise, the pinning step does not alter the average configurational properties of the system. This obviously represents a major simplification when studying these quenched-disordered models, which, over the years, have been considered in various fields of liquid-state theory.

In the theory of adsorption in disordered porous solids, they appear as special cases of fluids adsorbed in depleted or templated matrices. Indeed, the first class corresponds to models of porous solids obtained by freezing equilibrium configurations of a fluid and removing at random a fraction of its particles [3,4], the second one to models obtained by freezing equilibrium configurations of a binary mixture and removing one component called the template [5–8]. Thus, it is clear that a homogeneous PP system corresponds to a depleted or templated fluid-matrix system in which the depleted or template particles are reinjected as the confined fluid [2]. Both the depleted and templated models are extensions of the concept of the quenched-annealed (QA) system, first introduced by Madden and Glandt [9], in which the porous matrix is obtained by simply quenching equilibrium configurations of a fluid without any subsequent particle removal. The properties of these different models have been compared by Van Tassel *et al.* [3–8]. An interesting finding that will be useful in the

following is that, for a given matrix density, the depleted and templated matrices generically show stronger correlations and a more open accessible volume than a quenched matrix obtained by simply freezing equilibrium configurations of a fluid [3–5].

In computational studies of the dynamics of fluids in confinement have also regularly appeared PP systems [1, 10–21]. Here, the use of these models can turn out highly advantageous. Indeed, the configurations of the fluid particles obtained immediately after the pinning step are automatically equilibrium configurations by construction [1,2]. Hence, an acceptable initial configuration is always readily available, while finding one can be a very nontrivial task for other models (see Ref. [22] for the case of QA systems), and it does not have to be equilibrated [1,12]. This is clearly very interesting for studies of dense and/or glass-forming systems.

Following insightful pioneering works where PP systems had been investigated in order to measure the spatial extent of dynamical correlations in confined glass-forming liquids [1,12–14], it was realized that these systems could also be used to probe the existence of nontrivial static correlations in bulk glassy liquids. The key here is to consider the pinning process as a constraint imposed on the fluid and to measure how the configurations of the constrained particles influence the accessible states of the free ones, via the computation of point-to-set correlations [23–27], for instance. Numerous studies of PP systems have recently appeared along this line [28–36].

In this Rapid Communication, we contribute to the study of the glassy dynamics in PP systems and report on the predictions of the mode-coupling theory (MCT) [37] for the dynamical arrest scenarios in the homogeneous case. To this end, we use a recent extension of the MCT to fluids imbibed in disordered porous solids [38–41] and compute the dynamical phase diagram of the pinned system deriving from the one-component hard-sphere fluid, which, thanks to the overall mathematical structure of the theory and the robustness of the associated bifurcation schemes [37], can be expected to be a representative example.

The motivation for this work is twofold. First, computer simulation studies of the slow dynamics of fluids confined in disordered porous matrices have recently appeared [19–22,42,

43], allowing comparisons with the previous predictions of the MCT [38–41]. In many respects, they demonstrate that the MCT offers a consistent, though idealized, picture of the dynamics of these systems. For instance, the observed changes in the density fluctuation relaxation pattern with increasing confinement agree well with the theoretical picture of a crossover from type B (bulk-like) bifurcation scenarios at low matrix densities to type A (Lorentz-gas-like) scenarios at high matrix densities. However, a potential issue with the theory is also pointed out. Indeed, while the MCT predicts a dynamical reentry phenomenon at high matrix densities for the equisized hard-sphere QA system, no sign of it is actually visible in the simulation data [19–22,42,43]. But, interestingly, Kim *et al.* do clearly find such a reentry phenomenon in their results for the PP variant of this system [19–21]. One aim of the present work is to show, after some clarifications, that, at least at the level of the self-dynamics, the MCT is in complete agreement with the results of the computer simulations with respect to this reentry phenomenon at high matrix densities, even with the seemingly negative result obtained for the hard-sphere QA system. Difficulties however remain for the collective dynamics.

Second, the effect of pinned particles on the glass transition has recently been investigated in the framework of the random first-order transition (RFOT) theory [44,45] by Cammarota and Biroli, who find a very interesting and nontrivial scenario [34]. It is widely believed that the RFOT theory and the MCT have strong connections, because they have some mathematical structures in common. However, actual calculations sometimes reveal major discrepancies, like when the spatial dimension is changed [46–48]. It thus seems very natural to wonder how the two theories compare in the present context. As we will show, it turns out that the scenario predicted by the MCT differs very significantly from the one obtained by Cammarota and Biroli.

The application of the MCT scheme to fluid-matrix systems results in self-consistent equations for the time evolution of $\phi_q(t)$, the normalized connected autocorrelation function of the fluid density fluctuations, and $\phi_q^s(t)$, the autocorrelation function of the tagged-particle density fluctuations, at wave vector modulus q [40,41]. In the infinite time limit, from which the state of the system can be determined, they reduce to equations for the nonergodicity parameters $f_q = \lim_{t\to\infty} \phi_q(t)$ and $f_q^s = \lim_{t\to\infty} \phi_q^s(t)$, which read

$$\frac{f_q}{1-f_q} = \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left[V_{\mathbf{q,k}}^{(2)} f_k f_{|\mathbf{q-k}|} + V_{\mathbf{q,k}}^{(1)} f_k \right], \quad (1)$$

$$\frac{f_q^s}{1-f_q^s} = \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left[v_{\mathbf{q,k}}^{(2)} f_k^s f_{|\mathbf{q-k}|} + v_{\mathbf{q,k}}^{(1)} f_k^s \right], \quad (2)$$

$$V_{\mathbf{q,k}}^{(2)} = \frac{1}{2} n_f S_q^c \left[\frac{\mathbf{q \cdot k}}{q^2} \hat{c}_k^c + \frac{\mathbf{q \cdot (q-k)}}{q^2} \hat{c}_{|\mathbf{q-k}|}^c \right]^2 S_k^c S_{|\mathbf{q-k}|}^c, \quad (3)$$

$$V_{\mathbf{q,k}}^{(1)} = n_f S_q^c \left[\frac{\mathbf{q \cdot k}}{q^2} \hat{c}_k^c + \frac{\mathbf{q \cdot (q-k)}}{q^2} \frac{1}{n_f} \right]^2 S_k^c S_{|\mathbf{q-k}|}^d, \quad (4)$$

$$v_{\mathbf{q,k}}^{(2)} = n_f \left[\frac{\mathbf{q \cdot (q-k)}}{q^2} \right]^2 \left[\hat{c}_{|\mathbf{q-k}|}^c \right]^2 S_{|\mathbf{q-k}|}^c, \quad (5)$$

$$v_{\mathbf{q,k}}^{(1)} = \left[\frac{\mathbf{q \cdot (q-k)}}{q^2} \right]^2 \hat{h}_{|\mathbf{q-k}|}^d, \quad (6)$$

where n_f is the fluid density, S_q^c the connected structure factor, S_q^d the disconnected structure factor, $\hat{h}_q^d$ the Fourier transform of the disconnected total pair correlation function, and $\hat{c}_q^c$ the Fourier transform of the connected direct correlation function defined as $n_f \hat{c}_q^c = 1 - 1/S_q^c$ [49–52]. All these quantities depend on both the fluid and matrix densities.

While Refs. [38–41] explicitly mention QA systems, no assumption about the statistics of the disordered solid is actually made during the derivation of the MCT. It follows that it can be applied to any fluid-matrix model, and in particular to homogeneous PP systems. Such a possibility has sometimes been challenged [1,16]. Indeed, as already mentioned, the pinning protocol exactly preserves the configurational properties of the bulk fluid from which a PP system is prepared [1,2], but it can have a strong effect on the dynamics [1,10–21]. At first sight, this looks incompatible with the use of the MCT, which is known to take only structural quantities as input in order to predict the dynamics. The present worked example should clearly show that no such difficulty actually exists, the key being that, for fluid-matrix systems, one has to properly take into account the fact that the correlation functions generically split into connected and disconnected components in the presence of quenched disorder, due to broken symmetries at the microscopic level [53]. In other words, even if this is not visible at the level of simple configurational properties, the structure does actually change in a pinned system (see the Supplementary Material of Ref. [35] for an illustration) and this is reflected in an explicit dependence of the connected and disconnected correlation functions on the pinning fraction. Similar care should be taken when discussing the thermodynamics of PP systems, since the thermodynamic susceptibilities such as the heat capacity and the compressibility are given by connected correlations in disordered systems [52,54].

In addition to the results for the equisized hard-sphere PP system, those for the analogous QA system, which already appeared in Refs. [38–41], will be recalled for comparison. The difference between the two models is subtle. In both cases, the fluid and the matrix consist of hard-sphere particles of the same diameter and the control parameters are the volume fractions occupied by the fluid and matrix particles, denoted by ϕ_f and ϕ_m, respectively. Only the preparation protocol is slightly modified. In the QA system, the configurations of the matrix particles are drawn from those of an equilibrated hard-sphere fluid of compacity ϕ_m, then a fluid of mobile particles with compacity ϕ_f is inserted in the obtained disordered samples. In the PP system, a hard-sphere fluid of compacity $\phi_f + \phi_m$ is first equilibrated, then a fraction $x = \phi_m/(\phi_f + \phi_m)$ of the particles is pinned down to form the porous matrix of compacity ϕ_m, while the remaining ones become the mobile fluid component with compacity ϕ_f. So, while the matrix in the QA system is prepared independently of the fluid that will be adsorbed in it, the matrix in the PP system is prepared under its direct influence. This is the reason for the generically stronger correlations and more open accessible volume of the matrix in the PP system [3–5]. Note that by construction, both models coincide when ϕ_f or ϕ_m vanish.

The required structural input is computed for both systems within the Percus-Yevick approximation [49–51,55], which
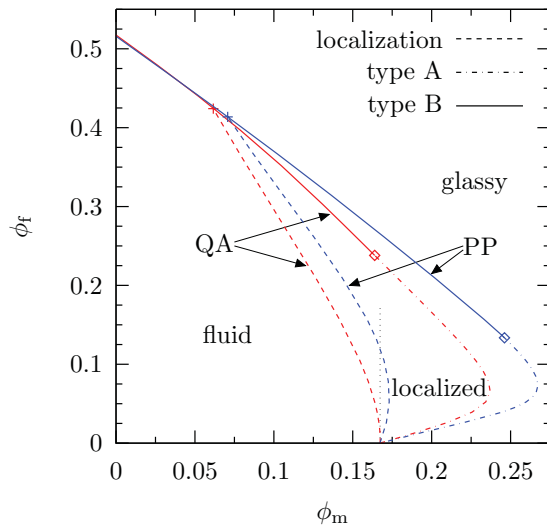
FIG. 1. (Color online) Mode-coupling dynamical phase diagrams of the equisized hard-sphere quenched-annealed (QA) and partly pinned (PP) fluid-matrix models. ϕ_f and ϕ_m denote the volume fractions occupied by the fluid and matrix particles, respectively. The dotted vertical line is a guide to emphasize the reentrant behavior of the diffusion-localization line of the PP system.

leads to analytic expressions for the hard-sphere PP system, as was recently realized [2]. The numerical procedure used to solve the above equations is described in Refs. [40,41], to which the interested reader is referred for technical details, together with Ref. [56] where an in-depth study of the difficulties arising from the presence of an infrared divergence in the MCT equations is reported.

The resulting phase diagrams are plotted in Fig. 1. Both have much in common. Three phases are found: fluid when $f_q = f_q^s = 0$ is the only solution, localized when $f_q = 0$ and $f_q^s \neq 0$, and glassy when $f_q \neq 0$ and $f_q^s \neq 0$. The systems enter the glassy phase by crossing an ideal glass transition line, which corresponds to the freezing of the collective dynamics and along which the bifurcation scenario changes from type B to type A as the confinement gets stronger. Furthermore, this line is preceded at high enough matrix densities by a diffusion-localization transition line associated with a continuous arrest of the self-dynamics only. For the PP system, there might be a very short glass-glass transition line at the junction between the type A and B glass transition branches, but given its very limited extent and the possible sources of inaccuracies in the numerical calculations, such as the discretization of the integrals in Eqs. (1) and (2), no definite conclusion can be reached at this point. In practice, both systems can be said to have phase diagrams with the same topology.

Now considering the shape of the phase diagrams, another shared feature of both models is a significantly reentrant ideal glass transition line, the effect being more pronounced for the PP system. But, and quite remarkably, the MCT delivers contrasting predictions with respect to the behavior of the diffusion-localization transition lines. Indeed, no reentry phenomenon is visible in Fig. 1 for the QA system, while one is clearly present for the PP system [57]. For both types of transition, the overall trend, which is that the ergodicity-breaking events occur at higher densities in the PP system, is perfectly in line with the fact that it is characterized by a more open accessible volume [3–5], thus confirming an argument by Kim *et al.* [19–21].

The diffusion-localization transition lines are precisely those investigated by Kim *et al.* in their simulations [19–21]. Indeed, the criterion they use to define the dynamically arrested phase is based on the mean-squared displacement, a self dynamical quantity. This is confirmed by the careful analysis of the interplay between the collective and self dynamics led by Kurzidim *et al.* [22,43]. Hence, the conclusion: The MCT does correctly capture the fact that the subtle structural differences between the QA and PP models result in different reentry behaviors of their *self* dynamics in the high matrix density regime, in full agreement with the simulation results [19–21].

Unfortunately, the situation is less clear with respect to the *collective* dynamics, which has been studied in detail for the QA system only [22,42]. Indeed, in qualitative agreement with the prediction of distinct transition lines for the self and collective dynamics at moderate and high matrix densities, a wide separation of time scales between the two dynamics is observed in simulations, but with no sign of a reentrant collective dynamics. The problem might lie in the theory or in the simulations. Indeed, on the one hand, it is possible that the standard formulation of the MCT based on equilibrium quantities breaks down if the particles cannot redistribute themselves across the system, as is precisely the case in a localized state. On the other hand, the simulations, which are very difficult in this density regime, do not seem consistent with the expectation that the collective and self dynamics coincide in the limit of vanishing fluid density. So, more work is definitely needed to clarify this point. Here, the PP systems could be very useful, thanks to the possibility to easily generate equilibrated samples even at high densities.

We now turn to the comparison with the predictions of the RFOT theory [34]. For this, it is more convenient to consider the dynamical phase diagram of the PP system in the plane defined by the total volume fraction $\phi_f + \phi_m$ and the pinning fraction x, as in Fig. 2. In passing, we note that, thanks to this representation, the total densities at the various transitions are clearly shown to be decreasing functions of x, in line with the rather natural expectation that pinning down particles slows down the dynamics.

Within the RFOT theory, the PP systems are found to occupy a very special place [34]. Indeed, at different levels of calculation (mean-field or real-space renormalization group analysis), the theory consistently predicts for them ideal glass transition lines starting at $x = 0$ (bulk limit) and ending at some critical value $x_c < 1$. This means that one can go from the fluid to the glassy phases without crossing any transition, simply by moving around this terminal point as one can do with the critical point of the liquid-gas transition. However, this is an isolated case. For other types of random environments, including QA systems, it is predicted that the glass transition never disappears, but only becomes continuous for strong disorder, very much like in the MCT scenario with the type A bifurcations. It is then impossible to reach the glassy phase without crossing a transition.

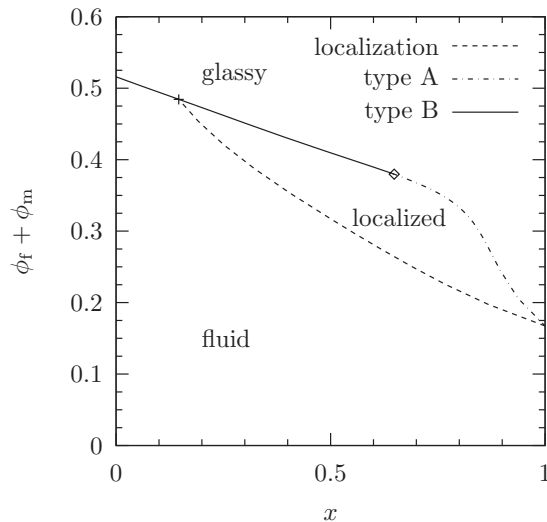So, while the predictions of the MCT and the RFOT theory tend to agree for generic cases of fluids in disordered

FIG. 2. Mode-coupling dynamical phase diagram of the equisized hard-sphere partly pinned fluid-matrix model. $\phi_f + \phi_m$ is the total volume fraction occupied by both the fluid and matrix particles; $x = \phi_m/(\phi_f + \phi_m)$ is the pinning fraction.

environments, they are in stark contrast for the PP systems. Indeed, within the MCT, the PP systems appear as rather ordinary fluid-matrix systems, as illustrated by Fig. 1, and the transition lines run up to $x = 1$, so that the system always has to cross a dynamical transition line in order to enter the glassy phase, as shown in Fig. 2. The PP systems are thus a clear example of diverging predictions between the two considered theoretical schemes and, as such, would probably deserve much attention in the future. We note that, based on the presently available data and, in particular, on the joint study of QA and PP systems by Kim *et al.* [19–21], no obvious inconsistency with the MCT scenario has appeared yet, but, as pointed out above, some issues are still pending and more work is needed.

In summary, in this Rapid Communication, the predictions of the MCT for the slow dynamics of a homogeneous PP system have been worked out and compared to previous findings for a related QA system [38–41], to the results of recent computer simulation studies [19–21], and to the scenario derived within the framework of the RFOT theory [34]. It is found that the dynamical phase diagrams of the QA and PP systems are very similar within MCT. The only significant qualitative difference is in the shape of the diffusion-localization lines at high matrix densities, with a reentry phenomenon for the PP system but not for the QA model. In the light of these results, the simulation data, which could first appear as a challenge to the theory, are actually found to represent a nice confirmation of its predictions. Finally, the comparison between the MCT and the RFOT theory shows that the two approaches make predictions that are in stark contrast.

The latter result might have serious consequences. Indeed, two different points of view might be adopted on PP systems. On the one hand, seen from the theory of simple liquids and adsorption phenomena, they are fluid-matrix models among others [3–8]. They display interesting and possibly useful configurational properties [1,2], but these are merely accidental and no peculiar physics beyond confinement phenomena should be expected. This is what the MCT supports. On the other hand, seen from the theory of disordered systems, they are very special physical objects, ideally and uniquely suited to track hidden forms of order and the related phase transitions in amorphous systems [23–36]. This is what the RFOT theory illustrates. Finding which point of view is the most appropriate for glass-forming liquids could have a profound influence on future directions in glass transition theory.

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