

Critical Dynamical Heterogeneities Close to Continuous Second-Order Glass Transitions

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We analyze, using inhomogeneous mode-coupling theory, the critical scaling behavior of the dynamical susceptibility at a distance ϵ from continuous second-order glass transitions. We find that the dynamical correlation length ξ behaves generically as $\epsilon^{-1/3}$ and that the upper critical dimension is equal to six. More surprisingly, we find that ξ grows with time as $\ln^2 t$ exactly at criticality. All of these results suggest a deep analogy between the glassy behavior of attractive colloids or randomly pinned supercooled liquids and that of the random field Ising model.

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Several recent studies have revealed that the properties of the glass transition can be drastically modified by suitably tuning some control parameters. In the case of the colloidal glass transition, an attractive interaction on top of the hard-sphere repulsion can change the dynamical behavior and lead to glass-glass transitions and logarithmic relaxation [1]. This behavior is also expected for glassy liquids in porous media [2]. For generic glass-forming liquids, it was recently predicted [3,4] that randomly pinning a fraction of particles would transmute the glass transition in a continuous second-order phase transition akin to that of the random field Ising model (RFIM) [3–5] (for a review of the RFIM and its critical properties, see Refs. [6,7]). There are strong theoretical indications that these phenomena are in fact all related to the existence of a new kind of glassy critical point, first found within mode-coupling theory (MCT) as a higher-order singularity [8]. The physical contexts in which it appears are quite different: for attractive colloids it is a terminal point of a glass-glass transition line, for glass-forming liquids either pinned or trapped in porous media it corresponds to the locus where the mode-coupling transition and the ideal glass transition lines merge. In the former case the glass transition line stops at this new critical point [1,3], whereas in the latter it carries on and becomes continuous [2]. In all of these physical situations, localized particle rearrangements not described by MCT are expected to be of greatly diminished importance. In particular, for attractive colloids such processes are suppressed by the formation of strong short-ranged bonds that push the system into an attractive glass phase [8], while in the case of pinned systems the temperature at which an entropy

crisis occurs is expected to merge with the critical temperature of MCT [3], which implies that the region where MCT fails to describe glassy dynamics shrinks to zero. Hence, MCT might become *quantitatively* accurate in these situations. The dynamical behavior of the two-point functions at this new glassy critical point, that we will call A_3 using Götze's terminology, was predicted by MCT computations [8] and confirmed later both numerically and experimentally in colloids [1]. The static properties of the fluctuations of the overlap field between two equilibrium configurations were recently investigated in Refs. [3,5] and, using simulations, in Ref. [9]. A complete theory of dynamical correlations is, however, still lacking. The aim of this Letter is to develop such a theory by extending the “inhomogeneous” MCT (IMCT) formalism, which was developed by some of us [10] to describe dynamical heterogeneities at the usual MCT transition. We shall obtain the mean-field values of the critical exponents, the upper critical dimension, and derive the critical behavior, which turns out to be very different from the usual one. We find, in particular, that dynamical length scales grow logarithmically with time, which strongly bolsters the relationship with the RFIM [3,5].

In order to grasp the main properties of the A_3 critical point, it is useful to focus on the mean-field Landau-like potential $V(f; \epsilon)$, called the Franz-Parisi (FP) potential in the present context [11]. The arguments ϵ and f are, respectively, the vector of all control parameters that can be tuned (e.g., the temperature, the range of attraction between particles, and the fraction of pinned particles) and the glassy (nonergodic) order parameter, which measures

how far the dynamics can displace the system away from its initial configuration. For usual glass transitions, the FP potential has a unique minimum $f_0 = 0$ at high temperatures; it corresponds to a complete loss of memory of the initial condition as normal in a liquid. A secondary minimum appears for $f = f^* > 0$ below a certain transition temperature T_c , see Fig. 1. An important achievement of the last decades was to establish that T_c actually coincides with the MCT transition, where locally stable, long-lived amorphous structures, corresponding to the secondary minimum of $V(f; \epsilon)$, appear. Roughly speaking, the connection between the FP potential and MCT may be expressed as $V'(f; \epsilon) = (f/1-f) - \mathcal{F}_\epsilon[f]$, where $\mathcal{F}_\epsilon[f]$ is the memory function of the MCT equations [12]. (The full-fledged MCT calculation deals with wave-vector dependent order parameters $f_{\vec{q}}$, we will return to this below.) An A3 critical point corresponds to the merging of the minima f_0 and f^* [13,14]. It is of codimension 2, much as the liquid-gas critical point; i.e., one needs to tune at least two control parameters to reach it, as found for systems with quenched pinning sites and for hard sphere systems, where one tunes the short-range attractive interactions and the density, see inset of Fig. 1.

Technically, the existence of an underlying thermodynamical formulation has been extremely useful to understand that the MCT transition is necessarily accompanied by the divergence of a length scale, which governs the spatial extent over which dynamical fluctuations are correlated, a feature that was hard to anticipate within the original framework of Götze *et al.* This diverging length scale is in fact a direct consequence of the vanishing of the curvature $V''(f^*; \epsilon)$ of the FP potential at T_c (see Fig. 1) [15–17]. The IMCT formalism allowed us to make a series of precise predictions about the space-time scaling of dynamical heterogeneities in supercooled liquids close to T_c [10]. One finds in particular that the dynamical correlation length diverges as $|T - T_c|^{-1/4}$ as the critical

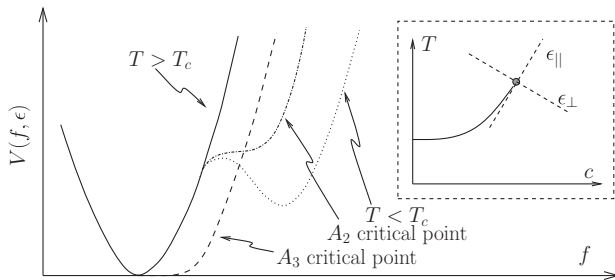


FIG. 1. Schematic presentation of the FP potential. The second minimum that develops below the transition point becomes flat as the usual MCT (A_2) transition point is approached from below. The two minima coalesce into one at the A_3 point. Inset: Schematic phase diagram obtained for random pinning glass transitions. In the temperature (T)-pinning fraction (c) plane the MCT transition line ends in an A_3 critical point [3]. The two special directions mentioned in the text are shown.

point is approached. Although the IMCT predictions are only expected to be correct far enough from T_c below 8 dimensions, many general predictions appear to be confirmed, sometimes quantitatively, by large scale computer simulations, see Ref. [18] and references therein. These developments, and others, strongly support a quantitative theory of supercooled liquids built using the mean-field scenario as a starting point, much as Curie-Weiss theory provides a foundation for the modern theory of critical phenomena [19,20].

In the following we first explain our results in an informal and simple way based on the behavior of the FP potential; we then sketch the complete IMCT derivation. We will denote as $V^{(n)}$ the n th derivative of $V(f; \epsilon)$ with respect to f ($V' = V^{(1)}$, etc.), and $\vec{\nabla}_\epsilon V$ the gradient of V with respect to the parameters. The expansion of $V'(f; \epsilon)$ around the transition point $f = f_c$, $\vec{\epsilon} = 0$ reads, with $\delta f = f - f_c$:

$$V'(f; \epsilon) \approx V_c^{(2)} \delta f + \frac{1}{2} V_c^{(3)} \delta f^2 + \frac{1}{3} V_c^{(4)} \delta f^3 + \vec{\nabla}_\epsilon V_c' \cdot \vec{\epsilon} + \vec{\nabla}_\epsilon V_c^{(2)} \cdot \vec{\epsilon} \delta f + \dots,$$

where we have used that by definition, at the transition, $V'(f_c; 0) \equiv 0$. The standard MCT transition (the A_2 critical point) occurs when the secondary minimum of $V(f)$ just appears, implying $V_c^{(2)} = 0$ (see Fig. 1). The next order singularity (A_3) occurs when $V_c^{(3)}$ concomitantly vanishes as well (see Fig. 1). Looking for the new location f^* of the minimum away from the transition, one finds, to leading order in $\epsilon = |\vec{\epsilon}|$: $f^* - f_c \sim \sqrt{\epsilon}$, as is familiar for the A_2 case and $f^* - f_c \sim \sqrt[3]{\epsilon}$ in the generic A_3 case. There is, however, a subtlety here: since the A_3 point requires at least two parameters to be varied simultaneously, one needs to include the case where the chosen trajectory in parameter space is precisely perpendicular to $\vec{\nabla}_\epsilon V_c'$, in which case one finds again the weaker singularity $f^* - f_c \sim \sqrt{\epsilon_\parallel}$, where ϵ_\parallel is the distance to the critical point along that special direction. The motivation for the notation ϵ_\parallel stems from the liquid-glass phase diagram, sketched in the inset of Fig. 1 in the case of random pinning glass transitions [3]. In general, A_3 is the terminal point of the line of A_2 critical points. The special direction found above is the one tangent to the A_2 line. In consequence, we also introduce the notation ϵ_\perp for the magnitude of the component of $\vec{\epsilon}$ perpendicular to the A_2 line (i.e., parallel to $\vec{\nabla}_\epsilon V_c'$).

The main idea of IMCT [10] is to perturb the system with a small spatially periodic external potential $\propto \cos(\vec{q}_0 \cdot \vec{x})$, whose spatial profile varies over the length scale $1/q_0$. The characteristic value of q_0^* at which the external perturbation starts to act differently from a uniform, $q_0 = 0$, perturbation allows one to obtain the correlation length of dynamical heterogeneities as $\xi = 1/q_0^*$. Since all the physics of the

slowing down is governed by the vanishing of the curvature of the FP potential, the crucial point is to work out how the periodic perturbation (of zero mean) changes this curvature. Because the system is rotationally invariant, it is reasonable to assume that the extra contribution to the curvature is $\sim q_0^2$. Therefore, one has

$$V''(f^*, \varepsilon) \approx V_c^{(3)}(f^* - f_c) + \frac{1}{2} V_c^{(4)}(f^* - f_c)^2 + \vec{\nabla}_\varepsilon V_c^{(2)} \cdot \vec{\varepsilon} + \Gamma q_0^2.$$

Close to an A2 critical point, $V_c^{(3)} \neq 0$ and $f^* - f_c \sim \sqrt{\varepsilon}$, which shows that the characteristic value of q_0 beyond which the relaxation time substantially changes is $\sim \varepsilon^{1/4}$, leading to $\xi \sim \varepsilon^{-1/4}$, in agreement with the result of Ref. [10]. Upon approaching an A3 critical point, $V_c^{(3)} = 0$, leading to $\xi \sim \varepsilon_\perp^{-1/3}$ in the generic case, and to $\xi \sim \varepsilon_\parallel^{-1/2}$ in the special case where $\varepsilon_\perp = 0$. These results are fully confirmed, and made more precise, by the IMCT analysis that we now briefly present.

The IMCT formalism starts from an exact equation for the inhomogeneous dynamic structure factor $F(\mathbf{q}_1, \mathbf{q}_2, t) = \langle \rho_{\mathbf{q}_1}(t) \rho_{-\mathbf{q}_2}(0) \rangle$ in the presence of an inhomogeneous external field $u(\mathbf{q}_0)$:

$$\frac{\partial F(\mathbf{q}_1, \mathbf{q}_2, t)}{\partial t} + \Omega_{q_1} F(\mathbf{q}_1, \mathbf{q}_2, t) + \sum_{\mathbf{k}} \int_0^t M(\mathbf{q}_1, \mathbf{k}, t - t') \frac{\partial F(\mathbf{k}, \mathbf{q}_2, t')}{\partial t'} dt' = \mathcal{T}_u(\mathbf{q}_1, \mathbf{q}_2, t), \quad (1)$$

where $\Omega_{q_1} \equiv q_1^2 k_B T / S_{q_1}$ is a frequency term, $M(\mathbf{q}_1, \mathbf{k}, t)$ is the memory kernel and $\mathcal{T}_u(\mathbf{q}_1, \mathbf{q}_2, t)$ contains all the terms generated due to the external potential. The dynamical susceptibility is defined as $\chi_{q_0}(\mathbf{q}_1, t) = \delta F(\mathbf{q}_1, \mathbf{q}_1 + \mathbf{q}_0, t) / \delta u(\mathbf{q}_0)|_{u \rightarrow 0}$. This object obeys a linear equation (that we do not write here, see Ref. [10]) obtained by taking the derivative of Eq. (1) with respect to $u(\mathbf{q}_0)$. In the long-time limit, this linear equation reads

$$\sum_{\mathbf{k}} (\delta_{\mathbf{k}, \mathbf{q}} - C_{\mathbf{q}; \mathbf{k}}(\mathbf{q}_0)) \chi_{q_0}(\mathbf{k}, t \rightarrow \infty) = S(\mathbf{q}, \mathbf{q}_0), \quad (2)$$

where $S(\mathbf{q}, \mathbf{q}_0)$ is a nonsingular source term and $C_{\mathbf{q}; \mathbf{k}}(\mathbf{q}_0)$ is a \mathbf{q}_0 dependent matrix that can be fully computed in terms of the memory kernel of the model, see Ref. [10]. Note that $\chi_{q_0}(\mathbf{k}, t \rightarrow \infty)$ is nothing else than the variation of the nonergodic parameter $f_{\mathbf{k}} = F(\mathbf{k}, t \rightarrow \infty)$ due to the external potential. In order to analyze Eq. (2) we recall (see Ref. [13]) that $C_{\mathbf{q}; \mathbf{k}}(0) = (1 - f_{\mathbf{k}})^2 \partial \mathcal{F}_{\mathbf{q}, \varepsilon}[\{f_{\mathbf{k}}\}] / \partial f_{\mathbf{k}}$. The properties of the operator $\delta_{\mathbf{k}, \mathbf{q}} - C_{\mathbf{q}; \mathbf{k}}(0)$, which is akin to $V'(f; \varepsilon)$, are reported in Ref. [13]: at distance ε from the transition, one finds $f_{\mathbf{k}} = f_{\mathbf{k}}^c + (1 - f_{\mathbf{k}}^c)^2 g_{\mathbf{k}}$, where $g_{\mathbf{k}} \propto \sqrt[3]{\varepsilon} \psi_{\mathbf{k}}^R$ and $\psi_{\mathbf{k}}^R$ is the (right) zero mode of $\mathbb{1} - \hat{C}(0)$ evaluated at the transition point. This scaling holds when approaching

the A3 critical point in any direction other than the one parallel to the line of usual A2 transitions that approach the A3 point. In this case one finds $g_{\mathbf{k}} \propto \sqrt{\varepsilon} \psi_{\mathbf{k}}^R$. Away from criticality, the smallest eigenvalue of the matrix $\mathbb{1} - \hat{C}(0)$ is not exactly zero: the deviations are of order, respectively, $\varepsilon_\perp^{2/3}$ and ε_\parallel close to an A3 point, depending on the direction of approach to criticality. Coming back to our original problem, we remark that the eigenvalues of $C_{\mathbf{q}; \mathbf{k}}(\mathbf{q}_0)$ can be computed using perturbation theory. Because of rotational invariance, one finds that all eigenvalues of $C_{\mathbf{q}; \mathbf{k}}(0)$ are shifted by an amount $\propto q_0^2$. The smallest eigenvalue of $\mathbb{1} - \hat{C}(\mathbf{q}_0)$ is therefore equal to $\alpha |\varepsilon_\perp|^{2/3} + \Gamma q_0^2$, where α and Γ are numbers, and $\varepsilon_\perp \neq 0$. The solution of Eq. (2) will be dominated by this very small eigenvalue, and thus reads

$$\chi_{q_0}(\mathbf{k}, t \rightarrow \infty) \approx \frac{\langle \psi^L | S \rangle \psi_{\mathbf{k}}^R}{\alpha |\varepsilon_\perp|^{2/3} + \Gamma q_0^2}, \quad (3)$$

where $\psi^{L,R}$ are the left and right largest eigenvectors of $C_{\mathbf{q}; \mathbf{k}}$ at criticality. From this expression, one directly demonstrates the existence of a diverging susceptibility and a diverging length scale within MCT, which is intimately due to the vanishing of the curvature of the FP potential. Close to an A3 point, this length scale diverges as $|\varepsilon_\perp|^{-1/3}$, as announced above. The time dependent analysis is more cumbersome and will be presented in detail elsewhere [21]. The final result is

$$\chi_{q_0}(\mathbf{k}, t) \simeq \frac{V_\perp(k) \xi_\perp^2}{1 + \Gamma(q_0 \xi_\perp)^2} \mathcal{G}_\perp \left(\frac{\ln t}{\sqrt{\xi_\perp}}, q_0 \xi_\perp \right), \quad \varepsilon_\perp \neq 0,$$

$$\chi_{q_0}(\mathbf{k}, t) \simeq \frac{V_\parallel(k) \xi_\parallel^2}{1 + \Gamma(q_0 \xi_\parallel)^2} \mathcal{G}_\parallel \left(\frac{\ln t}{\sqrt{\xi_\parallel}}, q_0 \xi_\parallel \right), \quad \varepsilon_\perp = 0,$$

where $V_{\parallel, \perp}(k)$ are certain functions, and $\xi_\perp = |\varepsilon_\perp|^{-1/3}$, $\xi_\parallel = |\varepsilon_\parallel|^{-1/2}$, as indeed anticipated by the simple arguments above.

By usual scaling arguments one finds that the $q_0 \rightarrow 0$ limit is well behaved provided $\mathcal{G}_{\parallel, \perp}(u, v = 0) \sim u^4$, and therefore, at criticality, $\chi_{q_0}^c(\mathbf{k}, t) \sim \ln^4 t$, a result that is compatible with the simulation results of attractive colloids reported in Ref. [22]. Away from criticality, the $\ln^4 t$ behavior only persists up to a time τ_ξ such that $\ln \tau_\xi \sim \sqrt{\xi_\parallel}$ (or $\ln \tau_\xi \sim \sqrt{\xi_\perp}$ along the special line $\varepsilon_\parallel = 0$). Another interesting limit is when the system is critical $\varepsilon_{\parallel, \perp} = 0$ and perturbed at a nonzero spatial frequency, $q_0 \neq 0$. In order to retain a nontrivial dynamics, one must now have $\mathcal{G}_{\parallel, \perp}(u \rightarrow 0, v \rightarrow \infty) = g(u^2 v)$, where $g(x)$ is a certain function behaving as x^2 for small x , and saturating to a constant for $x \rightarrow \infty$. This leads to $\chi_{q_0}^c(\mathbf{k}, t) = q_0^{-2} g(q_0 \ln^2 t)$, which shows that at criticality, the dynamical length grows without bound, as $\xi_c(t) \sim \ln^2 t$ [23]. This should be compared with the corresponding result for the A2 point, where $\xi_c \sim t^{a/2}$, where a is the MCT exponent for the β regime. As the A3 point is approached, the value of a

tends to zero and the power law crosses over to a logarithmic behavior. Note that there is no α regime at the A3 point, contrarily to the usual phenomenology of the A2 transition. In the latter case, the q_0 dependence of χ_{q_0} for $q_0 \gg \xi^{-1}$ crosses over from q_0^{-2} in the short time, β regime, to q_0^{-4} in the long time, α regime. For the A3 transition, on the other hand, only the q_0^{-2} behavior survives. We have checked all of these results numerically by solving exactly the dynamical equation obeyed by $\chi_{q_0}(t)$ in the so-called schematic limit where all \mathbf{k} dependence is discarded. We have chosen the F_{13} model for which the memory kernel is $\mathcal{F}_\epsilon[f] = \epsilon_1 f + \epsilon_3 f^3$. The salient features of the above scaling predictions for $\chi_{q_0}(t)$ are confirmed in Fig. 2. From a purely phenomenological point of view, the most important points are as follows. (1) The growth of χ_{q_0} is a logarithmic function of the relaxation time thus implying that dynamical heterogeneities increase much slower close to an A3 critical point than close to an A2 one; this might explain the numerical data of Ref. [24]. (2) The shape of χ_{q_0} is markedly different from the one found at an ordinary MCT transition. In particular, both the maximum and the long-time limit of χ_{q_0} diverge as the transition is approached, at variance with what happens close to an A2 point, where the long-time limit of χ_{q_0} remains bounded.

All the above results should be only be valid in high enough dimensions. In order to assess the effect of critical fluctuations on mean-field results one has to focus on the 4-point density correlations often called $G_4(\mathbf{r})$, which is related (in Fourier space) to the *square* of the dynamical susceptibility χ_{q_0} defined above (see Ref. [25] for a full justification of this relation). Since χ_{q_0} behaves as q_0^{-2} at

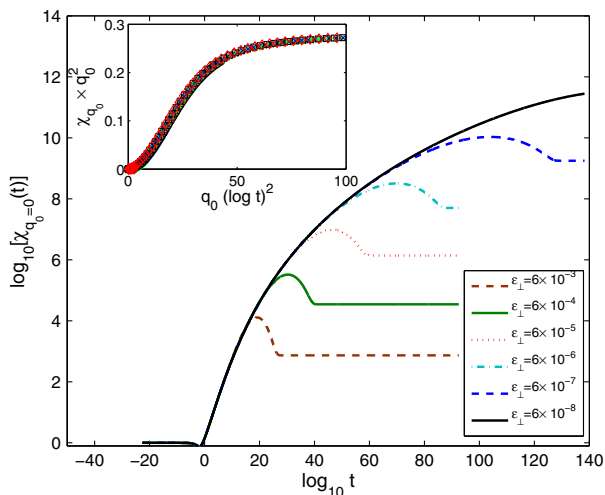


FIG. 2 (color online). Numerical results obtained by solving the schematic F_{13} equations. $\chi_{q_0=0}$ as a function of time approaching the A3 critical point, which is indeed found to rescale as $|e_{\perp}|^{-2/3}$ times a function of $|e_{\perp}|^{1/6} \times \ln t$. Inset: Scaling collapse, for different q_0 and t , of $q_0^2 \chi_{q_0}^c$ as a function of $q_0 \ln t$ at criticality, as predicted from the theory.

criticality, $G_4(r)$ is found to decay as $1/r^{d-4}$ up to distances of order ξ . This allows one to estimate the intensive fluctuations of the order parameter f^* in a region of size ξ^d , which is found to be $\sqrt{\langle \delta f^2 \rangle} \sim \xi^{-(4-d)/2} = e^{\nu(4-d)/2}$, which must be compared to $f^* - f_c \sim e^{\beta}$. Since $\beta = \nu = 1/3$ for A3 transitions (or $\beta = \nu = 1/2$ along the special direction) we conclude that the upper critical dimension d_c below which critical fluctuations change the nature of the transition is $d_c = 6$, instead of $d_c = 8$ as found for usual MCT transitions [25,26]. Therefore, in physical dimensions $d = 3$, one expects that these critical fluctuations will considerably affect the above predictions, at least close enough to the critical point. The relative influence of these fluctuations, and the quantitative size of the Ginzburg region where critical fluctuations are strong, are expected to depend on the model. But one consequence of these fluctuations that is physically relevant is the violation of the Stokes-Einstein relation, relating the relaxation time of the system to the diffusion constant of probe particles. Simulations of thermal and athermal [27,28] systems appear to conform to the prediction $d_c = 8$ for usual MCT transitions, yielding, for example, a Stokes-Einstein violation exponent that vanishes linearly as $8 - d$ [29]. Our above analysis demonstrates that near an A3 singularity the upper critical dimension of fluctuations is shifted down to a value $d_c = 6$. This result is indeed qualitatively consistent with the fact that some aspects of dynamical heterogeneity (such as bimodality of particle displacement distributions) are suppressed as one tunes, via the introduction of short-ranged attractions, a supercooled hard-sphere suspension to a regime dominated by higher-order singularities [30]. More numerical work on this and other aspects of our theory would be welcome, using simulations of attractive hard spheres in higher dimensions, along the lines of Ref. [28].

Finally, the alert reader will have recognized, both from the evolution of the FP potential shown in Fig. 1 and the value of the exponent $\beta = 1/3$ in generic directions and $\beta = 1/2$ in a special direction, that the A3 critical point is akin to an Ising transition, where ϵ_{\parallel} is a magnetic-field-like perturbation and ϵ_{\perp} is a temperaturelike perturbation. The behavior of the correlation function $G_4(r) \sim r^{4-d}$, the corresponding value $d_c = 6$ and, especially, the logarithmic relation between time and length-scale point towards the universality class of the RFIM, in line with previous static treatments [3,5]. Indeed, the A2 line is analogous to the spinodal line of the RFIM [26], which terminates at the A3 RFIM critical point. More concretely, in the physical case of random pinning sites that induce higher order singularities, the FP potential will acquire a random spatial component. Given that the standard A2 transition is like a spinodal of the FP potential, spatial fluctuations of the FP potential create an A3 second-order critical point when the minima of the FP potential merge. All of these results strengthen the analogies with the physics of the RFIM.

Remarkably, the logarithmic behavior of the correlation function and of the dynamical correlation length that we found is usually a manifestation of *activated events* which are indeed expected for the RFIM at criticality, but were thought to be impossible to grasp within a MCT formalism. On the other hand, from the study of the RFIM we know that there are two qualitatively different kinds of activated processes: the critical ones emerging at the transition and the ones responsible for nucleation. The former are correctly described by the Landau theory above $d_c = 6$ (at least at a static level), whereas the analysis of the latter requires to go beyond mean-field theory. In agreement with that, MCT appears to be able to capture activated processes at the $A3$ transition, possibly in a quantitatively correct way above $d > 6$, but fails to describe the ones that destabilize the glass phase below the MCT transition.

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