Cooperativity beyond Caging: Generalized Mode-Coupling Theory

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The validity of mode-coupling theory (MCT) is restricted by an uncontrolled factorization approximation of density correlations. The factorization can be delayed and ultimately avoided, however, by explicitly including higher order correlations. We explore this approach within a microscopically motivated schematic model. Analytic tractability allows us to discuss in great detail the impact of factorization at arbitrary order, including the limit of avoided factorization. Our results indicate a coherent picture for the capabilities as well as limitations of MCT. Moreover, including higher order correlations systematically defers the transition and ultimately restores ergodicity. Power-law divergence of the relaxation time is then replaced by continuous but exponential growth.

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The dynamic scaling in dense supercooled liquids and colloidal suspensions is a subject surrounded by controversy. Mode-coupling theory (MCT) has shaped our understanding through detailed and remarkably successful predictions [1]. A salient result of MCT is the existence of an ideal glass transition. The latter, however, is predicted to occur at substantially higher temperatures or lower densities than that observed in the laboratory [2]. Furthermore, it has been suggested that there are "activated processes" not accounted for by MCT which restore ergodicity and thus round off the ideal transition [3,4]. In order to remedy such shortcomings one should scrutinize the approximations made within MCT.

A fundamental quantity in MCT is the two-point correlator $F_k(t) = \mathcal{N}^{-1}\langle \rho_{-\boldsymbol{k}}(0)\rho_{\boldsymbol{k}}(t)\rangle$, with $\rho_{\boldsymbol{k}}$ the Fourier transform of spatial particle density fluctuations. For $\mathcal{N}$ particles interacting through a pair potential one finds, using standard projection operator techniques [1,5],

$$\ddot{F}_k(t) + \mu_k F_k(t) + (\gamma_{\boldsymbol{k}} * \dot{F}_k)(t) = 0. \qquad (1)$$

Here $(f * g)(t) = \int_0^t d\tau f(\tau) g(t-\tau)$ and dots indicate time derivatives. Further $\mu_k = k_B T k^2/mS_k$, where k_B is the Bolzmann constant, T the temperature, m the particle mass, and $S_k = F_k(0)$ the static structure factor. The memory function $\gamma_{\boldsymbol{k}}(t)$ is related to the autocorrelation of the fluctuating force. Within MCT it is assumed [1] that its dominant contribution arises from pair-density modes $\rho_{\boldsymbol{q}}\rho_{\boldsymbol{k}-\boldsymbol{q}}$. A projection of the fluctuating force onto this pair subspace gives $\gamma_{\boldsymbol{k}}(t) \approx \sum_{\boldsymbol{q},\boldsymbol{q}'} V^*_{\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q}} V_{\boldsymbol{q}',\boldsymbol{k}-\boldsymbol{q}'} \times F_{\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q},\boldsymbol{q}',\boldsymbol{k}-\boldsymbol{q}'}(t)$, where $V_{\boldsymbol{p},\boldsymbol{q}}$ represents static projections of the fluctuating force and $F_{\boldsymbol{p},\boldsymbol{q},\boldsymbol{p}',\boldsymbol{q}'}(t) = \langle \rho_{-\boldsymbol{p}}(0)\rho_{-\boldsymbol{q}}(0)\rho_{\boldsymbol{p}'}(t)\rho_{\boldsymbol{q}'}(t)\rangle$ is a four-point correlator with projected dynamics. On an *ad hoc* basis, static and dynamic correlations are then subjected to Gaussian factorization. An additional convolution approximation for the statics reduces the memory function to $\gamma_{\boldsymbol{k}}(t) \approx \int d\boldsymbol{q} w_{\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q}} F_q(t) F_{|\boldsymbol{k}-\boldsymbol{q}|}(t)$, with a weight-factor $w_{\boldsymbol{p},\boldsymbol{q}}$ containing only static information. Under these approximations for $\gamma_{\boldsymbol{k}}(t)$ Eq. (1) is closed in $F_k(t)$ and we have arrived at standard MCT.

Various versions of extended mode-coupling theories (EMCT) have been put forward to account for activated processes [4]. These theories perturbatively invoke a coupling to current modes. But recent simulations of Brownian systems [6], where the momentum current should not effectively couple to slow relaxation, have shown that deviations from MCT can be as large as they are in Newtonian systems. Also, recent theoretical work has cast grave doubt on the role of current modes in EMCT [7]. It would appear that perturbative coupling to currents cannot provide the local physics necessary to restore ergodicity deep within the activated regime.

Within a theory entirely based on density modes one is thus lead to reconsider the applicability of Gaussian factorization [8–10]. Proceeding in the projection operator approach one shows that the actual evolution of four-point correlations is governed by an equation analogous to (1) that, in turn, couples to six-point correlations, and so on. The factorization approximation may thus be delayed, a perturbative framework we refer to as generalized mode-coupling theory (GMCT). Recent implementations of GMCT [9,10] have demonstrated that the inclusion of higher order density correlations appears to systematically lower the transition temperature (or raise the transition volume fraction), suggesting that some aspect of activated behavior is captured.

In this Letter we illustrate the potential of GMCT to account for activated processes, meaning—here and throughout—the processes discarded in MCT due to factorization. This may be achieved through the *nonperturbative* limit where the full multipoint basis of dynamical correlations is included [11]. Our discussion is based on a schematic model as its analytic tractability provides deeper insights than could be obtained from numerical analysis of more realistic systems. To motivate its form we briefly return to Eq. (1) and its higher order generalizations. It is useful to normalize $\phi_k(t) = F_k(t)/S_k$. For

simplicity and following [9,10] we focus on diagonal contributions $\boldsymbol{q} = \boldsymbol{q}'$ in the memory function $\gamma_{\boldsymbol{k}}(t)$. Treating static correlations as in MCT but retaining the dynamic four-point correlations $\phi_{\boldsymbol{k}_1,\boldsymbol{k}_2}(t) = F_{\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_1,\boldsymbol{k}_2}(t)/F_{\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_1,\boldsymbol{k}_2}(0)$ leads to $\gamma_{\boldsymbol{k}}(t) \approx \int d\boldsymbol{q} \Lambda_{\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q}} \phi_{\boldsymbol{q},\boldsymbol{k}-\boldsymbol{q}}(t)$, where $\Lambda_{\boldsymbol{p},\boldsymbol{q}} = S_p S_q w_{\boldsymbol{p},\boldsymbol{q}}$ with the same weight-factor $w_{\boldsymbol{p},\boldsymbol{q}}$ as in MCT. At second-lowest order the projection operators yield [9,10]

$$\ddot{\phi}_{\boldsymbol{k}_1,\boldsymbol{k}_2}(t) + \mu_{k_1,k_2} \phi_{\boldsymbol{k}_1,\boldsymbol{k}_2}(t) + (\gamma_{\boldsymbol{k}_1,\boldsymbol{k}_2} * \dot{\phi}_{\boldsymbol{k}_1,\boldsymbol{k}_2})(t) = 0,$$

with $\mu_{k_1,k_2} \approx \mu_{k_1} + \mu_{k_2}$. The fluctuating force for pair densities is essentially a product of three density modes. We perform a corresponding projection and again consider only diagonal wave-vector terms in $\gamma_{\boldsymbol{k}_1,\boldsymbol{k}_2}(t)$. Simple treatment of the static projections as before then gives $\gamma_{\boldsymbol{k}_1,\boldsymbol{k}_2}(t) \approx [\mu_{k_1} \int d\boldsymbol{q} \Lambda_{\boldsymbol{q},\boldsymbol{k}_1-\boldsymbol{q}} \phi_{\boldsymbol{q},\boldsymbol{k}_1-\boldsymbol{q},\boldsymbol{k}_2}(t) + \mu_{k_2} \times \int d\boldsymbol{q} \Lambda_{\boldsymbol{q},\boldsymbol{k}_2-\boldsymbol{q}} \phi_{\boldsymbol{q},\boldsymbol{k}_1,\boldsymbol{k}_2-\boldsymbol{q}}(t)]/(\mu_{k_1} + \mu_{k_2})$ and $\phi_{\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3}(t)$ a normalized six-point correlation function.

This procedure may be continued indefinitely and induces a hierarchy of evolution equations for dynamical multipoint correlations. Our schematic model is obtained by dropping the wave-vector indices. We replace $\phi_k(t) \mapsto \phi_1(t)$, $\phi_{\boldsymbol{k}_1,\boldsymbol{k}_2} \mapsto \phi_2(t)$, etc., and since we effectively do not discriminate between different wave vectors, $\mu_{k_1} + \cdots + \mu_{k_n} \mapsto \mu n$. The memory functions naturally [12] become $\gamma_{\boldsymbol{k}_1,\ldots,\boldsymbol{k}_n}(t) \mapsto \mu \Lambda \phi_{n+1}(t)$. Neglecting inertial effects and including the bare viscosities arising from the short-time portion of the memory functions we arrive at the schematic hierarchy

$$\dot{\phi}_n(t) + \mu n \phi_n(t) + \mu \Lambda (\phi_{n+1} * \dot{\phi}_n)(t) = 0. \qquad (2)$$

From now on we set $\mu = 1$ as it can be absorbed by a rescaling of time. Note that Eq. (2) reduces to the F_2 model when closed with $\phi_2(t) = \phi_1^2(t)$ [1,13]. The drastic simplifications inherent in our schematic hierarchy clearly prohibit a quantitative interpretation of its predictions. Nevertheless, one would expect that Eq. (2) captures the general phenomenology of GMCT in a similar way as the F_2 model does for MCT.

The entire analysis of our schematic hierarchy is based on one central identity. It applies in Laplace transformed representation, $\hat{\phi}_n(s) = \mathcal{L}\{\phi_n(t)\}$, where Eq. (2) reads

$$\hat{\phi}_n(s) = \left[s + \frac{n}{1 + \Lambda \hat{\phi}_{n+1}(s)} \right]^{-1}. \qquad (3)$$

Recursion of this equation produces a continued fraction. Using various transformations (all mathematical details will be given in [14]), however, we derive an identity that directly relates any two functions $\hat{\phi}_m$ and $\hat{\phi}_n$:

$$\Omega_m(s) = \Omega_n(s) \quad \text{with} \quad \Omega_n(s) = \Gamma(n) \frac{v_n(s)}{u_n(s)}, \qquad (4)$$

where $\Gamma(z)$ denotes the Gamma function and

$$u_n(s) = \Psi(s+1, s+n+1; \Lambda) + \hat{\phi}_n(s) \Psi(s, s+n; \Lambda),$$
$$v_n(s) = \Phi(s+1, s+n+1; \Lambda) - \hat{\phi}_n(s) \Phi(s, s+n; \Lambda).$$

The latter expressions contain the (regularized) confluent hypergeometric functions $\Phi(a, b; z) = {}_1F_1(a, b; z)/\Gamma(b)$ and $\Psi(a, b; z)$ [15]. Note that $\Omega_m(s) = \Omega_n(s)$ for any m, n implies that $\Omega_n(s)$ is independent of n and thus invariant under (3). So any single $\hat{\phi}_m(s)$ determines the invariant $\Omega(s) \equiv \Omega_m(s)$ and from that, in turn, all $\hat{\phi}_n(s)$ follow by simply rearranging $\Omega(s) = \Omega_n(s)$.

In GMCT the hierarchy (2) is closed through factorization at some order $N \geq 2$. In general this amounts to $\phi_N(t) = \phi_{n_1}^{m_1}(t) \phi_{n_2}^{m_2}(t) \cdots \phi_{n_k}^{m_k}(t)$. Here orders n_i appear with multiplicity $m_i \geq 1$ such that $N = n_1 m_1 + n_2 m_2 + \cdots + n_k m_k$. In schematic MCT $\phi_2 = \phi_1^2$, while GMCT closures considered previously correspond to $\phi_3 = \phi_1 \phi_2$ [9] and $\phi_4 = \phi_1^2 \phi_2$ [10]. It turns out that for *any* factorization there is a critical coupling Λ_c above which the $\phi_n(t)$ do not relax fully, i.e., $q_n = \phi_n(t \to \infty) > 0$. This can be deduced from Eq. (4): associated with each closure is a polynomial, say, in q_1, whose real roots are dynamical fixed points. Some examples of critical couplings Λ_c, where real roots appear, and the corresponding plateau heights q_1 for low order closures are listed in Table I. We observe that within each order N the value of Λ_c is lowest for the factorization $\phi_N = \phi_1^N$ while it is largest for $\phi_N = \phi_n \phi_{N-n}$ with n the integer part of $N/2$. More importantly, there is an overall increase in Λ_c as we raise N. Indeed, for the factorization $\phi_N = \phi_1^N$ it can be shown that $\Lambda_c \sim N$ for $N \gg 1$. Including higher order correlations does not merely affect Λ_c quantitatively but in fact allows us to increase its value *unboundedly*. Implications of this result will become clear in the following.

The dynamics of GMCT under higher order factorization closures, which has never been investigated, can be obtained (schematically) by numerical integration of Eq. (2). Because of factorization of $\phi_N(t)$ the hierarchy reduces to a set of coupled nonlinear integrodifferential equations in $\{\phi_1(t), \ldots, \phi_{N-1}(t)\}$. It is efficiently integrated by the algorithm of [16]. The results in GMCT with $N \geq 3$ turn out to be remarkably similar to those of MCT. For any given factorization, $\phi_1(t)$ displays two-step relaxation when Λ approaches the relevant critical value Λ_c from below. As $\Lambda \to \Lambda_c$ the α-relaxation time τ diverges and $\phi_1(t)$ assumes the nonzero limit $q_1 = \phi_1(t \to \infty)$. We have performed a scaling analysis, which is based on Eq. (4) and $\hat{\phi}_N(s) = \mathcal{L}\{\prod_i \phi_{n_i}^{m_i}(t)\}$, to precisely charac-

TABLE I. Critical couplings Λ_c and associated plateaus $q_1 = \phi_1(t \to \infty)$ for all factorizations of $\phi_N(t)$ with $N = 2, 3, 4$.

	ϕ_1^2	ϕ_1^3	$\phi_1\phi_2$	ϕ_1^4	$\phi_1^2\phi_2$	ϕ_2^2	$\phi_1\phi_3$
Λ_c	4	4.4922	4.8284	4.9398	5.2359	5.6946	5.6046
q_1	$\frac{1}{2}$	0.5453	0.5858	0.5839	0.6205	0.6782	0.6565

terize the critical properties near Λ_c. It can be shown that the scaling equations for our hierarchy are in fact formally identical to those for usual schematic models [1]. Hence β relaxation follows the power laws $\phi_1(t) - q_1 \sim t^{-a}$ and $q_1 - \phi_1(t) \sim t^b$ in the early and late regimes, respectively, and the α-relaxation time diverges like $\tau \sim 1/(\Lambda_c - \Lambda)^\gamma$. Strikingly the exponents a, b, and γ for these scaling laws are *identical* for *all* factorization closures $\phi_N = \prod_i \phi_{n_i}^{m_i}$. Their values exactly match those of the F_2 model [1], that is, $a \approx 0.395$, $b = 1$, and $\gamma = \frac{1}{2a} + \frac{1}{2b}$, regardless of closure.

While tempting, the above results cannot be interpreted as supporting the standard MCT picture. The occurrence of the robust critical scaling under higher order closures is systematically deferred along with the critical point $\Lambda_c \sim N$ itself. We turn to the nonperturbative limit $N \to \infty$ of avoided factorization to develop a deeper understanding. According to our general result (4) all functions $\hat{\phi}_n(s)$ are known once we have determined the invariant $\Omega(s)$. *A priori* the latter depends on how we close the hierarchy since we may write $\Omega(s) = \Omega_N(s)$. However, analysis of $\Omega_N(s)$ shows that for all physically reasonable closures the invariant of the infinite hierarchy vanishes $\Omega(s) = \lim_{N\to\infty}\Omega_N(s) = 0$. Thus the infinite hierarchy has a *unique* solution. Rearranging $\Omega_n(s) = \Omega(s) = 0$ then produces the exact result

$$\hat{\phi}_n(s) = \frac{\Phi(s+1, s+n+1; \Lambda)}{\Phi(s, s+n; \Lambda)}. \tag{5}$$

The regularized confluent hypergeometric functions Φ are analytic in s. Therefore the only singularities in Eq. (5) are (first order) poles at points $\{s_i\}$ where the denominator $\Phi(s_i, s_i + n; \Lambda) = 0$ vanishes. The relaxation spectrum $\{s_i\}$ is infinite, discrete, and contained in the negative real axis. Consequently, the inverse Laplace transform of Eq. (5) is of the form $\phi_n(t) = \sum_{i=0}^{\infty} r_i e^{s_i t}$ with r_i the residues at $s = s_i$. Numerical evaluation of both $\{s_i\}$ and $\{r_i\}$ is straightforward. Plots of the solutions $\phi_1(t)$ of the infinite hierarchy are shown in Fig. 1. Upon increasing Λ a single component s_0 of the spectrum approaches the origin. Hence the shape of α relaxation in our schematic hierarchy is exponential with τ defined by $s_0 t = -t/\tau$. From $\Phi(s_0, s_0 + 1; \Lambda) = 0$ one shows that $\tau \sim e^{\Lambda}/\Lambda$ for $\Lambda \gg 1$. There is *no* MCT *transition* in the infinite hierarchy at any finite Λ. Instead, the relaxation time τ essentially grows *exponentially* in Λ. Remarkably, this hallmark of nonperturbative behavior [3] emerges from our microscopically motivated dynamical approach.

We now discuss how the above results merge into a consistent picture. In the infinite hierarchy, memory effects first appear in the relaxation of $\phi_1(t)$ but gradually spread into the hierarchy upon increasing Λ. This causes the exponential slowdown in the dynamics of $\phi_1(t)$. The MCT closure $\phi_2 = \phi_1^2$, while appropriate for small Λ, generally overestimates the relaxation time of $\phi_2(t)$. As

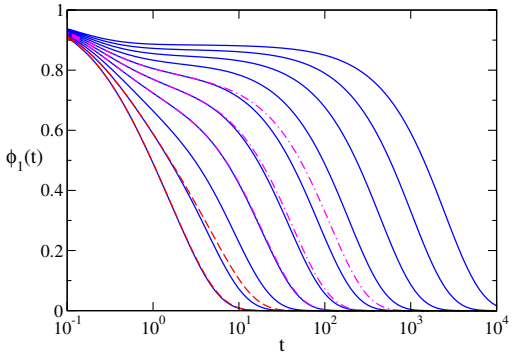

FIG. 1 (color online). Exact solutions of the infinite hierarchy [Eq. (5)] at $\Lambda = 1, 2, \ldots, 10$ (solid curves), MCT with closure $\phi_2 = \phi_1^2$ at $\Lambda = 1, 2$ (dashed curves), and GMCT under $\phi_7 = \phi_3\phi_4$ at $\Lambda = 4, 5, 6$ (dash-dotted curves). See text for discussion.

shown in Fig. 1 the MCT solution matches that of the infinite hierarchy at $\Lambda = 1$ but already deviates at $\Lambda = 2$. When Λ approaches $\Lambda_c = 4$ the feedback through the closure drives the MCT solution into a spurious transition, while in the infinite hierarchy the relaxation of $\phi_1(t)$ develops only a shoulder (see Fig. 1 at $\Lambda = 4$). This is also typically observed in experiments and simulations at the point where MCT *predicts* a glass transition [2]. If, however, the factorization is delayed to higher order correlations, then so is the appearance of feedback through the closure. We refer again to Fig. 1, which also shows GMCT solutions under factorization $\phi_7 = \phi_3\phi_4$. These perfectly match the infinite hierarchy even at $\Lambda = 4$, but again deviate as Λ is raised further towards the critical value $\Lambda_c \approx 8.1049$ of this closure. Of course the validity of GMCT may be extended to any value Λ by raising N. In the nonperturbative limit the transition is deferred to $\Lambda_c \sim N \to \infty$ and we arrive at the ergodic solution (5) of the infinite hierarchy. The fact that the $\phi_n(t)$ derived from Eq. (5) do not obey any factorization corresponds to underlying *non-Gaussian* dynamical fluctuations [11]. In microscopic systems these are a natural consequence of cooperative, heterogeneous dynamics—not fully accounted for in MCT due to Gaussian factorization. However, microscopic GMCT offers the potential to systematically capture non-Gaussian fluctuations and thus the underlying processes that cause them.

Strikingly the solutions of the infinite hierarchy nevertheless carry some important features consistent with MCT. Figure 2 shows GMCT solutions under factorization at orders $N = 2, 3, \ldots, 7$ at the corresponding critical couplings Λ_c. From our discussion of the dynamics in GMCT we know that each of these functions approaches its relevant plateau q_1 (see Table I) like a power law t^{-a} with the MCT exponent $a \approx 0.395$. Now compare these critical solutions to those of the infinite hierarchy (Fig. 2) at the same values of Λ: apparently there is an excellent match throughout the early β-relaxation regime. In this sense the

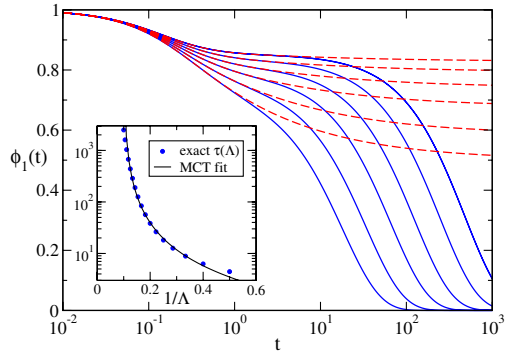

FIG. 2 (color online). Main panel: Critical solutions (dashed lines) for closures ϕ_1^2, $\phi_1\phi_2$, ϕ_2^2, $\phi_2\phi_3$, ϕ_3^2, $\phi_3\phi_4$ at $\Lambda_c \approx 4$, 4.8, 5.7, 6.5, 7.3, 8.1, respectively, and solutions of the infinite hierarchy at the same Λ (solid lines). Inset: α-relaxation time of the infinite hierarchy (dots) for $\Lambda = 2, 2.5, \ldots, 10$ and an MCT fit (solid curve), $\tau_{\text{fit}} = A/(\Lambda^{-1} - \Lambda_c^{-1})^{\gamma}$, with $\gamma \approx 1.765$ fixed but $A \approx 0.7$ and $\Lambda_c \approx 10.1$ fitted.

hierarchy *appears* to approach criticality with a β-relaxation exponent a as predicted by MCT. Likewise, we could extract the MCT exponent $b = 1$ characterizing late β relaxation where $\phi_1(t)$ drops below its plateau. A consistent MCT picture would then require power-law scaling of the relaxation time τ with a particular exponent γ. For fitting purposes this power law is usually expressed in $T - T_c$ rather than $\Lambda_c - \Lambda$, which is equivalent (asymptotically) close to criticality. Noting that $1/\Lambda$ is generally an increasing function of T [1] (and thus represents a temperaturelike variable) we write $\tau_{\text{fit}} = A/(\Lambda^{-1} - \Lambda_c^{-1})^{\gamma}$. Most remarkably, this power law with the MCT exponent $\gamma \approx 1.765$ indeed fits the relaxation time of the infinite hierarchy well over $3 \leq \Lambda \leq 9$ covering, as in simulations [2], about two to three decades in τ; see inset of Fig. 2. Thus, within this limited dynamical range the solutions of the infinite hierarchy exhibit power-law behaviors characterized by the MCT exponents a, b, and γ. The fitted $\Lambda_c \approx 10.1$, lying well above the MCT value $\Lambda_c = 4$, is consistent with correspondingly lower T_c extrapolated in experiments and simulations [2]. However, contrary to MCT these power laws do not reflect asymptotic scaling in the infinite hierarchy. In fact, there is no divergence of the relaxation time $\tau \sim e^{\Lambda}/\Lambda$ at any finite Λ, and, in particular, not at $\Lambda_c \approx 10.1$ as extrapolated from the power-law fit.

The entire phenomenology discussed above emerges purely from the structure of our schematic hierarchy (2). The qualitative differences between the nonperturbative and MCT solutions of Eq. (2) in several aspects resemble those found when contrasting experimental or simulation data [2,6,17] with microscopic ($\boldsymbol{k}$-dependent) MCT, e.g., predicted versus fitted T_c or the apparent power-law behaviors and deviations away from them. Our analysis resolves these discrepancies, leading to a coherent picture based upon non-Gaussian dynamical fluctuations. To describe the relaxation of supercooled liquids in full detail, including, e.g., stretched exponentials, numerical analysis of microscopic ($\boldsymbol{k}$-dependent) GMCT will be necessary. One would hope that such an approach preserves the successful features of MCT, for instance, accurate prediction of the plateau values. Beyond that, however, it might not only offer a possibility to systematically improve the predictions of MCT, particularly through the nonperturbative limit, but also provide a kinetic approach to capture dynamical heterogeneity. The implications of the work presented here on related issues, such as the breakdown of the Stokes-Einstein relation, will be explored in future work.

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