

Mode-coupling theory of the glass transition for polydisperse systems

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We develop a covariant formulation of the mode-coupling theory (MCT) for the glass transition in polydisperse suspensions. We show that the Mori-Zwanzig equations of motion and the MCT memory functional are invariant under arbitrary changes of basis in the space of species-resolved density fluctuations, provided the vertices are transformed consistently. Exploiting this symmetry, we construct a mode basis via a diameter-moment expansion and an orthonormalization with respect to the structure factor. In this basis, the leading mode is the total density, and higher modes encode systematic size-contrast corrections. Truncating to a few modes yields a controlled, numerically stable reduction of the multicomponent MCT while retaining quantitative accuracy for the total intermediate scattering function. Benchmarking against a ten-component discretization of continuously polydisperse hard-sphere-like fluids (Schulz-Zimm and inverse-cubic distributions), we find that three to six modes reproduce the critical packing fraction and the full shape of the total correlator with large computational savings. This reformulation provides an efficient framework for the MCT of polydisperse systems with controlled discretization and basis-truncation errors.

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I. INTRODUCTION

Glasses are amorphous solids that exhibit short-range but no long-range order and occupy an intermediate state between crystalline solids and liquids. They form when a liquid is cooled or compressed rapidly enough to avoid crystallization and enters a supercooled regime in which small changes in temperature or density cause large variations in transport coefficients. Upon further approach to the glass transition, transport ceases on accessible timescales and a structural arrest occurs. The dynamics near this arrest display characteristic features such as dynamical heterogeneity and a two-step relaxation [1–5].

Colloidal suspensions provide a versatile setting to investigate this phenomenology. At high concentrations they undergo a glass transition accompanied by a drastic viscosity increase and loss of equilibration on experimental timescales. In addition, the size of colloidal particles (typically 0.01–1 μm) allows for optical investigation, and their hard-sphere-like interactions make them a benchmark system for theory and simulation [6–12]. A defining feature of experimental and simulated colloids is size polydispersity, either inherited from synthesis or deliberately introduced to suppress crystallization. Its impact on structure and dynamics has been characterized in numerous experiments and simulations [13–17].

The mode-coupling theory (MCT) of the glass transition captures many qualitative and semiquantitative aspects of

slow structural relaxation using only static two-point correlations as input [4,5,18]. Near the transition, MCT predicts the emergence of a two-step relaxation and scaling laws for correlation functions and timescales [4,19,20]. The central observable is the intermediate scattering function (ISF), the time-dependent density-density correlator, which encodes the slow cage dynamics. For colloids, MCT has been formulated for overdamped (Smoluchowski) dynamics [21], and the resulting long-time structural relaxation is controlled by the equilibrium structure rather than by the short-time microscopic dynamics [22–25].

Multicomponent generalizations of MCT are well established and have been applied successfully to binary mixtures and to complex glass-forming melts [26–35]. However, the numerical effort grows rapidly with the number of components because the MCT memory kernel couples pairs of density modes across species. This presents a bottleneck for polydisperse systems, which are commonly approximated by discretizing the particle-size distribution into many species [36,37]. Recently, continuously polydisperse hard-sphere fluids have been investigated within MCT [37], but the attainable number of bins remains constrained by computational cost, with the memory-kernel evaluation scaling steeply with component number.

In this work, we address this challenge by exploiting a symmetry of the theory. We show that the Mori-Zwanzig equations of motion and the MCT memory functional are covariant under arbitrary changes of basis in the space of species-resolved density fluctuations, provided the vertices are transformed consistently. Building on this, we construct a mode basis from size moments around the mean diameter and perform an orthonormalization. In this basis, the leading mode is the total density and higher modes encode systematic size-contrast corrections. Truncating the hierarchy

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at a small number of modes yields a reduced set of coupled equations that retains the total ISF with high accuracy while drastically lowering the computational load. For mixtures evolving under Smoluchowski dynamics, the partial correlators are completely monotone [38,39], which supports the safety of such a truncation.

The remainder of the paper is organized as follows. In Sec. II we define the observables and present the multicomponent MCT equations of motion. In Sec. III we establish the covariance under basis changes, construct the transformation matrix via Gram-Schmidt orthogonalization, and derive the transformed equations, including the correct transformation of the vertices. Section IV details the numerical implementation and the generation of discretized diameter sets. In Sec. V we present results for the critical packing fraction as a function of polydispersity and mode cutoff, and compare total ISFs obtained with different truncations against full multicomponent calculations and against a naive reduction in component number. We conclude in Sec. VI with a discussion and outlook.

II. THEORY AND OBSERVABLES

We consider a size-polydisperse colloidal system comprising s species with particle numbers N_α and diameters σ_α for $\alpha = 1, \dots, s$, enclosed in a box of volume V . The total particle number is $N = \sum_{\alpha=1}^s N_\alpha$, and the species concentrations are $x_\alpha = N_\alpha/N$. The thermodynamic limit $N \rightarrow \infty$ and $V \rightarrow \infty$ at fixed number density $n = N/V$ and fixed x_α is anticipated. A configuration is specified by the particle positions $\mathbf{\Gamma} = (\mathbf{r}_{\alpha,i_\alpha} : \alpha = 1, \dots, s, i_\alpha = 1, \dots, N_\alpha) \in V^N$.

In this work, we focus on overdamped dynamics and neglect hydrodynamic interactions. The conditional probability density $\mathbb{P}(\mathbf{\Gamma}, t | \mathbf{\Gamma}_0)$ to find the system in configuration $\mathbf{\Gamma}$ at time t given $\mathbf{\Gamma}_0$ at $t = 0$ obeys the Smoluchowski equation

$$\partial_t \mathbb{P} = \Omega \mathbb{P}, \quad (1)$$

with the Smoluchowski operator

$$\Omega = \Omega(\mathbf{\Gamma}) = \sum_{\alpha=1}^s \sum_{i_\alpha=1}^{N_\alpha} D_\alpha^0 \frac{\partial}{\partial \mathbf{r}_{\alpha,i_\alpha}} \cdot \left[\frac{\partial}{\partial \mathbf{r}_{\alpha,i_\alpha}} + \frac{1}{k_B T} \frac{\partial U(\mathbf{\Gamma})}{\partial \mathbf{r}_{\alpha,i_\alpha}} \right], \quad (2)$$

where D_α^0 is the short-time diffusion coefficient of species α and $U(\mathbf{\Gamma})$ is the potential energy.

For two observables $A(\mathbf{\Gamma})$ and $B(\mathbf{\Gamma})$ the time-correlation function is

$$C_{AB}(t) = \int d\mathbf{\Gamma} \int d\mathbf{\Gamma}_0 \delta A(\mathbf{\Gamma})^* \mathbb{P}(\mathbf{\Gamma}, t | \mathbf{\Gamma}_0) \delta B(\mathbf{\Gamma}_0) \mathbb{P}_{\text{eq}}(\mathbf{\Gamma}_0), \quad (3)$$

with fluctuations $\delta A = A - \langle A \rangle$ and canonical averaging $\langle \cdot \rangle$ over the equilibrium distribution $\mathbb{P}_{\text{eq}}(\mathbf{\Gamma}) = Z^{-1} \exp[-U(\mathbf{\Gamma})/k_B T]$. Using the formal solution $\mathbb{P}(\mathbf{\Gamma}, t | \mathbf{\Gamma}_0) = \exp(\Omega t) \delta(\mathbf{\Gamma} - \mathbf{\Gamma}_0)$ and integration by parts yields

$$C_{AB}(t) = \int d\mathbf{\Gamma} \delta A(\mathbf{\Gamma}, t)^* \delta B(\mathbf{\Gamma}) \mathbb{P}_{\text{eq}}(\mathbf{\Gamma}), \quad (4)$$

where $\delta A(\mathbf{\Gamma}, t) = \mathcal{R}(t) \delta A(\mathbf{\Gamma})$ and $\mathcal{R}(t) = \exp(\Omega^\dagger t)$ evolves observables with the adjoint Smoluchowski operator. The

adjoint takes the form [27]

$$\Omega^\dagger = \sum_{\alpha=1}^s \sum_{i_\alpha=1}^{N_\alpha} D_\alpha^0 \left[\frac{\partial}{\partial \mathbf{r}_{\alpha,i_\alpha}} - \frac{1}{k_B T} \frac{\partial U(\mathbf{\Gamma})}{\partial \mathbf{r}_{\alpha,i_\alpha}} \right] \cdot \frac{\partial}{\partial \mathbf{r}_{\alpha,i_\alpha}}. \quad (5)$$

We write correlation functions succinctly as

$$C_{AB}(t) = \langle A(t) | B \rangle = \langle A | \mathcal{R}(t) | B \rangle, \quad (6)$$

where $\langle A | B \rangle$ denotes the Kubo scalar product

$$\langle A | B \rangle := \int d\mathbf{\Gamma} \delta A(\mathbf{\Gamma})^* \delta B(\mathbf{\Gamma}) \mathbb{P}_{\text{eq}}(\mathbf{\Gamma}). \quad (7)$$

We consider the species-resolved density modes at wave vector $\mathbf{q} \neq \mathbf{0}$,

$$\rho_\alpha(\mathbf{q}, t) = \sum_{i_\alpha=1}^{N_\alpha} \exp[i\mathbf{q} \cdot \mathbf{r}_{\alpha,i_\alpha}(t)]. \quad (8)$$

For $\mathbf{q} \neq \mathbf{0}$ the average vanishes and $\delta \rho_\alpha(\mathbf{q}) = \rho_\alpha(\mathbf{q})$. The partial ISFs are

$$S_{\alpha\beta}(\mathbf{q}, t) := \frac{1}{N} \langle \rho_\alpha(\mathbf{q}, t) | \rho_\beta(\mathbf{q}) \rangle, \quad (9)$$

which by isotropy depend only on $q = |\mathbf{q}|$. The static partial structure factors are $S_{\alpha\beta}(q) := S_{\alpha\beta}(q, 0)$, and the total ISF is $S(q, t) = \sum_{\alpha=1}^s \sum_{\beta=1}^s S_{\alpha\beta}(q, t)$ with static counterpart $S(q) = \sum_{\alpha=1}^s \sum_{\beta=1}^s S_{\alpha\beta}(q)$. The species-resolved short-time diffusion matrix follows from

$$D_{\alpha\beta}(q) := -\frac{1}{N} \langle \rho_\alpha(\mathbf{q}) | \Omega^\dagger \rho_\beta(\mathbf{q}) \rangle = q^2 D_\alpha^0 x_\alpha \delta_{\alpha\beta}. \quad (10)$$

We assemble $S_{\alpha\beta}(q, t)$ and $D_{\alpha\beta}(q)$ into $s \times s$ matrices $\mathbf{S}(q, t)$ and $\mathbf{D}(q)$. Both are symmetric and $\mathbf{D}(q)$ is diagonal in the species basis since hydrodynamic interactions are ignored.

Following the standard MCT projection scheme [4,21,40], the matrix of partial ISFs obeys

$$\begin{aligned} \dot{S}_{\alpha\beta}(q, t) &+ \sum_{\gamma=1}^s \sum_{\delta=1}^s D_{\alpha\gamma}(q) [\mathbf{S}^{-1}(q)]^{\gamma\delta} S_{\delta\beta}(q, t) \\ &+ \sum_{\gamma=1}^s \sum_{\delta=1}^s \int_0^t \mathcal{M}_{\alpha\gamma}(q, t-t') [\mathbf{D}^{-1}(q)]^{\gamma\delta} \dot{S}_{\delta\beta}(q, t') dt' \\ &= 0, \end{aligned} \quad (11)$$

with the memory kernel matrix $\mathcal{M}(q, t)$. In the preceding equation, we formally introduced upper (contravariant) and lower (covariant) indices as familiar from linear algebra in the context of basis transforms in vector spaces and their duals. Note that contractions are always between one contravariant and one covariant index. Thus, inverses of matrices with covariant indices naturally acquire contravariant indices.

Introducing the contravariant reduced memory kernel

$$M^{\alpha\beta}(q, t) = \sum_{\gamma=1}^s \sum_{\delta=1}^s [\mathbf{D}^{-1}(q)]^{\alpha\gamma} \mathcal{M}_{\gamma\delta}(q, t) [\mathbf{D}^{-1}(q)]^{\delta\beta}, \quad (12)$$

or equivalently in matrix form

$$\mathbf{M}(q, t) := \mathbf{D}^{-1}(q) \mathcal{M}(q, t) \mathbf{D}^{-1}(q), \quad (13)$$

we obtain the compact matrix form

$$\dot{\mathbf{S}}(q, t) + \mathbf{D}(q)\mathbf{S}^{-1}(q)\mathbf{S}(q, t) + \int_0^t \mathbf{D}(q)\mathbf{M}(q, t-t')\dot{\mathbf{S}}(q, t')dt' = 0. \quad (14)$$

Within MCT, the kernel is approximated by a bilinear functional of the ISFs that enforces wave-vector conservation,

$$M^{\alpha\beta}(q, t) = \frac{n^2}{2q^2N} \sum_{\mathbf{q}_1+\mathbf{q}_2=\mathbf{q}} \sum_{\gamma=1}^s \sum_{\delta=1}^s \sum_{\kappa=1}^s \sum_{\lambda=1}^s \mathcal{V}^{\alpha,\gamma\delta}(\mathbf{q}, \mathbf{q}_1\mathbf{q}_2) \times S_{\gamma\kappa}(q_1, t) S_{\delta\lambda}(q_2, t) \mathcal{V}^{\beta,\kappa\lambda}(\mathbf{q}, \mathbf{q}_1\mathbf{q}_2)^*. \quad (15)$$

Here, the sum over the wave vectors $\mathbf{q}_1, \mathbf{q}_2$ is restricted such that $\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{q}$, reflecting the translational invariance. The components of the vertices $\mathcal{V}(\mathbf{q}, \mathbf{q}_1\mathbf{q}_2)$ are

$$\mathcal{V}^{\alpha,\gamma\delta}(\mathbf{q}, \mathbf{q}_1\mathbf{q}_2) = \frac{\mathbf{q}}{qx_\alpha} \cdot [\mathbf{q}_1 \delta_\alpha^\delta c^{\alpha\gamma}(q_1) + \mathbf{q}_2 \delta_\alpha^\gamma c^{\alpha\delta}(q_2)], \quad (16)$$

where $c^{\alpha\beta}(q)$ are the matrix elements of the multicomponent direct correlation function defined via the Ornstein-Zernike relation (specified in Sec. III). Equations (14)–(16) close the MCT for the partial ISFs once the static inputs $S_{\alpha\beta}(q)$ [or equivalently $c^{\alpha\beta}(q)$] are provided.

In principle, Eqs. (14)–(16) allow one to compute the full matrix $\mathbf{S}(q, t)$ at each time, wave number, and species pair [37]. Direct evaluation, however, is computationally demanding because the memory kernel involves six sums over all species, leading to a complexity of $O(s^6)$. Even after exploiting the matrix symmetries, the computational effort remains substantial for many-component systems.

III. COVARIANCE OF THE MCT EQUATIONS AND TRANSFORMATION MATRIX

We now demonstrate that the multicomponent MCT equations are covariant under arbitrary changes of basis in the space of species-resolved density fluctuations, provided the vertices are transformed consistently. This enables us to construct a mode basis adapted to size polydispersity and to introduce a controlled truncation.

Consider, for each wave vector, an invertible transformation between the species basis $[\rho_1(\mathbf{q}), \dots, \rho_s(\mathbf{q})]$ and a new basis $[\tilde{\rho}_1(\mathbf{q}), \dots, \tilde{\rho}_s(\mathbf{q})]$. Then the old basis vectors can be expanded in the new basis as

$$\rho_\alpha(\mathbf{q}, t) = \sum_{a=1}^s \tilde{\rho}_a(\mathbf{q}, t) L_\alpha^a(\mathbf{q}), \quad (17)$$

with matrix elements $L_\alpha^a(\mathbf{q}) := [\mathbf{L}(\mathbf{q})]_\alpha^a$ of the transformation matrix $\mathbf{L}(\mathbf{q})$, which will be defined in detail below. We reserve Greek indices $(\alpha, \beta, \dots)$ for the species basis and use Latin indices $(a, b, \dots)$ for the new basis. The inverse transformation $\mathbf{L}^{-1}(\mathbf{q})$ has elements $(L^{-1})_a^\alpha(\mathbf{q}) \equiv [\mathbf{L}^{-1}(\mathbf{q})]_a^\alpha$, and by construction

$$\sum_{a=1}^s (L^{-1})_a^\alpha(\mathbf{q}) L_\beta^a(\mathbf{q}) = \delta_\beta^\alpha, \quad \sum_{\alpha=1}^s L_\alpha^a(\mathbf{q}) (L^{-1})_b^\alpha(\mathbf{q}) = \delta_b^a. \quad (18)$$

Two-index covariant objects [e.g., the ISF $S_{\alpha\beta}(q, t)$ or the diffusion matrix $D_{\alpha\beta}(q)$, etc.]

$$\tilde{S}_{ab}(q, t) = \sum_{\alpha=1}^s \sum_{\beta=1}^s S_{\alpha\beta}(q, t) (L^{-1})_a^\alpha(\mathbf{q}) (L^{-1})_b^\beta(\mathbf{q}), \quad (19)$$

or, in matrix form, $\tilde{\mathbf{S}}(q, t) = \mathbf{L}^{-T}(\mathbf{q})\mathbf{S}(q, t)\mathbf{L}^{-1}(\mathbf{q})$, where $\mathbf{L}^{-T}(\mathbf{q}) \equiv [\mathbf{L}^{-1}(\mathbf{q})]^T$. By construction,

$$\tilde{S}_{ab}(q, t) = \frac{1}{N} \langle \tilde{\rho}_a(\mathbf{q}, t) | \tilde{\rho}_b(\mathbf{q}) \rangle, \quad (20)$$

i.e., the definition of the ISF is preserved in the new basis.

In contrast, two-index contravariant objects [e.g., the reduced memory kernel $M^{\alpha\beta}(q, t)$, the inverse static structure factor $[\mathbf{S}^{-1}(q)]^{\alpha\beta}$, or the inverse diffusion matrix $[\mathbf{D}^{-1}(q)]^{\alpha\beta}$] transform as

$$\tilde{M}^{ab}(q, t) = \sum_{\alpha=1}^s \sum_{\beta=1}^s L_\alpha^a(\mathbf{q}) L_\beta^b(\mathbf{q}) M^{\alpha\beta}(q, t), \quad (21)$$

with matrix form

$$\tilde{\mathbf{M}}(q, t) = \mathbf{L}(\mathbf{q})\mathbf{M}(q, t)\mathbf{L}^T(\mathbf{q}). \quad (22)$$

Applying $\mathbf{L}(\mathbf{q})$ from the left and $\mathbf{L}^T(\mathbf{q})$ from the right to Eq. (14) yields the same form in the new basis,

$$\dot{\tilde{\mathbf{S}}}(q, t) + \tilde{\mathbf{D}}(q)\tilde{\mathbf{S}}^{-1}(q)\tilde{\mathbf{S}}(q, t) + \int_0^t \tilde{\mathbf{D}}(q)\tilde{\mathbf{M}}(q, t-t')\dot{\tilde{\mathbf{S}}}(q, t')dt' = 0, \quad (23)$$

so covariance of the MCT equation of motion is guaranteed by the specified transformation laws of $\mathbf{S}$, $\mathbf{D}$, and $\mathbf{M}$.

To preserve form invariance also at the level of the MCT closure, the memory kernel in the new basis must retain the same bilinear structure. With the three-wave-vector representation and wave-vector conservation $\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{q}$, we write

$$\tilde{M}^{ab}(q, t) = \frac{n}{2q^2} \sum_{\mathbf{q}_1+\mathbf{q}_2=\mathbf{q}} \sum_{c=1}^s \sum_{d=1}^s \sum_{\kappa=1}^s \sum_{\lambda=1}^s \tilde{\mathcal{V}}^{a,cd}(\mathbf{q}; \mathbf{q}_1, \mathbf{q}_2) \times \tilde{S}_{ck}(q_1, t) \tilde{S}_{dl}(q_2, t) \tilde{\mathcal{V}}^{b,kl}(\mathbf{q}; \mathbf{q}_1, \mathbf{q}_2)^*, \quad (24)$$

provided the vertices transform according to

$$\tilde{\mathcal{V}}^{a,bc}(\mathbf{q}; \mathbf{q}_1, \mathbf{q}_2) = L_\alpha^a(\mathbf{q}) L_\beta^b(\mathbf{q}_1) L_\gamma^c(\mathbf{q}_2) \mathcal{V}^{\alpha,\beta\gamma}(\mathbf{q}; \mathbf{q}_1, \mathbf{q}_2), \quad (25)$$

at its own wave vector.

In the species basis, the vertex assumes the simple form given in Eq. (16), which involves the direct correlation function $c^{\alpha\beta}(q)$ defined via the multicomponent Ornstein-Zernike relation [27,41–43]

$$[\mathbf{S}^{-1}(q)]^{\alpha\beta} = [\mathbf{X}^{-1}]^{\alpha\beta} - nc^{\alpha\beta}(q), \quad [\mathbf{X}^{-1}]^{\alpha\beta} = \frac{1}{x_\alpha} \delta^{\alpha\beta}, \quad (26)$$

so that $c^{\alpha\beta}(q)$ is contravariant in this convention.

Comparing Eq. (16) with the transformed structure shows that the explicit vertex expression is not form invariant. Likewise, the inverse number-concentration matrix $\mathbf{X}^{-1}$ entering the Ornstein-Zernike relation is diagonal only in the species basis. Hence, the species basis is a distinguished basis for deriving the MCT closure. Once the transformation law (25)

is imposed, however, the theory is covariant and the closure retains its bilinear form in the new basis.

We exploit the covariance to construct a mode basis adapted to size polydispersity. Following a diameter-moment expansion [44], we define the Vandermonde matrix

$$E_a^\alpha = \left(\frac{\sigma_\alpha - \bar{\sigma}}{\bar{\sigma}} \right)^{a-1}, \quad \bar{\sigma} = \sum_{\alpha=1}^s x_\alpha \sigma_\alpha, \quad (27)$$

and the corresponding unnormalized moment modes

$$\mu_a(\mathbf{q}, t) = \sum_{\alpha=1}^s \rho_\alpha(\mathbf{q}, t) E_a^\alpha. \quad (28)$$

Since $E_1^\alpha = 1$, the lowest moment mode is the total density, $\mu_1(\mathbf{q}, t) = \sum_{\alpha} \rho_\alpha(\mathbf{q}, t)$. The static correlator matrix in the moment basis is

$$S_{ab}^E(q) := \frac{1}{N} \langle \mu_a(\mathbf{q}) | \mu_b(\mathbf{q}) \rangle = \sum_{\alpha=1}^s \sum_{\beta=1}^s S_{\alpha\beta}(q) E_a^\alpha E_b^\beta. \quad (29)$$

In particular, the second moment mode ($a = 2$) carries the first centered size contrast,

$$\mu_2(\mathbf{q}, t) = \sum_{\alpha=1}^s \left(\frac{\sigma_\alpha - \bar{\sigma}}{\bar{\sigma}} \right) \rho_\alpha(\mathbf{q}, t). \quad (30)$$

It is the density mode weighted by deviations of the species diameters from the mean. Equivalently, μ_2 probes fluctuations of the local average diameter at fixed overall density. For a binary mixture with species A and B this reduces to

$$\begin{aligned} \mu_2(\mathbf{q}, t) &= \frac{\sigma_A - \bar{\sigma}}{\bar{\sigma}} \rho_A(\mathbf{q}, t) + \frac{\sigma_B - \bar{\sigma}}{\bar{\sigma}} \rho_B(\mathbf{q}, t) \\ &\propto (\sigma_A - \sigma_B) c(\mathbf{q}, t), \end{aligned} \quad (31)$$

where $c(\mathbf{q}, t)$ is the concentration mode.

Direct use of the Vandermonde matrix can be ill-conditioned for large a because its entries scale as powers of a , Eq. (27). Consequently, a change of basis using this matrix yields covariant objects with small entries and contravariant ones with correspondingly large entries, which makes numerical handling more challenging. It is thus preferable to restrict the transformations to rotations, which preserve the norm. To obtain a numerically more stable basis with simple static correlations, we therefore perform a $S(q)$ orthonormalization, where the matrix-valued static structure factor defines the inner product (and hence the metric). Let $T(\mathbf{q})$ be a lower-triangular matrix (e.g., from Cholesky or Gram-Schmidt) such that

$$\sum_{c=1}^s \sum_{d=1}^s S_{cd}^E(q) T_a^c(\mathbf{q}) T_b^d(\mathbf{q}) = \delta_{ab}. \quad (32)$$

We then define the final modes and the single change-of-basis matrix by

$$\tilde{\rho}_a(\mathbf{q}, t) = \sum_{b=1}^s \mu_b(\mathbf{q}, t) T_a^b(\mathbf{q}), \quad (L^{-1})_a^\alpha(\mathbf{q}) = \sum_{b=1}^s E_b^\alpha T_a^b(\mathbf{q}). \quad (33)$$

By construction, the static correlator in the mode basis is the identity,

$$\tilde{S}_{ab}(q) \equiv \frac{1}{N} \langle \tilde{\rho}_a(\mathbf{q}) | \tilde{\rho}_b(\mathbf{q}) \rangle = \delta_{ab}, \quad (34)$$

and the dynamical correlators transform as in Eq. (19) with $L(\mathbf{q}) = T^{-1}(\mathbf{q}) E^{-1}$. In particular, the first orthonormalized mode is proportional to the total density, $\tilde{\rho}_1(\mathbf{q}, t) \propto \sum_{\alpha} \rho_\alpha(\mathbf{q}, t)$, and therefore

$$\tilde{S}_{11}(q, t) = \frac{1}{N} \langle \tilde{\rho}_1(\mathbf{q}, t) | \tilde{\rho}_1(\mathbf{q}) \rangle = \frac{S(q, t)}{S(q)}, \quad (35)$$

i.e., it equals the normalized total intermediate scattering function. If desired, one can rescale the first mode by $\sqrt{S(q)}$ to make $\tilde{S}_{11}(q, t) = S(q, t)$ while keeping $\tilde{S}_{ab}(q) = \delta_{ab}$ for $a, b \geq 2$.

In the mode basis, the equations of motion retain their form (23) with the memory kernel (24). Empirically, the transformed correlator matrix $\tilde{S}(q, t)$ is dominated by its first diagonal element, while off-diagonal elements and higher modes are substantially smaller. This motivates a truncation (cutoff) at a modest number of modes. The truncation is safe in the sense that the Cauchy-Schwarz inequality bounds the off-diagonal elements by the diagonal ones, $|\tilde{S}_{ab}(q, t)|^2 \leq \tilde{S}_{aa}(q, 0) \tilde{S}_{bb}(q, 0) = 1$, and the complete monotonicity of partial correlators for Smoluchowski dynamics [38,39] supports the stability of the reduced dynamics. The quantitative performance of this truncation is assessed in Sec. V.

IV. NUMERICS

The influence of different shapes of size distributions on the glass transition has been studied previously [15,37,45]. To demonstrate that our method is not restricted by the choice of distribution, we apply the cutoff approximation to two conventional size distributions: the Schulz-Zimm distribution [46,47] and an inverse-cubic distribution.

For the Schulz-Zimm distribution we use

$$P(\sigma) = \frac{1}{\Gamma(z+1)} \left(\frac{z+1}{\bar{\sigma}} \right)^{z+1} \sigma^z \exp \left(-\frac{z+1}{\bar{\sigma}} \sigma \right), \quad (36)$$

where $\bar{\sigma}$ is the mean diameter and $z = 1/\delta^2 - 1$ is fixed by the polydispersity δ , given by the standard deviation of the size distribution normalized by $\bar{\sigma}$. For the inverse-cubic case, we take

$$P(\sigma) = \frac{A}{\sigma^3}, \quad \sigma \in [\sigma_{\min}, \sigma_{\max}], \quad (37)$$

with normalization constant $A = 2(\sigma_{\min}^{-2} - \sigma_{\max}^{-2})^{-1}$. The interval $[\sigma_{\min}, \sigma_{\max}]$ is chosen to match the target δ . Both distributions are plotted in Fig. 1 for $\delta = 0.1, 0.2, 0.3$.

Discretization into s components proceeds by binning $P(\sigma)$ into s equal-probability bins. The concentration of bin α is $x_\alpha = 1/s$, and its effective diameter is set to the bin average

$$\sigma_\alpha = \frac{1}{x_\alpha} \int_{\text{bin}\alpha} \sigma P(\sigma) d\sigma, \quad (38)$$

which preserves the first moment exactly under discretization. Following Ref. [37], we rescale the set $\{\sigma_\alpha\}$ to enforce

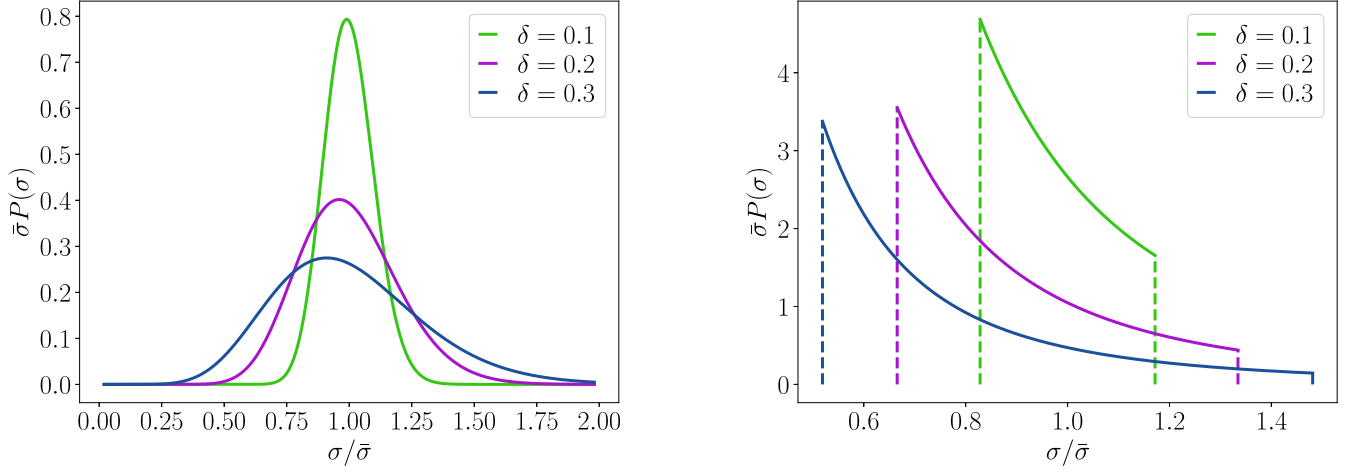


FIG. 1. Schulz-Zimm (left) and inverse-cubic distributions (right) for polydispersities $\delta = 0.1, 0.2, 0.3$.

$\bar{\sigma} = \sum_{\alpha} x_{\alpha} \sigma_{\alpha} = 1$. For the Schulz-Zimm distribution, δ directly sets $\{\sigma_{\alpha}\}$, while for the inverse-cubic distribution, δ determines $(\sigma_{\min}, \sigma_{\max})$ (and thus A) implicitly.

The partial static structure factors $S_{\alpha\beta}(q)$ are obtained by solving the multicomponent Ornstein-Zernike equation with the Percus-Yevick (PY) closure for additive hard spheres. In practice we use the known multicomponent PY solution [41,48]. The resulting $S_{\alpha\beta}(q)$ define $c^{\alpha\beta}(q)$ via Eq. (26). The packing fraction is calculated as follows:

$$\varphi = \frac{\pi}{6} n \sum_{\alpha=1}^s x_{\alpha} \sigma_{\alpha}^3. \quad (39)$$

For the short-time transport, we choose species-dependent diffusion coefficients proportional to the inverse particle size according to Stokes-Einstein scaling, i.e.,

$$D_{\alpha}^0 = D_0 \frac{\bar{\sigma}}{\sigma_{\alpha}}, \quad (40)$$

with D_0 as the reference diffusion coefficient for the average diameter $\bar{\sigma}$. Then $\bar{\sigma}^2/D_0$ sets the natural timescale of the system. The wave-vector grid uses $N_q = 100$ equidistant points on $q \in [q_{\min}, q_{\max}]$ with $q_{\min} = 0.2$ and $q_{\max} = 40.2$. The grid was constructed using these values because an increase in either N_q or $q_{\max}$ yields no quantitative differences. Time integration is performed on a standard logarithmically spaced grid, starting from a short initial step Δt_0 and doubling it repeatedly to cover several decades in t [37,49–51].

The matrices are first initialized in the species basis. We then construct the mode basis via $\mathbf{L}(q)$ as described in Sec. III. In the mode basis, a cutoff to m modes can be applied. If a cutoff is set, we evaluate only the upper-left $m \times m$ blocks of all transformed objects and propagate the reduced system. However, the transformed quantities, especially the vertex, are initialized fully and only afterwards reduced in size. The transformed static correlator is $\tilde{\mathbf{S}}(q) = \mathbf{I}$ by construction, which simplifies the initial conditions. If no cutoff is applied, we compute the full dynamics in the mode basis and, if needed, transform back to recover the partial ISFs.

Unless stated otherwise, we use $s = 10$ components and polydispersities $\delta \in \{0.1, 0.2, 0.3\}$ to span moderate to high polydispersities [44].

V. RESULTS

We assess the performance of the mode-basis cutoff scheme against the full multicomponent calculation. Section V A quantifies how the critical packing fraction depends on the number of modes, and Sec. V B presents the corresponding ISFs. For context, we also compare our complexity reduction (via the mode-basis cutoff) with a baseline simplification in which the size-polydisperse system is represented by a mixture with fewer components.

A. Nonergodicity parameter and critical packing fraction

The nonergodicity parameter distinguishes the glass from the liquid state at fixed packing fraction and thus determines the critical packing fraction. It is defined as the long-time limit of the ISF, $F(q) := \lim_{t \rightarrow \infty} S(q, t)$. In the mode basis it is obtained from

$$\tilde{\mathbf{F}}(q) = \tilde{\mathbf{S}}(q) - [\tilde{\mathbf{S}}^{-1}(q) + \tilde{\mathcal{F}}(q)]^{-1}, \quad (41)$$

where $\tilde{\mathcal{F}}(q)$ denotes bilinear functionals of the nonergodicity parameters [26]. The iterative determination of $\tilde{\mathbf{F}}(q)$ converges monotonically [39]. A nonzero entry in the non-ergodicity-parameter matrix signals a glass state. This criterion is equivalent in the mode and species bases. In the following, we examine how the critical packing fraction inferred from the nonergodicity parameter shifts with the cutoff.

Table I reports the critical packing fractions for the discretized Schulz-Zimm diameter sets [Eq. (36)] as a function of polydispersity and number of included modes m . As expected, the critical packing fraction increases with polydispersity because a larger size dispersion enhances an effective entropic attraction that stabilizes the liquid phase [52–55]. For large values of m , the critical packing fractions remain close to the ten-component reference. At low m , the accuracy depends sensitively on δ . For $\delta = 0.1$, retaining as few as three modes yields critical packing fractions within about 0.7% of

TABLE I. Critical packing fractions for different polydispersities and modes m for the Schulz-Zimm distribution.

Modes m	$\delta = 0.1$	$\delta = 0.2$	$\delta = 0.3$
10	0.51533	0.51736	0.52222
8	0.51535	0.51740	0.52254
6	0.51537	0.51824	0.52573
4	0.51614	0.52597	0.54327
3	0.51900	0.54027	0.56665
2	0.52786	0.55679	0.58135
1	0.55454	0.63006	/

the ten-mode result. For $\delta = 0.3$, retaining only four modes already yields a deviation of about 4%, and a single-mode truncation does not produce a physically reasonable critical packing fraction. Hence, $\delta = 0.3$ represents a sensible upper bound for reliable truncation at low mode numbers.

These trends reflect the expected dependence on size dispersion. When the discretized diameters are narrowly distributed (small δ), few collective modes capture most of the dynamics, and low cutoffs perform well. As δ increases, the broader spread of diameters requires more modes to maintain accuracy, and agreement between truncated and full (ten-component) results degrades at lower cutoffs.

The inverse-cubic distribution (Table II) shows the same overall behavior. For ten-component systems, cutoffs down to six modes reproduce the untruncated result very well. For smaller δ , acceptable accuracy persists for as low as three modes, while for larger δ , four to six modes are required.

B. Intermediate scattering function

The matrix-valued ISF fully characterizes the dynamics of the size-polydisperse system. Here, we analyze the impact of the cutoff on the total ISF, i.e., the first diagonal element of the transformed ISF matrix, $\tilde{S}_{11}(q, t) = S(q, t)/S(q)$.

We compare the total ISF across cutoff values using the distance to the glass transition, $\varepsilon = \varphi - \varphi_c$, to align states

TABLE II. Critical packing fractions for different polydispersities and modes m for the inverse-cubic distribution.

Modes m	$\delta = 0.1$	$\delta = 0.2$	$\delta = 0.3$
10	0.51529	0.51786	0.52517
8	0.51532	0.51788	0.52523
6	0.51534	0.51851	0.52653
4	0.51584	0.52433	0.53865
3	0.51753	0.53472	0.55926
2	0.52392	0.55028	0.56925
1	0.55380	0.61900	/

at comparable proximity to criticality [56]. We display ε in geometric progression, $\varepsilon = \pm 10^{-n}$ with $n = 1, 2, 3, 4$ in the liquid ($\varepsilon < 0$) and $n = 1, 2, 3$ in the glass ($\varepsilon > 0$). The choice of $\varepsilon = 0$ corresponds to setting $\varphi = \varphi_c$.

Figure 2 shows $S(q, t)/S(q)$ for both distributions at $\delta = 0.2$. For the Schulz-Zimm distribution, curves at the same $|\varepsilon|$ are very similar for $m \geq 3$, and essentially indistinguishable for large $|\varepsilon|$ (e.g., $\varepsilon = \pm 10^{-1}$). As $|\varepsilon|$ decreases, deviations emerge: truncation to $m = 1$ and $m = 2$ differ from the ten-component reference already far from criticality; truncation to $m = 3$ and $m = 4$ deviate only very close to the transition; and truncation to $m \geq 6$ remains very close to the untruncated result. The inverse-cubic case exhibits the same pattern: cutoffs below $m = 3$ perform poorly, $m = 3$ and $m = 4$ are reasonable except very near the transition, and $m \geq 6$ track the full result closely. For all cutoffs $m \geq 3$, the overall shape and time dependence closely resemble the ten-component reference.

Overall, a ten-component discretization can be well approximated by retaining a modest number of modes. Increasing the cutoff systematically improves agreement, while a cutoff of $m = 6$ already reduces the number of memory-kernel evaluations by about a factor of 20 and remains close to the full result. Some quantitative deviations are expected because MCT is sensitive to small input differences near the glass transition. Nevertheless, the cutoff approximation re-

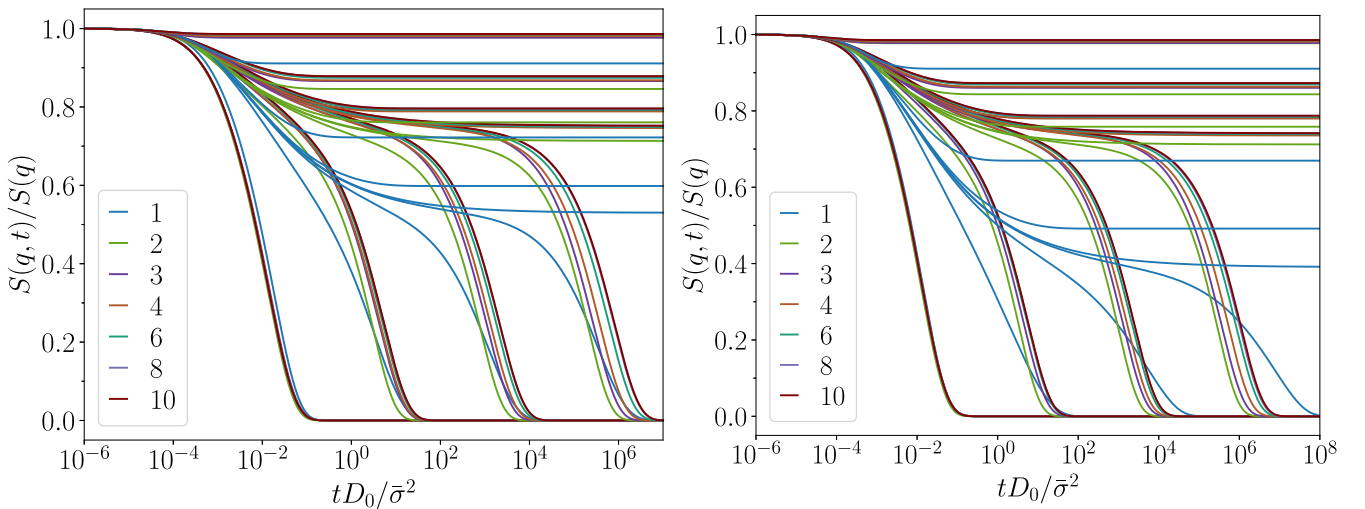


FIG. 2. Normalized total ISF in the mode basis for diameters generated according to the Schulz-Zimm (left) and inverse-cubic distributions (right) at $\delta = 0.2$ and $q\bar{\sigma} = 6.67$. Curves correspond to cutoffs to $m = 1, 2, 3, 4, 6, 8, 10$ and to distance parameters $\varepsilon = \pm 10^{-n}$ with $n = 1, 2, 3, 4$ for $\varepsilon < 0$, $\varphi = \varphi_c$ for $\varepsilon = 0$, and $n = 1, 2, 3$ for $\varepsilon > 0$ (left to right). Critical packing fractions are listed in Tables I and II.

TABLE III. Comparison of α -relaxation times τ_α for the Schulz-Zimm and inverse-cubic distributions at $q\bar{\sigma} = 6.67$, $\delta = 0.2$, and $\varepsilon = -10^{-4}$ in units of $\bar{\sigma}^2/D_0$ and in dependence of the modes m .

Modes m	Schulz-Zimm	Inverse-cubic
10	699289.4	901599.5
8	677299.2	840026.9
6	527765.6	690493.3
4	327654.5	450799.8
3	233096.5	305664.2
2	184718.0	247390.1
1	321057.4	5523946.4

covers the relevant dynamical information with substantially reduced cost, and the close agreement in shape for cutoffs to $m \geq 3$ suggests that a small retuning of φ can further align truncated and full curves. Although we show the total ISFs only at the first-neighbor peak, $q\bar{\sigma} = 6.67$, the same trends are observed for other values of q .

To quantify the differences, Table III compares α -relaxation times at $\delta = 0.2$ and $\varepsilon = -10^{-4}$. We define τ_α as the time at which the normalized total ISF drops to $1/e$ of its plateau at the transition. For the Schulz-Zimm distribution, a cutoff to $m = 8$ nearly reproduces the ten-component τ_α ; a cutoff to $m = 6$ yields about 75% of the reference, $m = 4$ about 50%, and $m = 3$ remains in the same decade but deviates strongly. Cutoffs to $m = 1$ and $m = 2$ are not sensible choices. Their curve shapes differ qualitatively from the ten-component reference, which is reflected in poor or misleading τ_α values (e.g., the apparent proximity of $m = 1$ is not meaningful given its dynamics).

The inverse-cubic results follow the same trend: a cutoff to $m = 8$ is close to the reference; a cutoff to $m = 6$ yields about 80%; a cutoff to $m = 4$ about 50%; and a cutoff to $m = 3$ about 30%. Again, cutoffs $m = 1$ and $m = 2$ are not comparable due to qualitatively different dynamics.

Although the total ISF is the most important quantity for discussing the glass transition, higher-order moments capture

additional features beyond the total ISF. In Fig. 3, we show the second moment $\tilde{S}_{22}(q, t)$ as a representative example, evaluated at the same distances from criticality and at the same critical packing fractions as previously discussed, for both distributions. Because at least the first two moments must be retained, the minimal truncation order is $m = 2$. For $m \geq 6$, the curves remain very close to the ten-component reference across all ε , similar to the total ISF. In contrast, $m = 3$ and $m = 4$ show more pronounced deviations than the total ISF, and $m = 2$ already deviates strongly far from the glass transition. Overall, the second-moment ISF exhibits the same truncation trends as the total ISF for both distributions and cutoffs with $m \geq 3$ providing reasonable agreement in curve shape with the ten-component reference. As expected, agreement deteriorates with increasing moment order, with higher-order moments requiring more modes for an accurate representation.

Finally, Fig. 4 contrasts two ways of reducing computational cost for the Schulz-Zimm distribution at $\delta = 0.2$: (i) starting from a ten-component mixture and applying the mode-basis cutoff to $m = 3$ or $m = 4$ modes (red), and (ii) representing the distribution by only $s = 3$ or $s = 4$ components (blue). Far from the glass transition, the cutoff curves are nearly indistinguishable from the ten-component reference when compared at the same ε . Closer to criticality, quantitative deviations increase, but the cutoff curves retain the characteristic shape and time dependence of the reference. In contrast, directly reducing the number of components introduces a nearly constant offset and yields dynamics that differ from the ten-component system already at early times. Given similar computational effort, the mode-basis cutoff more faithfully reproduces the shape and behavior of the full system while achieving a substantial reduction in complexity.

VI. SUMMARY AND OUTLOOK

We have shown that the multicomponent MCT equations are covariant under arbitrary changes of basis in species

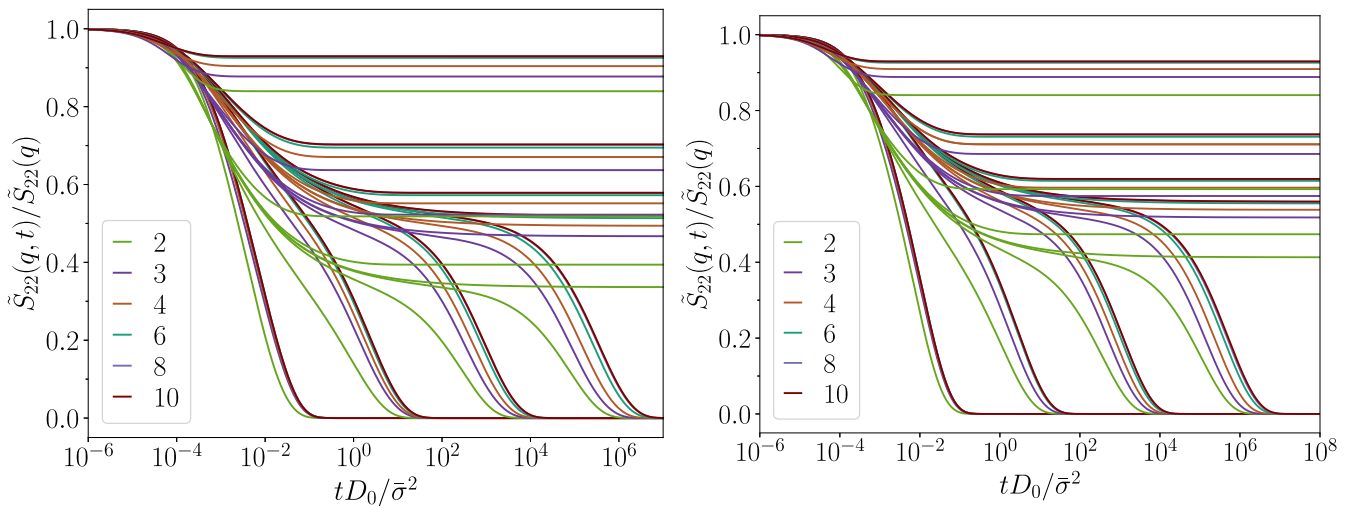


FIG. 3. Normalized second-moment ISF in the mode basis for diameters generated according to the Schulz-Zimm (left) and inverse-cubic distributions (right) at $\delta = 0.2$ and $q\bar{\sigma} = 6.67$. Curves correspond to cutoffs to $m = 1, 2, 3, 4, 6, 8, 10$ and to distance parameters $\varepsilon = \pm 10^{-n}$ with $n = 1, 2, 3, 4$ for $\varepsilon < 0$, $\varphi = \varphi_c$ for $\varepsilon = 0$, and $n = 1, 2, 3$ for $\varepsilon > 0$ (left to right). Critical packing fractions are listed in Tables I and II.

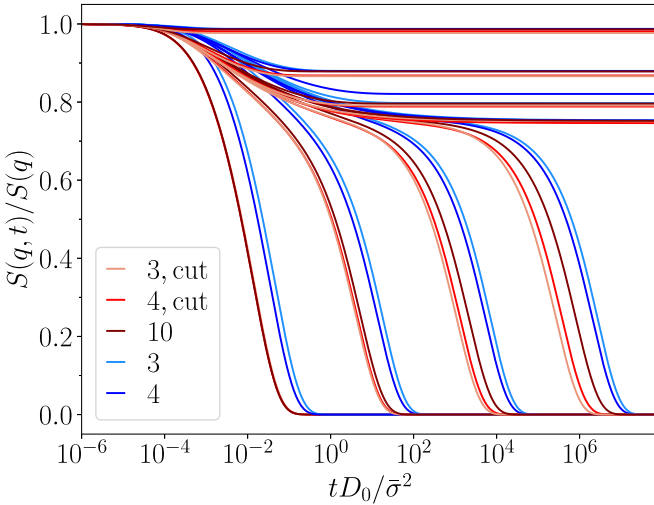


FIG. 4. Normalized total ISF for diameters generated according to the Schulz-Zimm distribution at $\delta = 0.2$ and $q\bar{\sigma} = 6.67$ with the cutoff approximation (red; starting from ten components and retaining three or four modes) and with a reduced number of mixture components (blue; discretized directly as three or four components). The brown curve is the ten-component reference.

space when the vertices are transformed consistently. This identifies the species basis as the natural frame for deriving the closure, while allowing a subsequent change to a problem-adapted basis without altering the structure of the equations. Building on this, we constructed a mode basis tailored to size polydispersity. Starting from a diameter-moment (Vandermonde) expansion and an $S(q)$ orthonormalization, we obtained a single, wave-vector-dependent transformation that renders the static correlator the identity in the mode basis. In this representation, the first mode is proportional to the total density, the second captures size-contrast concentration fluctuations, and higher modes encode successively higher diameter moments. Empirically, the transformed dynamical correlator matrix is dominated by its first diagonal element, which motivates a systematic truncation to a modest number of modes.

We validated the resulting mode-basis cutoff scheme for two sample distributions, the Schulz-Zimm and inverse-cubic diameter distributions, over a range of polydispersities. The cutoff accurately reproduces the critical packing fraction and the normalized total ISF over broad time windows, with performance governed by the number of retained modes and by the width of the size distribution. For moderate polydispersity, retaining as few as three to four modes already yields quantitative agreement near the transition and excellent agreement away from it; for higher polydispersity, six or more modes are typically sufficient. Compared with directly coarsening the mixture to fewer components, the mode-basis

cutoff better preserves the shape and time dependence of the ten-component reference at a matched distance from the glass transition. Since the computational cost scales as $O(s^6)$ per wave vector, reducing the number of components from ten to five yields a reduction by roughly a factor 60 while maintaining high accuracy.

Building on the covariance, the Newtonian case can be treated with virtually no modifications. Instead of transforming the diffusion matrix, one transforms the frequency matrix, which involves the species masses and the inverse structure factors. The closure and the bilinear memory kernel retain their form in the new basis.

The present implementation does not yet include the scattering-length weighted ISF. However, the transformation can be adapted such that the first mode aligns with the weight vector $[b^\alpha(q)]$, and consequently $\tilde{S}_{11}(q, t)$ would equal $S(q, t) = \sum_{\alpha=1}^s \sum_{\beta=1}^s b^\alpha(q) b^\beta(q) S_{\alpha\beta}(q, t)$. This modification preserves the covariance of the equations and the structure of the closure, and it should be straightforward to incorporate within the current framework.

Our strategy is not restricted to polydisperse hard spheres. All static input enters through the multicomponent structure factors, so any mixture for which $S_{\alpha\beta}(q)$ [or equivalently $c^{\alpha\beta}(q)$] is available can be treated in the same way. This includes systems with different pair interactions or thermodynamic state points, provided consistent static inputs are supplied. However, it is restricted to unimodal size distributions with a single, well-defined peak, specifically excluding multimodal distributions.

Finally, the mode construction could be further refined once the primary use cases are identified. Adaptive truncation and mild basis optimization, guided by error indicators for $\tilde{S}_{11}(q, t)$, can be pursued to reduce the number of retained modes without compromising accuracy, while keeping these refinements as a secondary layer atop the robust, covariance-based formulation.

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DATA AVAILABILITY

The data presented in this work and the program used to generate them are publicly available [57].

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