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Qualitatively different collective and single-particle dynamics in a supercooled liquid

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The equations of fluctuating nonlinear hydrodynamics for a two component mixture are obtained with a proper choice of slow variables which correspond to the conservation laws in the system. Using these nonlinear equations we construct the basic equations of the mode coupling theory (MCT) and consequent ergodic-nonergodic (ENE) transition in a binary mixture. The model is also analyzed in the one component limit of the mixture to study the dynamics of a tagged particle in the sea of identical particles. According to the existing MCT, dynamics of the single-particle correlation is slaved to that of the collective density fluctuations and, hence, both correlations freeze simultaneously at the ENE transition. We show here from a nonperturbative approach that at the ENE transition, characterized by the freezing of the long time limit of the dynamic correlation of collective density fluctuations to a nonzero value, the tagged-particle correlation still decays to zero. Our result implies that the point at which simulation or experimental data of the self-diffusion constant extrapolate to zero would not correspond to the ENE transition of simple MCT.

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I. INTRODUCTION

The self-consistent mode coupling theory (MCT) has been a useful tool for understanding slow dynamics in a dense liquid starting from the liquid side. The construction of this model involves a basic feedback mechanism affecting the transport properties of the liquid, arising from the coupling of slowly decaying density fluctuations in the supercooled state. The basic result of the model is that as the density of the liquid increases beyond a critical value, a dynamic transition from the ergodic liquid state to a nonergodic ideal glassy state occurs. The long time limit of the time correlation of density fluctuations is treated as an order parameter for this transition. This quantity, termed as the nonergodicity parameter (NEP), makes a discontinuous jump from being zero in the liquid state to a nonzero positive value at the ergodic-nonergodic (ENE) transition.

For understanding the mechanism of glass formation in liquids with simple interaction potentials like the hard sphere or Lennard-Jones type, computer simulation of a small number of particles moving under classical laws of motion has been a useful tool [1–3]. In such simulation studies often binary mixtures are the system of choice, since they can be tuned to avoid crystallization of the liquid and thus facilitate the study of the supercooled state [1]. For the two component systems, the self-consistent MCT with the prediction of an ENE transition has been studied by several authors [4–6] in the past. The approach adopted there is a straightforward generalization of the MCT for the one component case. In these works analysis of experimental and simulation data is made through schematic models or treating the various nonuniversal parameters in the theory as freely adjustable for data fitting. The model equations in these works predict the dynamic transition too prematurely even when the structural inputs for the MCT was taken from the simulations. This aspect of the mode coupling model for the binary fluid is indicated in the computer simulation results reported in Refs. [7,8] in which the authors simulated a binary Lennard-Jones system. In this paper we refer to these models of binary mixture as the "existing MCT."

In the current work we present a different formulation of MCT for a binary mixture by constructing the renormalized perturbation theory for the dynamics of the collective modes of a two component system. The collective modes represent the underlying conservation laws of mass and momentum and we use the equations of fluctuating nonlinear hydrodynamics (FNH) for describing their evolution in time. The basic conservation laws thus play a key role in the construction of the mode coupling model that we develop here. The self-consistent MCT is formulated using a Martin-Siggia-Rose (MSR) type field theory corresponding to the stochastic dynamical equations. Previous works [9–11] using similar techniques for one component systems have provided important insight in our understanding of MCT for such systems. At the simplest level irreversible dynamics of the slow modes is expressed using bare transport coefficients in the equations of fluctuating linear hydrodynamics [12]. The transport coefficients represent the role of short time or binary collision events in producing dissipation. The nonlinear couplings of the slow modes in the FNH equations represent the role of correlated motions of the particles at high density and give rise to renormalization of the bare frictional coefficients. The reversible part or the Euler terms in the fluctuating equations are obtained using the Poisson bracket relations between the microscopic variables. The nonlinearities in the dynamics which give rise to the feedback mechanism of the MCT and causes the ENE transition are present in this reversible part, namely the pressure term in the momentum conservation equation. In the present model formulation for the binary mixture we focus on the dynamic instability or the ENE transition as a first step. Within the self-consistent MCT, we consider the corrections to the transport coefficients using the mode coupling approximation of dominant density fluctuations and this is the key ingredient in producing a feedback to the transport properties. For the binary mixture, couplings to the concentration fluctuations also become equally relevant. In

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the MSR theory the renormalization of both the viscosity and the interdiffusion are expressed in a self-consistent form. The possible ENE transition, allowed by the model equations, is analyzed in terms of the solution of a set of integral equations for the NEPs. These equations are obtained from the long time limit of the corresponding correlation functions constructed from the MSR field theoretic model.

The equal time or structural correlations in a binary fluid are obtained with a proper free energy functional written in terms of the slow variables and is a required input for the study of the dynamics. It is obtained from the coarse graining of the microscopic Hamiltonian of the binary mixture. Following the method of Langer and Turski [13], the momentum density dependent part is obtained [14]. The so called interaction part of the free energy functional is taken in the standard form with expansion in terms of direct correlation functions [15]. In the present work we confine ourselves to a strictly Gaussian type free energy functional. The two-point direct correlation functions can be expressed in terms of static structure factors of the binary mixture through Ornstein-Zernike relations [16]. The thermodynamic properties of the fluid determined from the interaction potential of the particles thus enter the formulation of the dynamics. In the mode coupling model for the dynamics, computation of the mode coupling integrals appearing in the renormalized transport coefficients requires the static structure factor of the liquid as an input. In the one component case the Percus Yevick (PY) structure factor has been used mostly in similar situations. In case of the binary mixtures we use the extension of the PY models for a two component fluid by Lebowitz [17]. These structure factors are obtained as a function of the packing fraction η, size ratio α (of species 2 to that of species 1), and the relative abundance of the species 1 denoted by the variable x.

Using the field theoretic formulation for the dynamics of the collective modes for the binary system, we are able to consider the one component limit of the mixture by setting the size ratio and mass ratio of the two species to be unity. The process of self-diffusion can then be considered by taking the system as a mixture of a single (tagged) particle with $(N-1)$ particles. In the existing MCT, the time correlation $\phi(t)$ (say) of collective density fluctuations couples to time correlation of the single particle $\phi_s(t)$. Therefore, as the ϕ freezes at the ENE transition, so does ϕ_s, which is simply slaved to the former and, hence, the tagged-particle diffusion is zero at this point. In the present work we consider the implications of the ENE transition on the dynamics of a single particle in a sea of identical particles.

The paper is organized as follows. In the next section, we define the proper set of conserved densities for the two component system and obtain the equations of FNH for the slow variables. In Sec. III, we introduce the MSR field theory for treating the nonlinearities in the FNH equations and construction of the renormalized perturbation theory. Here we demonstrate how the theory can be renormalized in terms of the elements of the self-energy matrix defined with the so called Dyson equation. In Sec. IV we discuss the ENE transition and the resulting equations for the NEPs at the one loop order renormalization. In order to clearly indicate the difference of the present approach from existing MCT for binary systems, we also discuss here the approximations involved in obtaining the latter. In Sec. V, we consider the existing MCT model and discuss the approximations involved in reaching the same with the use of the MSR approach. In Sec. VI we demonstrate through a nonperturbative analysis developed in Ref. [18] that the single-particle dynamics decouples from the collective correlation's behavior near the ENE transition. In the final section we evaluate our results in the background of the existing MCT for binary systems.

II. GENERALIZED HYDRODYNAMICS FOR A BINARY MIXTURE

The dynamics of the many-particle system is studied in terms of the time evolution of a set of slow modes. The latter arise as a consequence of underlying conservation laws, broken symmetries [19,20], or a specific physical property of the system in consideration. The equations of motion of these microscopically conserved variables are obtained in terms of generalized Langevin equations. Using standard formulations [21], the Langevin equations for the coarse grained densities $\{\psi_i\}$ are obtained in the generalized form (we adopt the notation that the repeated indices are summed over),

$$\frac{\partial \psi_\alpha}{\partial t} = \left[Q_{\alpha\nu} - \Gamma^0_{\alpha\nu}\right]\frac{\delta F}{\delta \psi_\nu} + \zeta_\alpha, \tag{1}$$

where ζ_α denotes the thermal noise which is assumed to be Gaussian and white. Correlation of the noise is related through standard fluctuation-dissipation relations (FDRs) to the bare or short time transport matrix $\Gamma^0_{\alpha\sigma}$ and introduces the irreversible dynamics for the collective modes. $\Gamma^0_{\alpha\sigma}$ in Eq. (1) is related to the correlation of the thermal noise ζ_α with FDRs,

$$\langle \zeta_\alpha(t)\zeta_\sigma(t')\rangle = 2\beta^{-1}\Gamma^0_{\alpha\sigma}\delta(t-t'), \tag{2}$$

where $\beta = (k_B T)^{-1}$ is the inverse of temperature T times the Boltzmann constant k_B. $F[\psi]$ is identified as the free energy functional of the local densities $\{\psi_\alpha(\mathbf{x},t)\}$ and determines the equal time correlations or susceptibility matrix $\chi^{-1}_{\alpha\sigma}$, with $\alpha,\sigma \in$ the set of slow modes for the system. $F[\psi]$ is expressed in terms of the slow modes. Thus, equilibrium averages of the fields ψ_α at equal times are given by

$$\langle \psi_\alpha \psi_\sigma\rangle = \frac{\int D(\psi) e^{-\beta F[\psi]}\psi_\alpha\psi_\sigma}{\int D(\psi) e^{-\beta F[\psi]}}, \tag{3}$$

where $D(\psi)$ indicates a functional integral over the fields $\{\psi_\alpha\}$. The stationary solution of the Fokker-Planck equation corresponding to Langevin equations for fluctuating hydrodynamics is $e^{-\beta F}$. In the following we consider the set of equations for a binary mixture which forms the basis for the model of self-diffusion we consider here. For the binary mixture elements of the bare transport matrix include the viscosities and interdiffusion coefficients. $Q_{\alpha\nu} = \{\psi_\alpha,\psi_\nu\}$ in Eq. (1) is the Poisson bracket between the slow variables ψ_α and ψ_ν.

We consider here the FNH equations for a binary mixture of N_s identical particles of species s having mass m_s, diameter σ_s (for $s = 1,2$), respectively. We define $x_s = N_s/N$ and $n_s = N_s/V$ respectively as the concentration and the number density per unit volume of the particles of species s (for $s = 1,2$). The total number of particles is $N = N_1 + N_2$ and V is the volume of the mixture. For the binary system, we consider the following set of collective variables which are treated as slow due to the underlying microscopic conservation of the individual mass and sum of the total momentum of the two species, respectively. The individual mass densities ρ_s and the momentum densities $\mathbf{g}_s$ for the species s are respectively defined in terms of microscopic phase space variables as [22]

$$\rho_s(\mathbf{x},t) = m_s \sum_{i=1}^{N_s} \delta(\mathbf{x} - \mathbf{R}_s^i(t)), \tag{4}$$

$$\mathbf{g}_s(\mathbf{x},t) = \sum_{i=1}^{N_s} \mathbf{P}_s^i \delta(\mathbf{x} - \mathbf{R}_s^i(t)), \tag{5}$$

where m_1 and m_2 are the masses of particles of species 1 and 2, respectively. The phase space coordinates of position and momentum of the ith particle of the species s are denoted as $\{R_s^i(t), P_s^i(t)\}$. The individual coarse grained mass densities, respectively denoted as $\rho_s(\mathbf{x},t)$ are microscopically conserved. The individual momentum densities $\mathbf{g}_1$ and $\mathbf{g}_2$ are not conserved, but total momentum density defined as

$$\mathbf{g}(\mathbf{x},t) = \mathbf{g}_1(\mathbf{x},t) + \mathbf{g}_2(\mathbf{x},t) \tag{6}$$

is conserved. We work here with the following set of conserved variables: the total mass density $\rho(\mathbf{x},t)$, total momentum density $g(\mathbf{x},t)$, and the concentration variable $c(\mathbf{x},t)$ [12]. The mass and concentration densities are defined as follows:

$$\rho(\mathbf{x},t) = \rho_1(\mathbf{x},t) + \rho_2(\mathbf{x},t), \tag{7}$$

$$c(\mathbf{x},t) = x_2\rho_1(\mathbf{x},t) - x_1\rho_2(\mathbf{x},t). \tag{8}$$

We define the fluctuations of ρ and c, respectively as $\delta\rho = \rho - \rho_0$ and $\delta c = c$, since the average of c is zero when we consider the mass ratio of the constituent particles to be unity.

The generalized Langevin equation (1), leads to the equations of motion for the respective coarse grained densities $\psi_i \equiv \{\rho(\mathbf{x},t), \mathbf{g}(\mathbf{x},t), c(\mathbf{x},t)\}$ for a binary mixture. Following standard procedures [23], outlined in Appendix A we obtain

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{g} = 0, \tag{9}$$

$$\frac{\partial g_i}{\partial t} + \nabla_j \left[\frac{g_i g_j}{\rho}\right] + \rho \nabla_i \frac{\delta F_U}{\delta \rho} + c \nabla_i \frac{\delta F_U}{\delta c} + L_{ij}^0 \frac{g_j}{\rho} = \theta_i, \tag{10}$$

$$\frac{\partial c}{\partial t} + \nabla \cdot \left[c \frac{\mathbf{g}}{\rho}\right] + \gamma_0 \nabla^2 \frac{\delta F_U}{\delta c} = f_c. \tag{11}$$

F_U is the so called potential part of the free energy functional $F[\psi]$ introduced in the generalized Langevin equation (1). F is expressed as

$$F[\rho,\mathbf{g},c] = F_K[\rho,\mathbf{g}] + F_U[\rho,c]. \tag{12}$$

The kinetic part F_K (dependent on the current density $\mathbf{g}$) is computed from the microscopic Hamiltonian. Considering partition function of the system and following the method of Langer and Turski [13,14] we obtain

$$F_K[\rho,\mathbf{g}] = \int d\mathbf{x} \frac{\mathbf{g}^2(\mathbf{x})}{2\rho(\mathbf{x})}. \tag{13}$$

$F_U[\rho,c]$ is taken here as a quadratic functional of the fields ρ and c and is related to the structure of the liquid. This is expressed in terms of the corresponding direct correlation functions $\{c_{\rho\rho}, c_{\rho c}, c_{cc}\}$ defined in the Ornstein-Zernike relations. See Appendix A for details on the structure of the mixture.

The various elements of the bare transport matrix $\Gamma_{\alpha\sigma}^0$ which appear in the generalized Langevin equation (1) for the binary mixture are defined in Appendix A. Thus, $\Gamma_{g_i g_j}^0 \equiv L_{ij}^0$ represents the matrix of bare or short time viscosities, while $\Gamma_{cc}^0 \equiv \gamma_0 \nabla^2$ is linked to the bare interdiffusion coefficient for the mixture. These two dissipative coefficients are related to the correlation of the Gaussian noises respectively in Eqs. (10) and (11),

$$\langle f_c(\mathbf{x},t) f_c(\mathbf{x}',t')\rangle = 2\beta^{-1}\gamma_0 \nabla^2 \delta(\mathbf{x} - \mathbf{x}')\delta(t - t'), \tag{14}$$

$$\langle \theta_i(\mathbf{x},t)\theta_j(\mathbf{x}',t')\rangle = 2\beta^{-1} L_{ij}^0 \delta(\mathbf{x} - \mathbf{x}')\delta(t - t'), \tag{15}$$

$$\langle f_c(\mathbf{x},t)\theta_i(\mathbf{x}',t')\rangle = 0. \tag{16}$$

Here β^{-1} determines the strength of the thermal noise correlations. For an isotropic system the bare viscosity matrix L_{ij}^0 involves two independent coefficients,

$$L_{ij}^0 = -L_0 \nabla_i \nabla_j - \eta_0(\delta_{ij}\nabla^2 - \nabla_i \nabla_j), \tag{17}$$

where L_0 and η_0 respectively denote the bare or short time longitudinal and shear viscosities.

Equations (9)–(11) represent the dissipative dynamics of the slow modes in a binary mixture due to nonlinear coupling of the these modes. The mode coupling model for slow dynamics of a mixture follows from these equations. To focus on the role of various nonlinearities we note the following terms in the above set of equations. In Eq. (10) for the momentum density $\mathbf{g}$, the second, third, and the fourth terms on the left-hand side (LHS) represent various contributions from the reversible part of the dynamics. The second term represents a convective nonlinearity and ensures Galilean invariance of the equations, while the third and the fourth terms correspond to nonlinear dynamics. Even with a Gaussian free energy functional defined in Eq. (A6), these two terms give rise to a nonlinear coupling of ρ and c. In Eq. (11) for concentration c, the second term on the LHS represents reversible dynamics and is the only nonlinearity. The third term represents the dissipative part that corresponds to a diffusive mode. The only nonzero Poisson bracket of the concentration variable is $\{c, g_i\}$ and since the the functional derivative of F with g_i is g_i/ρ, the only possible coupling in the reversible part of the c equation is between c and g_i/ρ. This is an important point to note and will be useful when we consider the renormalization of the dynamics due to the nonlinearities. The role of $1/\rho$ nonlinearities in the FNH equations will be ignored in this work to primarily focus on the ENE transition in particular.

At the linear level Eqs. (9)–(11) of fluctuating hydrodynamics for the mixture involve the characteristic speeds c_0 and υ_0, which are respectively expressed in terms of equilibrium structure factors, $c_0^2 = \rho_0 \chi_{\rho\rho}^{-1}$ and $\upsilon_0^2 = \rho_0 \chi_{\rho c}^{-1}$. The dissipative equations of linearized dynamics also include the bare transport coefficients which are, respectively, the longitudinal viscosity L_0 and the interdiffusion coefficient

$\nu_0 = \gamma_0 \chi_{cc}^{-1}$. The effects of the nonlinearities in the above FNH equations are accounted through renormalization of the bare transport coefficients (L_0, ν_0) as well as the speeds (c_0, υ_0). In particular, corrections of L_0 and ν_0 due to the slowly decaying hydrodynamic modes give rise to a nonlinear feedback mechanism which is key to producing the slow dynamics of the MCT.

III. MARTIN-SIGGIA-ROSE FIELD THEORY

In this section we describe the computation of the correlation and response functions of the conserved slow modes which are the prime quantities in describing the dynamics of the binary mixture and possible ENE transition in the system. The consequences of the nonlinearities in the equations of motion, i.e., the generalized Langevin equations for the slow variables, are worked out by using graphical methods of field theory. In the present work the renormalized perturbation theory is developed in self-consistent form, which is particularly useful for the discussion of the mode coupling model and the consequent slow dynamics. We follow here closely the methodology developed in Ref. [10] using the standard approach of MSR field theory [24–28]. We describe the scheme briefly below, and for more technical details we refer the reader to Ref. [10]. The renormalized theory for the binary mixture dynamics is developed in terms of the correlation functions and response functions respectively given by

$$G_{\alpha\beta}(12) = \langle \psi_\alpha(1)\psi_\beta(2)\rangle, \tag{18}$$

$$G_{\alpha\hat{\beta}}(12) = \langle \psi_\alpha(1)\hat{\psi}_\beta(2)\rangle. \tag{19}$$

The number 1 refers here to both space and time coordinates (x_1,t_1) and so on. The averages denoted here by the angular brackets are functional integrals over all the fields weighted by $e^{-\mathcal{A}}$. The action $\mathcal{A}$ is a functional of the field variables $\{\psi_i\}$ and the corresponding conjugate hatted fields $\{\hat{\psi}_i\}$ introduced in the MSR filed theory. Using the equations of motion (9)–(11) for the set of slow modes $\{\rho,c,\mathbf{g}\}$, the MSR action functional is obtained in the Appendix B as given in Eq. (B7). The correlation and response functions in the MSR field theory, respectively given by Eqs. (18) and (19), are suitably organized in terms of their contributions from the Gaussian and non-Gaussian parts of the action functional $\mathcal{A}$. Using the polynomial expansions of the linear and nonlinear kernel terms in the equations of motion the action functional $\mathcal{A}$ in Eq. (B7) is put in a schematic form:

$$\mathcal{A}_U[\Psi,\hat{\Psi}] = \frac{1}{2}\sum_{1,2}\Psi(1)G_0^{-1}(12)\Psi(2) + \frac{1}{3}\sum_{1,2,3}V(123)\Psi(1)\Psi(2)\Psi(3) + \frac{1}{4}\sum_{1,2,3,4}V(1234)\Psi(1)\Psi(2)\Psi(3)\Psi(4) - \sum_{1}\Psi(1)U(1). \tag{20}$$

In the above expression the set of slow modes $\{\psi_\alpha\}$ are represented in terms of a vector field $\Psi(1)$ having the different fields as its components. The nonlinearities in the equations of motion (9)–(11) give rise to non-Gaussian terms in the action Eq. (20) involving products of three or more field variables. The corresponding vertex functions $V(123)$, etc. [see Eq. (20) for the MSR action], are defined to be symmetric under the exchange of the indices. The simplest level form of the correlation functions are zeroth order quantities denoted by G_0, corresponding to the the action which is only quadratic order in the fields, all higher order vertices being ignored. Keeping only the Gaussian terms in the action functional (B7), the matrix G_0^{-1} defined in Eq. (20) is obtained in the block form

$$G_0^{-1} = \begin{bmatrix} \bigcirc & \mathcal{B}_0^\dagger \\ \mathcal{B}_0 & \mathcal{C}_0 \end{bmatrix}, \tag{21}$$

where the elements of matrix $\mathcal{B}_0$ are provided in Table I and the matrix $\mathcal{B}_0^\dagger$ is the transpose and complex conjugate of the matrix $\mathcal{B}_0$. The matrix $\mathcal{C}_0$ is defined as

$$[\mathcal{C}_0]_{\hat{\mu}\hat{\nu}} = 2\beta^{-1}\delta_{\hat{\mu}\hat{\nu}}[\delta_{\hat{\mu}\hat{g}}L_0 + \delta_{\hat{\mu}\hat{c}}\gamma_0]. \tag{22}$$

TABLE I. Elements of matrix $[G_0^{-1}]_{\hat{\alpha}\beta}$ defined in terms of the matrix $\mathcal{B}_0$. $\nu_0' = \chi_{\rho c}^{-1}\gamma_0$ and $\nu_0 = \chi_{cc}^{-1}\gamma_0$.

	ρ	c	g
$\hat{\rho}$	ω	0	$-q$
$\hat{c}$	$iq^2\nu_0'$	$\omega + iq^2\nu_0$	0
$\hat{g}$	$-qc_0^2$	$-q\upsilon_0^2$	$\omega + iq^2L^0$

The $\bigcirc$ on the right-hand side (RHS) of Eq. (21) represents the null matrix with all its elements equal to zero. The role of the non-Gaussian terms is to renormalize the correlation functions of the Gaussian theory and is expressed in a perturbation series expansion in terms of the corresponding vertices. The diagrammatic methods of field theories are used for this purpose. In the following we use the FDRs (28)–(31) to obtain important conclusions on the renormalized theory from a nonperturbative approach.

A. Fluctuation-dissipation relations

We now demonstrate that the correlation and response functions defined in Eqs. (18) and (19) are related through a set of fluctuation-dissipation relations. The derivation of these FDRs are based on the symmetry of the MSR action under time reversal transformation [29]. In Appendix A we demonstrate that the MSR action (B7) remains invariant under the following time transformation rules of the field ψ_i and its hatted conjugate $\hat{\psi}_i$:

$$\psi_i(\mathbf{x},-t) \to \epsilon_i\psi_i(\mathbf{x},t),$$
$$\hat{\psi}_i(\mathbf{x},-t) \to -\epsilon_i\left[\hat{\psi}_i(\mathbf{x},t) - i\beta\frac{\delta F}{\delta\psi_i(\mathbf{x},t)}\right]. \tag{23}$$

Applying this symmetry corresponding to the field $\psi_i \equiv g_i$, we obtain

$$\begin{aligned} g_i(\mathbf{x},-t) &\to -g_i(\mathbf{x},t), \\ \hat{g}_i(\mathbf{x},-t) &\to \hat{g}_i(\mathbf{x},t) - i\beta v_i(\mathbf{x},t). \end{aligned} \tag{24}$$

We denote the functional derivative of the free energy functional F with the field $\psi(\mathbf{x},t)$ as

$$\zeta_\psi(\mathbf{x}) = \frac{\delta F}{\delta\psi(\mathbf{x})}, \tag{25}$$

so that $\zeta_{g_i} = [\delta F/\delta g_i(\mathbf{x},t)] = g_i(\mathbf{x},t)/\rho(\mathbf{x},t) = v_i(\mathbf{x},t)$. Applying the above transformation rules to the correlation of $\hat{g}_i(\mathbf{x}_1,-t_1)$ with a field $\varphi(\mathbf{x}_2,t_2)$, we obtain

$$\begin{aligned} \langle \hat{g}_i(\mathbf{x}_1,-t_1)\varphi(\mathbf{x}_2,t_2)\rangle = \langle \hat{g}_i(\mathbf{x}_1,t_1)\varphi(\mathbf{x}_2,t_2)\rangle \\ - i\beta\langle v_i(\mathbf{x}_1,t_1)\varphi(\mathbf{x}_2,t_2)\rangle. \end{aligned} \tag{26}$$

For $t_1 > t_2$, the LHS is zero due to causality and obtains

$$G_{v_i\varphi}(\mathbf{x},t) = -i\beta^{-1}G_{\hat{g}_i\varphi}(\mathbf{x},t), \tag{27}$$

where $\mathbf{x} = \mathbf{x}_1 - \mathbf{x}_2$ and $t = t_1 - t_2$. Since the response functions by definition are time retarded due to causality principle, for the spatially Fourier transformed correlation function from the corresponding response function we obtain

$$G_{v_i\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\mathrm{Im}G_{\hat{g}_i\varphi}(\mathbf{q},\omega). \tag{28}$$

The $\mathbf{v}$ field has been introduced in the formulation to deal with the $1/\rho$ nonlinearity in the equations of motion. In the case of a one component liquid the latter plays a crucial role [10] in cutting off the sharp ENE transition in which density correlation function freezes at a nonzero value in the long time limit. In the present work we ignore the ergodicity mechanisms to focus on the implications of the ENE transition, in particular, in the binary system. Thus, we ignore the role of the $1/\rho$ nonlinearity and treat $\mathbf{g}$ and $\mathbf{v}$ with the linear relation $\mathbf{g} = \rho_0\mathbf{v}$ and work with the set of fields $\{\mathbf{g},\rho,c\}$. With this approximation the FDR given by Eq. (28) reduces to

$$G_{g_i\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\rho_0\mathrm{Im}G_{\hat{g}_i\varphi}(\mathbf{q},\omega). \tag{29}$$

Applying the same symmetries respectively for $\psi = \rho$ and c, we obtain the following set of FDRs:

$$G_{\zeta_c\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\mathrm{Im}G_{\hat{c}\varphi}(\mathbf{q},\omega), \tag{30}$$

$$G_{\zeta_\rho\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\mathrm{Im}G_{\hat{\rho}\varphi}(\mathbf{q},\omega), \tag{31}$$

where ζ_ρ and ζ_c are as defined in Eq. (25).

B. Renormalization

The role of the non-Gaussian parts of the action $\mathcal{A}$ on the correlation functions are quantified in terms of the self-energy matrix Σ which shows up in the equation satisfied by the response functions and that satisfied by the correlation functions. The self-energy matrix Σ is defined through the Schwinger-Dyson equation

$$\mathbf{G}^{-1} = \mathbf{G_0}^{-1} - \Sigma, \tag{32}$$

where $\mathbf{G_0}$ represents the Gaussian counterpart of $\mathbf{G}$ obtained by keeping only up to quadratic terms in the action $\mathcal{A}$. The matrix equations represented by Eq. (32) are solved to obtain the corresponding correlation and response functions in MSR field theory. For example, from the set of equations denoted by (32) we obtain that the response functions satisfy

$$\left[\left(G_0^{-1}\right)_{\hat{\alpha}\mu}(13) - \Sigma_{\hat{\alpha}\mu}(13)\right]G_{\mu\hat{\beta}}(32) = \delta(12)\delta_{\hat{\alpha}\hat{\beta}}. \tag{33}$$

Using diagrammatic methods the self-energies $\Sigma_{\hat{\alpha}\mu}$ as well as $\Sigma_{\hat{\alpha}\hat{\mu}}$ are expressed in perturbation theory in terms of the two-point correlation and response functions. The block form of the inverse Green's function matrix, both in the zeroth order and in the fully nonlinear theory, have a symmetric structure in the parts representing the response functions. Since the fields ψ and $\hat{\psi}$ are real, it readily follows from the MSR action functional that

$$\mathcal{A}^*[\psi,\hat{\psi}] = \mathcal{A}[\psi,-\hat{\psi}], \tag{34}$$

and it is straightforward to show [10] that the response function $G_{\hat{\alpha}\beta}$ satisfies the relation

$$G_{\hat{\alpha}\beta}(\mathbf{q},\omega) = -G^*_{\beta\hat{\alpha}}(\mathbf{q},\omega). \tag{35}$$

From the matrix form (21) for the G_0^{-1} matrix it is obvious that this is satisfied at the zeroth order. From the Dyson equation (32) it therefore also follows that the self-energies satisfy the relation

$$\Sigma_{\hat{\alpha}\beta}(\mathbf{q},\omega) = -\Sigma^*_{\beta\hat{\alpha}}(\mathbf{q},\omega). \tag{36}$$

Analyzing the structure of the full Green's function matrix from the Dyson equation in Appendix B 2, the response part of $G_{\alpha\hat{\beta}}$ is obtained as

$$G_{\alpha\hat{\beta}} = \frac{N_{\alpha\hat{\beta}}}{\mathcal{D}}. \tag{37}$$

The various elements of matrix $N_{\alpha\hat{\beta}}$ for the case of a binary mixture are obtained in Appendix B 2. The denominator $\mathcal{D}$ on the RHS of Eq. (37) is obtained in Eq. (B12). The $N_{\hat{\alpha}\beta}$ matrix satisfies the relation $N_{\hat{\alpha}\beta}(\mathbf{q},\omega) = N^*_{\beta\hat{\alpha}}(\mathbf{q},\omega)$. The various renormalized transport coefficients appearing in the RHS of Eq. (37) are expressed in terms of the corresponding response self-energies as listed in Eqs. (B13)–(B15). The correlation functions of the physical, unhatted field variables are defined as

$$G_{\alpha\beta} = -\sum_{\mu\nu} G_{\alpha\hat{\mu}}\mathcal{C}_{\hat{\mu}\hat{\nu}}G_{\hat{\nu}\beta}, \tag{38}$$

where greek letter subscripts take values ρ,c and the longitudinal components of the vector field $\mathbf{g}$. The self-energy matrix $\mathcal{C}_{\hat{\mu}\hat{\nu}}$ is listed in Table II. The double-hatted self-energies $\Sigma_{\hat{\mu}\hat{\nu}}$ as well as $\mathcal{C}_{\hat{\mu}\hat{\nu}}$ both vanish if either index corresponds to $\hat{\rho}$, since there is no noise or nonlinearity in the continuity equation (9). Therefore, from the general structure of Eq. (38) of the correlation and that of Eq. (37) for response functions we obtain

$$G_{\alpha\beta} = \frac{1}{\mathcal{D}\mathcal{D}^*}\sum_{\mu\nu} N_{\alpha\hat{\mu}}\mathcal{C}_{\hat{\mu}\hat{\nu}}N_{\hat{\nu}\beta}. \tag{39}$$

TABLE II. Elements of matrix $\mathcal{C}_{\hat{\alpha}\hat{\beta}}$.

	$\hat{\rho}$	$\hat{c}$	$\hat{g}$
$\hat{\rho}$	0	0	0
$\hat{c}$	0	$2\beta^{-1}q^2\gamma_0 - \Sigma_{\hat{c}\hat{c}}$	$-\Sigma_{\hat{c}\hat{g}}$
$\hat{g}$	0	$-\Sigma_{\hat{g}\hat{c}}$	$2\beta^{-1}q^2L_0 - \Sigma_{\hat{g}\hat{g}}$

From the above expression it is clear that the renormalized correlation functions involve both response type self-energies ($\Sigma_{\psi\hat{\psi}}$) and correlation type self-energies ($\Sigma_{\hat{\psi}\hat{\psi}}$), the latter being present in the matrix $\mathcal{C}_{\hat{\psi}\hat{\psi}}$. In order to demonstrate that the renormalized correlation functions can be expressed in terms of a set of renormalized transport coefficients we therefore need to establish a relation between the corresponding set of response and correlation type self-energies renormalizing the same transport coefficient. Here the fluctuation dissipation relations (29)–(31) between correlation and the response functions of the MSR field theory prove very useful. We are able to do this at the nonperturbative level here but in the hydrodynamic limit of small wave-vectors and frequencies.

C. Nonperturbative analysis

We consider the FDRs (29)–(31) obtained in the previous section to link the correlation and response self-energies. Using Eqs. (35) and (38), respectively, for the response function $G_{\hat{g}_i\varphi}$ and correlation function $G_{g_i\varphi}$ in the FDRs (29) and (30), we obtain the following set of relations:

$$N_{g_i\hat{\alpha}}\mathcal{C}_{\hat{\alpha}\hat{\gamma}} = -i\beta^{-1}\rho_0\big(\mathcal{D}\delta_{\hat{g}_i\hat{\gamma}} + N^*_{\hat{g}_i\varphi}G^{-1}_{\varphi\hat{\gamma}}\big), \tag{40}$$

$$\big\{\chi^{-1}_{\rho c}N_{\rho\hat{\alpha}} + \chi^{-1}_{cc}N_{c\hat{\alpha}}\big\}\mathcal{C}_{\hat{\alpha}\hat{\gamma}} = -i\beta^{-1}\big(\mathcal{D}\delta_{\hat{c}\hat{\gamma}} + N^*_{\hat{c}\varphi}G^{-1}_{\varphi\hat{\gamma}}\big). \tag{41}$$

The results in Eqs. (40) and (41) are further analyzed in the Appendix B to obtain a set of relations between the response and correlation self-energies. In the hydrodynamic limit which corresponds to the small wave numbers (q) and small frequencies (ω) we obtain

$$\gamma_{\hat{g}\hat{g}} = 2\beta^{-1}\rho_0\gamma'_{\hat{g}g}, \tag{42}$$

$$\gamma_{\hat{c}\hat{c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}\rho}}{\chi^{-1}_{\rho c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}c}}{\chi^{-1}_{cc}}. \tag{43}$$

The quantities $\{\gamma_{\hat{\alpha}\beta},\gamma_{\hat{\alpha}\hat{\beta}}\}$, which appear in the above relations are coming from the leading order contributions to the corresponding self-energies $\{\Sigma_{\hat{\alpha}\beta},\Sigma_{\hat{\alpha}\hat{\beta}}\}$. These leading order wave-vector dependencies for the different self-energies are listed in Appendix B. Justifications for using the properties of the different vertex functions V's in the action (20) are given in Appendix B. For the off-diagonal elements of the $\mathcal{C}_{\hat{\alpha}\hat{\delta}}$ matrix we also obtain

$$\gamma_{\hat{c}\hat{g}} = 2\beta^{-1}\frac{\gamma'_{\hat{g}c}}{\chi^{-1}_{cc}} = 2\beta^{-1}\frac{\gamma'_{\hat{g}\rho}}{\chi^{-1}_{\rho c}}. \tag{44}$$

The above relations between the self-energies are important for the renormalizability of the theory in terms of redefined transport coefficients. The renormalized longitudinal viscosity coefficient $L(q,z)$ is obtained in the form

$$\begin{aligned} L(q,z) &= L_0(q) + i\gamma'_{\hat{g}g}(\mathbf{q},z) \\ &= L_0(q) + \beta\rho_0^{-1}\int_0^\infty dte^{izt}\gamma_{\hat{g}\hat{g}}(\mathbf{q},t). \end{aligned} \tag{45}$$

From Eq. (43), we see that in the hydrodynamic limit both $\gamma'_{\hat{c}\rho}$ and $\gamma'_{\hat{c}c}$ are related to the self-energy $\gamma_{\hat{c}\hat{c}}$. This has important implications in the renormalization of the two transport coefficients appearing in the correlation function matrix. From Eqs. (B14) and (B15) and using the definitions of $\gamma_{\hat{c}c}$ and $\gamma_{\hat{c}\rho}$, we obtain, respectively, the renormalized expressions for $\nu(q,\omega)$ and $\nu'(q,\omega)$ as

$$\nu(q,\omega) = \chi^{-1}_{cc}\gamma_0(q) + \gamma'_{\hat{c}c}(q,\omega), \tag{46}$$

$$\nu'(q,\omega) = \chi^{-1}_{\rho c}\gamma_0(q) + \gamma'_{\hat{c}\rho}(q,\omega). \tag{47}$$

The renormalized quantities ν and ν' are respectively expressed as $\chi^{-1}_{cc}\gamma$ and $\chi^{-1}_{\rho c}\gamma$ in terms of a single renormalized quantity whose Laplace transform is defined as

$$\gamma(\mathbf{q},z) = \gamma_0(\mathbf{q}) + \beta\int_0^\infty dte^{izt}\gamma_{\hat{c}\hat{c}}(\mathbf{q},t), \tag{48}$$

involving the self-energy $\Sigma_{\hat{c}\hat{c}}$. To summarize, in hydrodynamic limit, the correlation and the response functions in the fully nonlinear theory are obtained in terms of the renormalized transport coefficients. The renormalization of the thermodynamic quantities c_0^2 and υ_0^2 follows from Eqs. (B16) and (B17). In Appendix B we demonstrate that for the Gaussian free energy considered in the present work these corrections are higher orders in q and vanish in the hydrodynamic limit.

IV. THE ERGODIC-NONERGODIC TRANSITION

We have identified above the respective self-energy matrix elements which contribute to the renormalized transport coefficients. The correlation and response functions of the fully nonlinear theory are expressed in terms of these renormalized quantities. The next step in this is to express these self-energies in terms of the correlation functions. This gives rise to a self-consistent scheme in which correlation functions satisfy nonlinear equations involving memory functions which are expressed in terms of the correlation functions themselves. This essentially constitutes the feedback mechanism of MCT and has been used extensively for understanding the slow dynamics in a dense supercooled liquid. As the density or the packing fraction of the system is increased, there is a critical density at which the density correlation function does not decay to zero in the long time limit and this is defined as an ENE transition. In the present case of the binary mixture, the transition is characterized by freezing of the correlations of ρ and c. The ENE transition in the mixture is characterized by correlation functions having a nonergodic solution, i.e., the long time limit of the different elements of the matrix of correlation functions $G_{\sigma\mu}(q,t)$, where $\sigma,\mu \in \{\rho,c\}$ are nonzero for all wave number q. This is equivalent to having the corresponding Laplace transform $G_{\sigma\mu}(q,z) \sim f_{\sigma\mu}(q)/z$ developing a pole at $z = 0$. Equivalently, the Fourier transform behaves like $G_{\sigma\mu}(q,\omega) \sim 2\pi\delta(\omega)f_{\sigma\mu}(q)$. In the following, the liquid is considered to have an ENE transition when a set of nonzero values is obtained for the $f_{\sigma\mu}(q)$'s, which are therefore called the NEPs.

Using the FDRs given by Eqs. (30) and (31), the expressions for the Fourier transform of the correlation functions are obtained in terms of the Laplace transforms of the correlation functions,

$$G_{\alpha\varphi}(q,z) = \frac{1}{\mathcal{D}}\sum_\nu N_{\alpha\hat{\nu}}(q,z)\chi_{\nu\varphi}. \tag{49}$$

The repeated index ν in Eq. (49) is summed over the set $\{\rho,c\}$. The above equation is rearranged to a form

$$\mathcal{D}\sum_{\alpha} N^{-1}_{\hat{\mu}\alpha}(q,z)G_{\alpha\varphi}(q,z)=\chi_{\mu\varphi}(q,z). \tag{50}$$

To sort out the dependence on structure and the dynamics, we define normalized correlation functions $\phi_{\alpha\sigma}$ as

$$\phi_{\alpha\sigma}(q,t)=\frac{G_{\alpha\sigma}(q,t)}{\sqrt{\chi_{\alpha\alpha}\chi_{\sigma\sigma}}}. \tag{51}$$

The corresponding NEP is then defined as the long time limit of the correlation functions $\phi_{\alpha\sigma}$; i.e., the NEPs $f_{\alpha\sigma}$ are obtained as

$$f_{\alpha\sigma}(q)=\lim_{t\to\infty}\phi_{\alpha\sigma}(q,t)=\lim_{z\to 0}z\phi_{\alpha\sigma}(q,z). \tag{52}$$

Taking the long time limit of the equations given by (49), we get the following equations for the NEPs:

$$\frac{f_{\alpha\sigma}(q)}{1-f_{\alpha\sigma}(q)}=\frac{a_{\alpha\sigma}L(q)}{\tilde{c}_0{}^2+(1-a_{\alpha\sigma})L(q)}. \tag{53}$$

The $a_{\alpha\sigma}$'s are obtained in terms of static correlations, $a_{\rho\rho}=1$, $a_{\rho c}=\chi$, and $a_{cc}=\chi^2$, where $\chi(k)=\chi_{\rho c}(k)/\sqrt{\chi_{\rho\rho}(k)\chi_{cc}(k)}$. $a_{\alpha\sigma}$ is symmetric in ρ and c. The quantity $\tilde{c}_0$ is obtained in terms of the sound speed c_0 as

$$\tilde{c}_0^2=c_0^2+v_0^2(\chi_{\rho c}/\chi_{\rho\rho}). \tag{54}$$

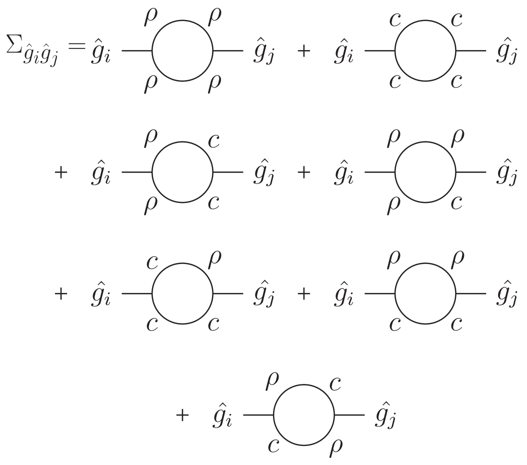

FIG. 1. One loop diagrams for $\Sigma_{\hat{g}_i\hat{g}_j}$ with vertices involving nonlinear couplings of density fluctuations.

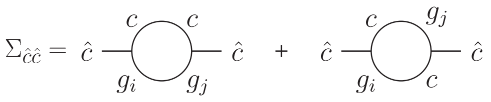

FIG. 2. One loop contributions to $\Sigma_{\hat{c}\hat{c}}$.

The function $L(q)$ in the RHS of Eq. (53) is the long time limit of the renormalized memory function $L(q,z)$ of the generalized viscosity. The memory function $L(q,z)$ involves a factor q^2 following from conservation laws. The memory functions for $L(q,t)$ as a nonlinear functional of the $f_{\alpha\sigma}$'s. In the standard MCT approach [10,30–32] the integral Eqs. (53) are closed by treating $L(q)$ as a nonlinear functional of the $f_{\alpha\sigma}$. This is determined by taking the long time limit of the corresponding self energies $\Sigma_{\hat{g}\hat{g}}$. Using the vertex functions appearing in the MSR action functional (B7) we show in the Appendix B 2 that the relevant diagrams (shown in Fig. 1) involving the slowly decaying correlations of ρ and c obtain the following one-loop contribution for $L(q,t)$,

$$L(q,t)=\frac{\beta^{-1}}{2\rho_0 q^2}\int\frac{d\mathbf{k}}{(2\pi)^3}V_{\hat{g}\sigma\sigma'}(\mathbf{q},\mathbf{k},\mathbf{k_1})V^{*}_{\hat{g}\mu\mu'}(\mathbf{q},\mathbf{k},\mathbf{k_1})$$
$$\times\,\varepsilon_{\sigma\mu}(k)\varepsilon_{\sigma'\mu'}(k_1)G_{\sigma\mu}(k,t)G_{\sigma'\mu'}(k_1,t), \tag{55}$$

where the repeated indices σ, μ, σ', and μ' are summed over the set $\{\rho,c\}$. The vertex function $V_{\hat{g}\sigma\mu}$ is given by

$$V_{\hat{g}\sigma\mu}(\mathbf{q},\mathbf{k},\mathbf{k_1})=[\hat{\mathbf{q}}.\mathbf{k}c_{\sigma\mu}(k)+\hat{\mathbf{q}}.\mathbf{k}_1c_{\sigma\mu}(k_1)], \tag{56}$$

and $\mathbf{k}_1=\mathbf{q}-\mathbf{k}$. $\varepsilon_{\sigma\mu}(k)$ is respectively equal to 1 or $\chi(k)$ for $\sigma=\mu$ and otherwise. The corresponding renormalization to the diffusion memory kernel $\gamma(q,t)$ is obtained from the self-energy element $\Sigma_{\hat{c}\hat{c}}$. The relevant one loop diagrams (shown in Fig. 2) involving the slowly decaying correlations of ρ and c obtain the following one loop contribution:

$$\gamma(q,t)=\frac{2}{\rho_0^2}\int\frac{d\mathbf{k}}{(2\pi)^3}\left[\frac{1}{k^2}\dot{G}_{cc}(k_1,t)\dot{G}_{\rho\rho}(k,t)\right.$$
$$\left.+\frac{1}{kk_1}\chi(k_1)\chi(k)\dot{G}_{\rho c}(k_1,t)\dot{G}_{\rho c}(k,t)\right]. \tag{57}$$

The dot implies a derivative with respect to time in the above equation. The memory kernel renormalizing the diffusion coefficient consists of the derivatives of the correlation functions and thus vanishes in the long time limit. The equations (53) obtained for the ENE transition in the mixture are same as obtained by Harbola and Das [14] for a binary system. The NEPs show a strong dependence on the size ratio $\alpha=\sigma_2/\sigma_1$, mass ratio $\kappa=m_2/m_1$, and concentration of the species x_1. The value of the total packing fraction $\eta=\pi(n_1\sigma_1^3+n_2\sigma_2^3)/6$ at the ENE transition point is the critical packing fraction and is denoted as η_c. We display in Fig. 3 how η_c, obtained from the

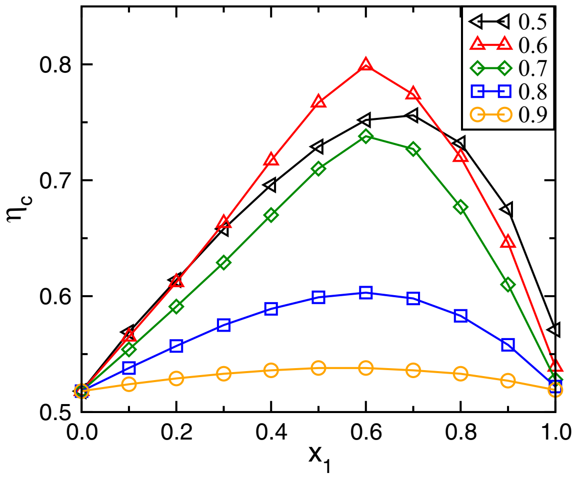

FIG. 3. (Color online) The critical packing fraction η_c (see text for definition) for the binary mixture as obtained from the solution of Eq. (53) vs concentration x_1 of smaller sized particles for different size ratios α, defined here as the ratio of smaller to bigger sized particles (having same mass) of the mixture.

solution of Eq. (53), changes with concentration x_1 of species 1 for different values of the size ratio α of the particles (with mass ratio $\kappa = 1$).

V. COMPARISON WITH THE EXISTING MCT MODEL

In this section we discuss a related model of MCT for a binary mixture which has been used extensively earlier in the literature and discuss its relevance in comparison to the present work. We briefly indicate how the method which we have used above for obtaining our model can also obtain the existing model and clarify the unphysical approximations made in this case. The existing version of the MCT [4] for binary systems is reproduced using the present method with a different choice for the set of slow variables. Here, in addition to the individual conserved densities $\{\rho_1,\rho_2\}$, the momentum densities of each component are treated as two *separate* slow variables. The microscopic definitions for the mass and momentum densities are given in Eqs. (4) and (5). The reversible parts of the corresponding equations for the slow modes are obtained by using the Poisson bracket relations among these four densities. However, assuming widely separated time scales, stochastic equations are written for each of the momentum densities $\mathbf{g}_s$ ($s = 1,2$). Using the same driving free energy functional F as in the Sec. II and further illustrated in Appendix A, the generalized Langevin equation corresponding to the four "slow" variables $\{\rho_s,\mathbf{g}_s\}$ for $s = 1,2$ are obtained [14]:

$$\frac{\partial \rho_s}{\partial t} + \nabla \cdot \mathbf{g}_s = 0, \tag{58}$$

$$\frac{\partial g_{is}}{\partial t} + \nabla_j \frac{g_{is} g_{js}}{\rho_s} + \rho_s \nabla_i \frac{\delta F_U}{\delta \rho_s} + L_{ij}^{ss'} \frac{\delta F}{\delta g_{js'}} = \xi_{is}. \tag{59}$$

In writing Eq. (58), the self- and interdiffusion of the two species in the density equations have been ignored. This is equivalent to writing $\mathbf{g}_s = (\rho_s/\rho)\mathbf{g}$. The thermal noise ξ_s in the equations for the momentum density $\mathbf{g}_s$ follows the FDR to the bare transport coefficients,

$$\langle \xi_{is}(\mathbf{x},t)\xi_{js'}(\mathbf{x}',t')\rangle = 2k_B T L_{ij}^{ss'} \delta(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{60}$$

Renormalization of the bare transport coefficients, as a result of the nonlinearities in the equations for the momentum currents is computed within the self-consistent mode coupling approximations of dominant density fluctuations. To the one loop order the contributions to the longitudinal component of the various elements of the transport coefficient matrix $\mathcal{L}_{ss'}$ are obtained as

$$\tilde{\mathcal{L}}_{ss'}(q,t) = \frac{n_0}{2n_s n_{s'}} \int \frac{d^3k}{(2\pi)^3} \sum_{\mu,\sigma,\mu',\sigma'} V_{s\mu\sigma}(\mathbf{q},\mathbf{k},\mathbf{k_1}) \times V_{s'\mu'\sigma'}(\mathbf{q},\mathbf{k},\mathbf{k_1}) G_{\mu\mu'}(k,t) G_{\sigma\sigma'}(k_1,t), \tag{61}$$

where $\mathbf{k}_1 = \mathbf{q} - \mathbf{k}$ and $n_0 (= n_1 + n_2)$ is the total number density with n_1 and n_2 being the individual number densities of species 1 and 2, respectively. The expression for the vertex function $V_{s\mu\sigma}$ is given by

$$V_{s\mu\sigma}(q,k) = \frac{n}{m_\mu m_\sigma}[\hat{\mathbf{q}} \cdot \mathbf{k}\delta_{s\sigma} c_{s\mu}(k) + \hat{\mathbf{q}} \cdot \mathbf{k}_1 \delta_{s\mu} c_{s\sigma}(k_1)]. \tag{62}$$

Starting from the above set of equations of motion, it is straightforward to obtain (using the MSR theory outlined in Sec. III) a set of nonlinear integro-differential equations or the so called memory function equations for the elements of the correlation function matrix $G_{ss'}$. The location of the ENE transition in the model is obtained by considering the long time limit of the equations for the time evolution of the correlation functions. The dynamical transition point in the previous version of MCT model is located by using a matrix equation similar to Eq. (53),

$$F(q) = \frac{1}{q^2}\mathcal{S}(q)\mathcal{L}(q)[\mathcal{S}(q) - F(q)], \tag{63}$$

where the matrix related to the structure is defined as $\mathcal{S}(q) = \sqrt{x_i x_j} S_{ij}$ and $\mathcal{L}_{ss'}(q)$ is the long time limit of the memory function $\mathcal{L}_{ss'}(q,t)$. These integral equations for the NEPs are similar to the equations we obtain in our model discussed in the earlier section. The actual form of the integral equations in the respective cases are determined from the wave-vector dependent structure factors or the equilibrium correlation functions which are used as an input in the model. These structural inputs are determined by the driving free energy functional F for the system. In this respect it is useful to note that *same free energy functional or wave-vector dependence* of the equilibrium correlations are used here for all the models. In the present model the ENE transitions corresponding to various choices for the thermodynamic parameters for mixture occur at higher packing fraction values than that predicted from Eqs. (63). This also agrees well with the results seen in the computer simulations on binary systems [7,8]. Details of such differences between the two types of mode coupling models have already been reported in Ref. [14].

In the present section we focus on the limiting case in which the previous MCT models [4] agree with predictions of our work with respect to the location of the ENE transition. It is clear from the deductions presented above that the primary difference between the two models come from the treatment of the momentum densities for the two different species of the mixture. The individual momentum densities $\{\mathbf{g}_1,\mathbf{g}_2\}$ are not conserved variables and there is no physical basis in assuming a separation of slow and fast time scales in their dynamics. However, the existing mode coupling model is obtained using separate Langevin equations for each of the momentum densities $\{\mathbf{g}_1,\mathbf{g}_2\}$. On the other hand, the total density $\mathbf{g} = \mathbf{g}_1 + \mathbf{g}_2$ is a conserved mode and has been treated as a slow mode in our model. For the Brownian particle, it is appropriate to write a Langevin equation for the momentum density of the single particle with high inertia. For the coarse grained tagged particle, the momentum density $\mathbf{g}^B(\mathbf{x},t)$ and the corresponding mass density $\rho^B(\mathbf{x},t)$ are related by the continuity equation,

$$\frac{\partial \rho^B}{\partial t} + \nabla \cdot \mathbf{g^B} = 0. \tag{64}$$

For tagged-particle momentum density $\mathbf{g}^B(\mathbf{x},t)$, the following equation is written

$$\frac{\partial g_i^B}{\partial t} + \rho^B \nabla_i \frac{\delta F}{\delta \rho^B} + \int d\mathbf{x}' \Gamma_0^B \frac{\delta F}{\delta g_i^B} = f_i^B. \tag{65}$$

The bare friction coefficient Γ_0^B is related to the noise $\mathbf{f}^B$ through the usual FDR. For the collective density ρ and

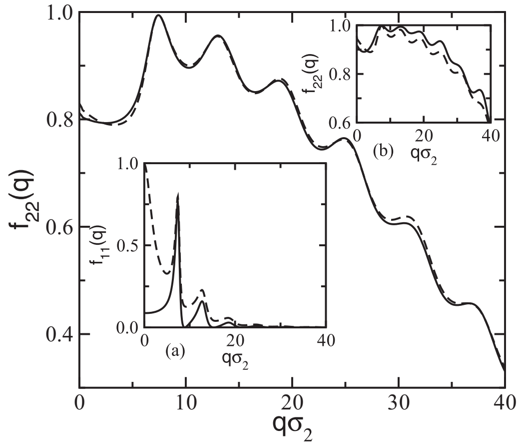

FIG. 4. Comparison between existing MCT model (mass ratio independent) and the present model in the Brownian limit of large mass ratio $\kappa = m_2/m_1 = 10^4$. For a mixture with size ratio $\alpha = \sigma_2/\sigma_1 = 10^2$, packing fraction $\eta = 0.6$, and for $x_2 = 0.1$, the NEPs $f_{22}(q)$ (main figure) and $f_{11}(q)$ (inset a) vs $q\sigma_2$. Inset (b) is the same as main figure for $x_2 = 0.01$. For all three figures, solid lines are results from Eq. (53) of present work and dashed lines are from Eqs. (63) of Ref. [4] of existing MCT.

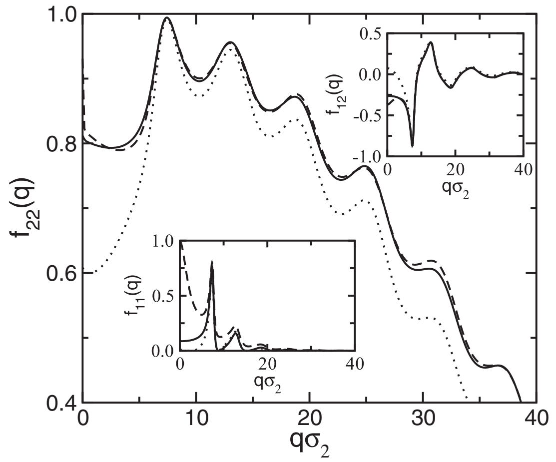

FIG. 5. Comparison between existing MCT model and the present model in the Brownian limit of the large mass ratio. Results are for the same mixture as in Fig. 4 with size ratio $\alpha = \sigma_2/\sigma_1 = 10^2$, packing fraction $\eta = 0.6$, and for $x_2 = 0.1$. The NEPs $f_{22}(q)$ (main figure) and $f_{11}(q)$ (lower inset) and $f_{12}(q)$ (upper inset) vs $q\sigma_2$. For all three figures, results from Eq. (53) of present work for mass ratio $\kappa = m_2/m_1 = 10^6$ (solid line), $= 10^2$ (dotted line), and from Eqs. (63) of Ref. [4] of existing MCT (dashed line).

current $\mathbf{g}$, the corresponding equations of motion follow from microscopic conservation laws; it is not so for $\mathbf{g}^B$. In this case the total momentum density $\mathbf{g}$ is approximated well by the rest of the "mixture." Therefore, *only* for the case of a Brownian particle can the set of four equations for the conserved densities (58) and (59) can be identified, respectively, with the set (9) and (10) and set (64) and (65) in our present model. Hence, in this case the results obtained in the existing MCT model comes close to the present formulation which keeps a proper account of the conservation laws. The mass ratio dependence of the ENE transition with the NEP Eqs. (53) and the wave-vector dependence of the corresponding NEPs has been reported in detail in an earlier work [33]. In the present section we demonstrate this equivalence of the two MCT models for the case of a single Brownian particle (large inertia) in a simple liquid being treated as a mixture. Using a very large mass ratio and very small concentration of the heavier particle, i.e., $\kappa \rightarrow$ large and $x \rightarrow$ small (signifying the Brownian limit), the results for the NEP in the present model for most wave-vector values are very close to the predictions of the existing model. For comparison of the two models, it is convenient to consider the nonergodicity parameters defined in terms of the corresponding species. The quantity $f_{ss'}$ is same as defined in Eqs. (51) and (52), but with $\{\alpha,\sigma\} \in s,s'$, for $s,s' = 1,2$. This is shown in Fig. 4. In the inset where f_{11} is shown, the $q \rightarrow 0$ behaviors of the two models are very different. To see this we need to take into account the behavior of the tagged-particle correlation ϕ_s in the two models. Taking the form of the Laplace transform $\phi_s(q,z)$ in the form of a diffusive pole with generalized diffusion coefficient $D_s(q,z)$, the long time limit of the correlation function or the so called NEP f_s is obtained as

$$\frac{f_s(q)}{1 - f_s(q)} = \frac{z}{iq^2 D_s(q,z)}. \tag{66}$$

Since for the existing MCT models, self-diffusion constant vanishes in the small frequency limit as $D_s \sim z$, we note from Eq. (66) that the NEP $f_s(q)$ gets pinned at the value 1 in the same limit. However, in our model D_s is finite in the $z \rightarrow 0$ limit, i.e., z/D_s for a fixed q is vanishing in this limit. The relation (66) implies vanishing of the NEP f_s for small wave numbers. For the existing MCT model, the predictions for the dynamics are, in fact, independent of mass ratio. In Fig. 5 we show another comparison of the two models at the same packing fraction $\eta = 0.6$ for a mixture having size ratio $\alpha = 10^2$ and $x_2 = 0.1$. The two mass ratios used here are $\kappa = 10^2$ and 10^4, respectively. Therefore, though in the Brownian limit the two MCT models are approximately matching, differences with our model show up at small q due to role of the conservation laws. For the other component f_{22}, both models essentially represent the collective correlations for a homogeneous liquid and, hence, they agree. However positivity of self-diffusion for interacting Brownian particles with hard core has been rigorously demonstrated [34].

VI. SINGLE-PARTICLE DYNAMICS

We now consider the implications of the MCT developed here on the dynamics of a tagged particle in a sea of identical particles. In this nonperturbative analysis we follow a method developed recently in Ref. [18] to establish, with the use of the available fluctuation dissipation relations in the MSR field theory, the long time behavior of time correlation functions. This was developed for analyzing the asymptotic dynamics of the correlation functions for the collective variables in a one component fluid. Here we apply this method for the binary mixture to prove an ENE transition beyond a critical density. Furthermore, the $1/\rho$ nonlinearities in the dynamics are ignored in this case. We then consider the model in the so called one component limit (to be explained below) to study the nature of a tagged-particle motion in a homogeneous liquid. This analysis demonstrates the decoupling between the collective and single-particle dynamics in a dense liquid.

A. The ENE transition in the mixture

We consider Eq. (38) for the correlation function for the MSR theory outlined above for a binary system. The ENE transition is characterized by the density and concentration correlation functions $\{G_{\rho\rho}(\omega), G_{\rho c}(\omega), G_{cc}(\omega)\}$, each developing a δ-function contribution. Using the one loop results (55) and (57), respectively, for the corresponding memory functions, we make the following observations.

(A) The generalized transport coefficient $L(\omega)$, which is the Laplace transform of $L(\mathbf{q},t)$ defined in Eq. (55), involves the correlation of the ρ and c. Hence, at the ENE transition, $L(\omega)$ has a singular part with $\delta(\omega)$ contribution. This conforms to the physics of the viscosity of the mixture diverging in the ideal glass phase. Equivalently, the self-energy $\Sigma_{\hat{g}\hat{g}}$ blows up at small frequencies and hence is written with a general nonperturbative expression:

$$\Sigma_{\hat{g}\hat{g}} = -A\delta(\omega) + \Sigma^R_{\hat{g}\hat{g}}. \tag{67}$$

The second term on the RHS represents parts of the self-energy contribution which are regular in the $\omega \to 0$ limit. In writing the above expression wave-vector dependence in the model is not ignored but suppressed to keep the notation simple.

(B) From Eq. (57), since it follows that the renormalization of the $\nu(q,z)$ involves only derivatives of the correlations of ρ and c, it has no singular contribution $[\sim\delta(\omega)]$ of $\nu(\omega)$ or $\nu'(\omega)$ in the small ω limit.

To test compatibility with the Dyson equation corresponding to the MSR action (B7), we substitute Eq. (67) back into Eq. (38). This involves setting both α and β equal to ρ in Eq. (38). It is straightforward to obtain that the singular contribution of $G_{\rho\rho}$ comes from that in the self-energy $\Sigma_{\hat{g}\hat{g}}$ in the form

$$G_{\rho\rho} = -AG_{\rho\hat{g}}\delta(\omega)G_{\hat{g}\rho} + \bar{\Sigma}^R_{\hat{g}\hat{g}}, \tag{68}$$

where $\bar{\Sigma}^R$ is the part of the correlation function contributed by the regular part Σ^R. For an ENE transition to occur, it is needed that the response function $G_{\rho\hat{g}}$ does not vanish as $\omega \to 0$. The response functions $G_{\alpha\hat{\beta}}$ are calculated from Eq. (37), where $N_{\alpha\hat{\beta}}$ are as given in Table III,

$$G_{\rho\hat{g}} = \frac{N_{\rho\hat{g}}}{\mathcal{D}} = \frac{q(\omega + iq^2\nu)}{\mathcal{D}}. \tag{69}$$

The RHS of the above equation is nonzero since $\nu(\omega)$ and $\nu'(\omega)$, as defined in Eqs. (46) and (47), respectively, are nonzero in the $\omega \to 0$ limit. In the same zero frequency limit, the determinant $\mathcal{D}$ does not blow up as ωL, c^2, and ν are finite. With these assumptions, $\mathcal{D}(\omega \to 0)$ is not infinite; hence, $G_{\rho\hat{g}} \neq 0$ in the low frequency limit. Therefore, the density correlation function $G_{\rho\rho}$ develops a $\delta(\omega)$ part. In an similar way it follows that the correlations $G_{\rho c}$ and G_{cc} each develop a singular part $[\sim\delta(\omega)]$ by coupling to $\Sigma_{\hat{g}\hat{g}}$. This is a consequence of the fact that both $\nu(\omega)$ and $\nu'(\omega)$ are nonzero in the small ω limit.

TABLE III. Elements of matrix $G^{-1}_{\hat{\alpha}\beta}$ in terms of the renormalized transport coefficients L, ν, and ν' respectively defined in Eqs. (45)–(47). The symbols υ^2 and c^2 are explained in the text.

	ρ	c	g
$\hat{\rho}$	ω	0	$-q$
$\hat{c}$	$iq^2\nu'$	$\omega + iq^2\nu$	$-\Sigma_{\hat{c}g}$
$\hat{g}$	$-qc^2$	$-q\upsilon^2$	$\omega + iq^2L$

In comparison to the above result, the correlation functions involving a momentum index $\mathbf{g}$ do not contain a δ-function peak at zero frequency. To demonstrate this, we note that if either of the indices α or β on the LHS of Eq. (38) is set equal to g, then the singular contribution in $\Sigma_{\hat{g}\hat{g}}$ is coupled to the response function $G_{g\hat{g}}$. From Table III, it follows that

$$G_{g\hat{g}} = \frac{\omega(\omega + iq^2\nu)}{\mathcal{D}(\omega)}, \tag{70}$$

which means that $G_{g\hat{g}}$ vanishes as $\omega \to 0$ as long as $\mathcal{D}(\omega = 0) \neq 0$. Therefore, the correlation functions involving a momentum index g do not show a δ-function peak at zero frequency.

To summarize, for a binary mixture all the three correlations $G_{\rho\rho}(\omega)$, $G_{\rho c}(\omega)$, and $G_{cc}(\omega)$ develop a singular piece $\sim\delta(\omega)$ or equivalently develop a nonzero long time limit signifying an ENE transition. It is important to note here that the possibility of the ENE transition requires that both $\nu(\omega)$ and $\nu'(\omega)$ are nonzero in the small ω limit.

B. Tagged-particle dynamics

We consider the system for which the two species are identical, i.e., the size ratio α and mass ratio κ are both equal to 1 and the number of the particles $N_1 = 1$ and $N_2 = N - 1$. This is referred to as the one component limit in the following. For large N, the relative fractions for the two species are $x_1 \to 0$ and $x_2 \to 1$. The concentration variable c, defined in Eq. (8) reduces to the tagged particle density ρ_s and hence the correlation function G_{cc} reduces to tagged-particle correlation function G_s [35]. The present theory therefore reduces to the MCT for the total and tagged-particle dynamics in a one component liquid. We first consider the behavior of the corresponding static susceptibilities $\chi_{\rho\rho}$, χ_{cc}, and $\chi_{\rho c}$. The inverse static susceptibilities are respectively obtained in the one component limit as

$$\beta\chi^{-1}_{\rho\rho}(\mathbf{k}) = \frac{1}{m^2nS(k)}, \quad \beta\chi^{-1}_{cc}(\mathbf{k}) = \frac{1}{m^2nx_1x_2},$$
$$\beta\chi^{-1}_{\rho c}(\mathbf{k}) = 0. \tag{71}$$

Here $S(k)$ denotes the static structure factor [16] of the one-component liquid. The above results also agree with the wave-vector dependent formulas [15] obtained by using Ornstein-Zernike relations [16,17,36] for the partial structure factors of a binary liquid. The key point here is that the off diagonal element $(\rho - c)$ vanishes and the susceptibility matrix is diagonal in the one component limit. From the definition (47) it directly follows that $\nu'(\omega)$ which is nonzero for a binary mixture, vanishes in this limit. As a result of this, we obtain $N_{c\hat{g}} \to 0$. The above analysis implies that the correlation function G_{cc} does not have any contribution coming from the singular part [$\sim\delta(\omega)$] in the self-energy $\Sigma_{\hat{g}\hat{g}}$. Hence, $G_s \rightarrow 0$ in the small frequency limit. However, with ν'(and hence ν_1) vanishing, we have

$$G_{\rho\hat{g}} = \frac{q}{\omega^2 - q^2c^2 + iq^2\omega L}. \tag{72}$$

Thus, $G_{\rho\hat{g}}$ is nonzero in the small ω limit. Applying the same argument used above with Eq. (68), the collective density correlation function $G_{\rho\rho}$ has the singular contribution from $\Sigma_{\hat{g}\hat{g}}$ self-energy. Thus, in the one component limit the collective density correlation function freezes at the ENE transition although the tagged particle correlation $G_s(q,t)$ goes to zero in long time limit. The corresponding self-diffusion coefficient for the tagged particle is nonzero in the hydrodynamic limit. Thus, the single particle dynamics decouples from the dynamics of collective variables. In this respect the conclusion of the present work differs fundamentally from existing mode coupling model for binary systems.

VII. DISCUSSION

We have studied here the mode coupling dynamics of a binary mixture in terms of the microscopically conserved densities for the two component system, namely the total density ρ, the concentration variable c, and the total momentum density **g**. The dynamics is described in terms of nonlinear Langevin equations for the modes with white noise. The correlations of the noise in the respective stochastic equations of motions define the bare transport coefficients for the system. In the present formulation, keeping consistency with the white noise, there are two transport coefficients, namely the bare viscosity L^0 and the γ_0 for the interdiffusion. Using a MSR field theory we have studied the effects of the nonlinearities in the Langevin equations of the slow variables on the long time limit of dynamic correlations. From a careful consideration of the renormalizability of the theory, we obtain the relevant mode coupling contributions which drive the system to an ENE transition. It is shown respectively in Eqs. (B14) and (B15) that the renormalized interdiffusion γ_0 couples through the respective static correlations χ^{-1}_{cc} and $\chi^{-1}_{\rho c}$. Two effective transport coefficients ν and ν' appear and for the mixture of two species, with finite concentrations of each, both are nonzero. For such a mixture, we have shown that the feedback mechanism from slowly decaying density fluctuations drives the system in to an ENE transition at which all three correlation functions $\{G_{\rho\rho}, G_{\rho c}, G_{cc}\}$ freeze in the long time limit.

We also demonstrate here the conditions in which our results agree with existing formulation of MCT for a mixture. In the existing theory the individual momentum densities are treated as slow variables with separation of time scales in its dynamics, though these are nonconserved modes. For the Brownian limit (with high inertia), however, it is appropriate to write a Langevin equation for the momentum density of the single particle. A key aspect of our formulation of the MCT is that the location of the ENE transition as well as the nature of the dynamic correlation is now dependent on the mass ratio of the constituent particles of the mixture [33]. This is also in agreement with computer simulation results [37]. Study of our model with large mass ratio and very low concentration of one of the species shows that its results agree closely with the corresponding prediction of the NEP in the existing MCT model. The distinct nature of the single-particle dynamics in a sea of *identical* particles, observed here, is primarily a consequence of taking into account the conservation laws properly in the present theory. The physics involved is very different from that for the situation where a tagged-particle diffusion differs from collective dynamics in a very asymmetric mixtures with sufficient size disparities [38] of the constituent particles. Hence, it is more a geometrical effect and is linked to the peculiarities of the physics of cage formation process in such mixtures having very dissimilar components. Finally, though somewhat speculative at this point, it is useful to note here that the observed decoupling of collective dynamics from single particle motion, is more indicative of the violation of the Stokes-Einstein relation [39] than the converse. In existing MCT, since divergences are driven by that of the relaxation time of the density correlations, the Stokes-Einstein relation is not violated.

Dynamical light-scattering experiments for a bidisperse mixture of colloidal particles [40] with size ratio of 0.6 interacting nearly by a hard-sphere potential indicate that, increasing the concentration of smaller particles beyond a certain value slows down the dynamics in the system. This so called plasticization effect, which has been observed in existing MCT [38], is also confirmed from our model equations. In Fig. 3, the plot of the critical packing fraction η_c, i.e., the minimum packing fraction at which the binary mixture undergoes an ENE transition for a particular size ratio and concentration, against the concentration of smaller particles x_1 is displayed for five different size ratios. The plasticization effect holds for all values of the size ratio. The effect is, however, more pronounced for mixtures with high size disparity.

The present formulation of MCT reduces to the dynamics of both collective as well as the tagged-particle density correlation functions for a one component system. In the final section of the paper we have considered this so called one component limit of our model. Our analysis demonstrates that the dynamics of the tagged particle correlation and the total density correlation are decoupled in this limit. The role of the static correlations is vital here. In the one component limit ν' becomes zero due to the vanishing of the susceptibility factor in Eq. (B15). In this case we show that the collective correlation freezes at the transition while the single-particle correlation decays to zero. Hence, the self-diffusion coefficient remains finite. In the existing MCT model for one component systems [30], with the same static correlation matrix, the tagged-particle dynamics is slaved to that of the total density correlation. According to these theories, at the ENE transition of the MCT, both time autocorrelation functions, collective as well as single particle, are simultaneously nonzero in the long time limit. This implies that the self-diffusion coefficient vanishes at the ENE transition. This is where our result is crucially different from existing MCT for one component systems. In this regard it would be useful to reexamine the practice of “locating” the ENE transition point in simulation studies or experiments are by *extrapolating* the diffusion constant of a tagged particle to zero. This so called MCT transition point does not agree with the same obtained from the integral equations of MCT [7] using equilibrium structure factor as an input.

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APPENDIX A: STRUCTURE AND DYNAMICS OF THE MIXTURE

In this Appendix we provide a brief description of the structure and dynamics of the binary mixture in terms of the hydrodynamic modes in the system. The first involves the free energy functional in terms of which equilibrium correlation are defined. The dynamics is described with stochastic nonlinear equations of generalized hydrodynamics.

1. Free energy functional

The equal time correlations of the hydrodynamic modes and the structural properties of the mixture are obtained from the averages taken in terms of the equilibrium probability distribution for the fluctuations [see Eq. (3) for a definition]. For this, the corresponding free energy functional expressed in terms of the slow modes is necessary. In the construction of the generalized Langevin equation (1), the the free energy F as a functional of the slow modes $\{\psi_\alpha\}$ is also needed. The functional F is generally divided into two parts: The momentum density dependent part is generally referred to as the kinetic part F_K and the rest as the potential part F_U:

$$F = F_K + F_U. \tag{A1}$$

The kinetic part F_K is given in Eq. (13). The potential part of the free energy functional F_U dependent on ρ and c is taken to be Gaussian in these variables at the simplest level in the present theory. The so called potential part $F_U[\rho,c]$ has two contributions,

$$F_U[\rho,c] = F_{\rm id} + F_{\rm in}, \tag{A2}$$

with $F_{\rm id}$ the so called ideal gas part for a noninteracting system. In terms of the slow variables $\{\rho_1,\rho_2\}$, the ideal gas part is obtained as

$$F_{\rm id} = \sum_{s,s'} \frac{1}{m_s} \int dx \rho_s(\mathbf{x}) \left[\ln\left(\frac{\rho_s}{\rho_{0s}}\right) - 1 \right]. \tag{A3}$$

The ideal part of free energy functional is non-Gaussian. However, the log term is approximated to have a Gaussian form. $F_{\rm in}$ is the interaction part. The standard density functional expansion [15] of the interaction part of the free energy $F_{\rm in}$ is obtained in terms of a functional Taylor series expansion involving the direct correlation functions $c_{ss'}$, where $s,s' = 1,2$ for the mixture,

$$F_{\rm ex} = F_0 - \sum_{s,s'=1}^{2} \frac{1}{2m_s m_{s'}} \int d\mathbf{x}_1 \int d\mathbf{x}_2 c_{ss'}(\mathbf{x}_1,\mathbf{x}_2;n_0) \times \delta\rho_s(\mathbf{x}_1)\delta\rho_{s'}(\mathbf{x}_2) + \cdots , \tag{A4}$$

where F_0 is the free energy of the uniform liquid state. The direct correlation functions $c_{ss'}$ are defined in the density functional formulation as

$$c_{ss'}(\mathbf{x},\mathbf{x}') = \left. \frac{\delta^2 F_{\rm in}[n(\mathbf{x})]}{\delta n_s(\mathbf{x}) \delta n_{s'}(\mathbf{x}')} \right|_0 , \tag{A5}$$

with the "0" implying that the functional derivative is evaluated for the equilibrium liquid state. In terms of the fields $\{\rho,c\}$ the functional F_U is written in the Gaussian form,

$$F_U[\rho,c] = \frac{1}{2} \sum_{\alpha,\sigma} \int d\mathbf{x} \int d\mathbf{x}' \delta\rho_\alpha(\mathbf{x}) \chi^{-1}_{\alpha\sigma}(\mathbf{x}-\mathbf{x}') \delta\rho_\sigma(\mathbf{x}'). \tag{A6}$$

In the above expression for the free energy the $\chi^{-1}_{\alpha\sigma}$ denotes the $\alpha\sigma$th element of the inverse of the equal time correlation matrix or the so called susceptibility matrix $\chi_{\alpha\sigma}$, where $\alpha,\sigma \in \{\rho,c\}$. The above free energy is also conveniently expressed as a Gaussian functional of the pair $\{\rho_1,\rho_2\}$ instead of the set $\{\rho,c\}$ as

$$F_U[\rho_s] = \frac{1}{2} \int d\mathbf{x} \int d\mathbf{x}' \sum_{s,s'=1}^{2} \delta\rho_s(\mathbf{x}) \chi^{-1}_{ss'}(\mathbf{x}-\mathbf{x}') \delta\rho_{s'}(\mathbf{x}'). \tag{A7}$$

Using the results (A2), (A3), and (A5), and doing a quadratic order expansion in density fluctuations, the $\chi_{ss'}$'s are expressed in terms the corresponding direct correlation functions $c_{ss'}$ for a mixture through Ornstein-Zernike relations [16,36]. The elements of the static susceptibility matrix $\chi_{\alpha\sigma}(q)$ are obtained in terms of the direct correlation functions $c_{\alpha\sigma}$, with $\alpha,\sigma \in \{\rho,c\}$, as

$$\chi^{-1}_{\rho\rho}(q) = \frac{\beta^{-1}}{m_1^2 n_0} \left[x_1 + \frac{x_2}{\kappa^2} - c_{\rho\rho}(q) \right], \tag{A8}$$

$$\chi^{-1}_{\rho c}(q) = \frac{\beta^{-1}}{m_1 m_2 n_0} \left[\kappa - \frac{1}{\kappa} - c_{\rho c}(q) \right], \tag{A9}$$

$$\chi^{-1}_{cc}(q) = \frac{\beta^{-1}}{m_2^2 n_0} \left[\frac{\kappa^2}{x_1} + \frac{1}{x_2} - c_{cc}(q) \right], \tag{A10}$$

where $c_{\rho\rho}(q)$, $c_{\rho c}(q)$, and $c_{cc}(q)$ are given by the following expressions:

$$c_{\rho\rho}(q) = x_1^2 \bar{c}_{11}(q) + 2(x_1 x_2/\kappa)\bar{c}_{12}(q) + \left(x_2^2/\kappa^2\right)\bar{c}_{22}(q), \tag{A11}$$

$$c_{\rho c}(q) = x_1 \kappa \bar{c}_{11}(q) + (x_2 - x_1)\bar{c}_{12}(q) - (x_2/\kappa)\bar{c}_{22}(q), \tag{A12}$$

$$c_{cc}(q) = \kappa^2 \bar{c}_{11}(q) - 2\kappa \bar{c}_{12}(q) + \bar{c}_{22}(q). \tag{A13}$$

We have used in Eq. (A11) the notation $\bar{c}_{ss'}(q) = n_0 c_{ss'}(q)$ for $s = 1,2$. The quantity κ is the mass ratio m_2/m_1 of the two species and the relative abundance is denoted as $x_s = N_s/N$ for $s = 1,2$.

2. The generalized Langevin equations

The Langevin equation (1) involves a deterministic or slow part expressed in terms of the variables ψ_α and a stochastic part. These are respectively given by (a) the first two terms on the RHS and (b) the third term on the RHS. The calculation of reversible part of the Langevin equations for the slow variables requires the Poisson bracket relations between the slow variables.

a. Poisson brackets

Using the basic Poisson bracket relations between the phase space coordinates and the microscopic Eqs. (4)–(8) for the slow variables $\rho(\mathbf{x},\mathbf{t})$, $c(\mathbf{x},t)$, and $g(\mathbf{x},t)$, we obtain

$$\{\rho(\mathbf{x}),g_i(\mathbf{x}')\} = -\nabla_i[\delta(\mathbf{x}-\mathbf{x}')\rho(\mathbf{x})], \tag{A14}$$

$$\{c(\mathbf{x}),g_i(\mathbf{x}')\} = -\nabla_i[\delta(\mathbf{x}-\mathbf{x}')c(\mathbf{x})], \tag{A15}$$

$$\{g_i(\mathbf{x}),g_j(\mathbf{x}')\} = -\nabla_j[\delta(\mathbf{x}-\mathbf{x}')g_i(\mathbf{x})] + \nabla'_i[\delta(\mathbf{x}-\mathbf{x}')g_j(\mathbf{x}')]. \tag{A16}$$

All other Poisson brackets between the different members of the set $\{\rho,c,\mathbf{g}\}$ are zero.

b. Bare dissipation coefficients

Next we consider the dissipative and stochastic contributions respectively given by the second and third terms in the RHS of Eq. (1). The bare transport matrix has been introduced as $\Gamma^0_{\alpha\beta}$ in Eq. (1) in the main text, with the superscript "0" signifying that it represents the short time uncorrelated dynamics. In the present section, we will suppress the superscript 0 of the Γ matrix to avoid cluttering with indices. This modified notation is adopted for this section only. The various elements of the matrix $\Gamma_{\alpha\sigma}$ are chosen keeping consistency with the structure of the Langevin equations. The continuity equation is maintained for the density equation. For the density field $\rho(\mathbf{x},t)$ and the corresponding current, i.e., the momentum density field $\mathbf{g}(\mathbf{x},t)$ is conserved, all the dissipative terms involving ρ, *viz.*$\Gamma_{\rho\psi} = \Gamma_{\psi\rho} = 0$. For determining the elements of the noise correlation matrix between g and c, the symmetry considerations and compatibility with the white noise both play important roles. In this respect we note that the dissipative tensor $\Gamma_{\alpha\sigma}$ should follow the time reversal symmetry given by

$$\Gamma_{\alpha\sigma}(-t) = \epsilon_\alpha\epsilon_\sigma\Gamma_{\alpha\sigma}(t), \tag{A17}$$

where $\epsilon_\alpha = \pm 1$ represents the time reversal signature of the slow variable ψ_α. Applying this to the diagonal elements Γ_{cc} and $\Gamma_{g_ig_j}$ we obtain $\Gamma_{cc}(-t) = \Gamma_{cc}(t)$ and $\Gamma_{g_ig_j}(-t) = \Gamma_{g_ig_j}(t)$. However, for the element Γ_{cg_i} the implications are different:

$$\Gamma_{cg_i}(-t) = -\Gamma_{cg_i}(t). \tag{A18}$$

The element Γ_{cg_i} is related to the correlations of noise f (say) in the c equation and that in the $\mathbf{g}$ equation, i.e., θ through a fluctuation-dissipation relation,

$$\langle f(\mathbf{x},t)\theta_i(\mathbf{x}',t')\rangle = 2\beta^{-1}\Gamma_{cg_i}\delta(t-t')\delta(\mathbf{x}-\mathbf{x}'). \tag{A19}$$

We note here that the symmetry (A18) cannot be maintained if $\Gamma_{\alpha\sigma}(t)$ represents white noise, i.e., is represented by a δ function. However, for construction of the FNH equations and validity of the formalism adopted here the white noise is an essential input. For the conserved modes considered here, the separation of time scales is valid. Therefore, to maintain consistency with the white noise we take $\Gamma_{cg_i} = \Gamma_{g_ic} = 0$. The noise correlation in the c and $\mathbf{g}$ equations is given by fluctuation dissipation relations:

$$\langle f(\mathbf{x},t)f(\mathbf{x}',t')\rangle = 2\beta^{-1}\Gamma_{cc}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'), \tag{A20}$$

$$\langle\theta_i(\mathbf{x},t)\theta_j(\mathbf{x}',t')\rangle = 2\beta^{-1}L^0_{ij}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'), \tag{A21}$$

$$\langle f(\mathbf{x},t)\theta_i(\mathbf{x}',t')\rangle = 0. \tag{A22}$$

For the diagonal element Γ_{cc}, for keeping consistency with conservation laws, we have $\Gamma_{cc} = \gamma_0\nabla^2$. On the other hand, $\Gamma_{g_ig_j}$ elements which appear in the momentum equation are represented in terms of the bare viscosities $\Gamma_{g_ig_j} = L^0_{ij}$. For an isotropic system the L^0_{ij} matrix is described in terms of two independent quantities; namely, the bulk viscosity ζ_0 and the shear viscosity η_0 are defined as

$$L^0_{ij} = -\left(\zeta_0 + \frac{\eta_0}{3}\right)\nabla_i\nabla_j - \eta_0\delta_{ij}\nabla^2. \tag{A23}$$

The longitudinal viscosity is defined as $L_0 = \zeta_0 + 4\eta_0/3$. Taking into account the Poisson bracket relations given by Eqs. (A14)–(A16) and Eqs. (13), (A6), and (A1) for the free energy functional F, we obtain the following Langevin equations respectively for the set of slow variables $\{\rho,c,\mathbf{g}\}$ stated in Eqs. (9) and (11).

APPENDIX B: THE MARTIN-SIGGIA-ROSE FIELD THEORY

1. The MSR action functional

In the standard MSR formalism, the correlation and response functions are determined using an action functional constructed for the field theory. For a set of fields $\{\psi_\alpha\}$ with equations of motion given by (1), the average of a functional $f[\psi]$ is obtained as [23]

$$\langle f[\psi]\rangle = \frac{\int D\psi \int D\hat\psi\, f[\psi]\exp[-\mathcal{A}[\psi,\hat\psi]]}{\int D\psi \int D\hat\psi\, \exp[-\mathcal{A}[\psi,\hat\psi]]}. \tag{B1}$$

The action functional $\mathcal{A}[\psi,\hat\psi]$ is obtained as

$$\mathcal{A}[\psi,\hat\psi] = \int d1 \int d2\hat\psi(1)\beta^{-1}\Gamma^0(12)\hat\psi(2) + i\int d1\hat\psi(1)\left\{\frac{\partial\psi(1)}{\partial t_1} + [Q+\Gamma^0]\frac{\delta F}{\delta\psi}\right\}, \tag{B2}$$

where we have not explicitly written the the field indices to avoid cluttering. The expression (B1) for the average of the functional $f(\psi)$ is used to write the averages of fields and higher order correlation functions in terms of a generating functional Z_ξ. Assuming $f(\psi) \equiv \psi$, we write

$$\langle\psi(1)\rangle = \frac{\delta}{\delta\xi(1)}\ln Z_\xi. \tag{B3}$$

with

$$Z_\xi = \int D\psi \int D\hat\psi\, \exp[-\mathcal{A}_\xi[\psi,\hat\psi]]. \tag{B4}$$

We have defined the generating functional Z_ξ by including a linear current term in the corresponding action $\mathcal{A}_\xi$ functional,

$$\mathcal{A}_\xi[\psi,\hat\psi] = \mathcal{A}[\psi,\hat\psi] - \int d1\xi(1)\psi(1). \tag{B5}$$

The multipoint correlation functions of the variables ψ's are obtained from the generating functional,

$$\langle \psi(1),\ldots,\psi(m)\rangle = \frac{1}{Z_\xi}\frac{\delta}{\delta\xi(1)}\cdots\frac{\delta}{\delta\xi(m)}Z_\xi\bigg|_{\xi=0}. \tag{B6}$$

From the expression for the MSR action (B2) and the equation of motion (1) it follows that the linear part of the dynamics produces a MSR action functional quadratic (Gaussian) in the fields. Using the explicit forms of the equations of motion (9)–(11) we obtain the corresponding MSR functional in the form

$$\begin{aligned}\mathcal{A} = \int dt \int d\mathbf{x}\Bigg(&\sum_{i,j}\hat{g}_i\beta^{-1}L^0_{ij}\hat{g}_j + \hat{c}\beta^{-1}\Gamma^0_{cc}\hat{c} + i\hat{\rho}\left[\frac{\partial\rho}{\partial t} + \nabla\cdot\mathbf{g}\right] + i\sum_i \hat{g}_i\Bigg[\frac{\partial g_i}{\partial t} + \rho\nabla_i\{\chi^{-1}_{\rho\rho}\delta\rho + \chi^{-1}_{\rho c}\delta c\}\\ &+ c\nabla_i\{\chi^{-1}_{cc}\delta c + \chi^{-1}_{\rho c}\delta\rho\} + \sum_j \nabla_j\{g_i \mathrm{v}_j\} + \sum_j L^0_{ij}\mathrm{v}_j\Bigg] + i\hat{c}\left[\frac{\partial c}{\partial t} + \nabla_i\{c v_i\} + \gamma_0\nabla^2\{\chi^{-1}_{\rho c}\delta\rho + \chi^{-1}_{cc}\delta c\}\right]\Bigg).\end{aligned} \tag{B7}$$

a. Invariance of the MSR action

Here we demonstrate that the MSR action functional given by Eq. (B7) is invariant under the transformation (23) for the set of slow modes $\{\rho,c,\mathbf{g}\}$. Changing t to $-t$ in Eq. (B7) and applying the time transformation rules for the above set and their corresponding hatted counterparts, the action reduces to

$$\begin{aligned}\mathcal{A}[\psi(-t),\hat{\psi}(-t)] &= \int dt\int d\mathbf{r}\Bigg[\left\{-\hat{g}_i + i\beta\frac{\delta F}{\delta g_i}\right\}\beta^{-1}L_{ij}\left\{-\hat{g}_i + i\beta\frac{\delta F}{\delta g_i}\right\} + \left\{\hat{c} - i\beta\frac{\delta F}{\delta c}\right\}\beta^{-1}\Gamma^0_{cc}\left\{\hat{c} - i\beta\frac{\delta F}{\delta c}\right\}\\ &\quad + i\left\{\hat{\rho} - i\beta\frac{\delta F}{\delta\rho}\right\}\left\{-\frac{\partial\rho}{\partial t} - Q_{\rho g_i}\frac{\delta F}{\delta g_i}\right\} + i\left\{-\hat{g}_i + i\beta\frac{\delta F}{\delta g_i}\right\}\left\{\frac{\partial g_i}{\partial t} + Q_{g_i g_j}\frac{\delta F}{\delta g_j} + Q_{g_i\rho}\frac{\delta F}{\delta\rho} + Q_{g_i c}\frac{\delta F}{\delta c} + L_{ij}\frac{\delta F}{\delta g_j}\right\}\\ &\quad + i\left\{\hat{c} - i\beta\frac{\delta F}{\delta c}\right\}\left\{-\frac{\partial c}{\partial t} - Q_{c g_i}\frac{\delta F}{\delta g_i} + \Gamma^0_{cc}\frac{\delta F}{\delta c}\right\}\Bigg]\\ &= \mathcal{A}[\psi,\hat{\psi}] + i\beta\int_{-\infty}^{\infty} d1\left\{\frac{\delta F}{\delta\psi_\alpha}Q_{\alpha\delta}\frac{\delta F}{\delta\psi_\delta}\right\} + \beta(F_\infty - F_{-\infty}).\end{aligned} \tag{B8}$$

The second term inside the curly brackets on the RHS vanishes since the dummy indices α and δ are summed over and the Poisson bracket $Q_{\alpha\delta}$ is odd under the interchange of the indices. The last term involving the F's also vanishes due to equilibrium. Hence, the MSR action is invariant under the time reversal transformations defined by Eq. (23).

2. Renormalized correlation functions

The construction of the field theoretical model fixes some basic characteristics of the structure of the correlation and the response functions. Let us first consider an important characteristic of the Green's function matrix G and G_0 in the MSR theory. For the cases in which both indices in the matrix Eq. (32) correspond to the unhatted fields, the following hold:

(a) $[\mathbf{G_0}^{-1}]_{\alpha\beta} = 0$, which follows from the action (B7) obtained in the MSR field theory;

(b) $\Sigma_{\alpha\beta} = 0$, which follows from causal nature of the response functions in MSR field theory.

From the Schwinger-Dyson equation (32) we obtain that the elements of the $\mathbf{G}^{-1}$ matrix corresponding to the unhatted fields, $[\mathbf{G}^{-1}]_{\alpha\beta} = 0$. Inverting the matrix $\mathbf{G}^{-1}$ which has the above structure, we obtain for the correlation functions of the physical, unhatted field variables,

$$G_{\alpha\beta} = -\sum_{\mu\nu} G_{\alpha\hat{\mu}}\mathcal{C}_{\hat{\mu}\hat{\nu}}G_{\hat{\nu}\beta}, \tag{B9}$$

where greek letter subscripts take values ρ,c and the longitudinal components of the vector field $\mathbf{g}$. The matrix $\mathcal{C}_{\hat{\mu}\hat{\nu}}$ is given by

$$\mathcal{C}_{\hat{\mu}\hat{\nu}} = [\mathcal{C}_0]_{\hat{\mu}\hat{\nu}} - \Sigma_{\hat{\mu}\hat{\nu}} \tag{B10}$$

and is listed in Table II. The double-hatted self-energies $\Sigma_{\hat{\mu}\hat{\nu}}$ and $\mathcal{C}_{\hat{\mu}\hat{\nu}}$ vanish if either index corresponds to $\hat{\rho}$, since there is no noise or nonlinearity in the continuity equation (9).

The response part of $G^{-1}_{\hat{\alpha}\beta}$ is obtained using Eq. (32) in terms of the corresponding elements of the matrix of $[\mathcal{B}_0 - \Sigma]$. The elements of the $G^{-1}_{\hat{\alpha}\beta}$ matrix are listed in Table III. The renormalized response function $G_{\alpha\hat{\varphi}}$ is obtained in the form

$$G_{\alpha\hat{\varphi}} = \frac{N_{\alpha\hat{\varphi}}}{\mathcal{D}}, \tag{B11}$$

where the elements of matrix $N_{\alpha\hat{\varphi}}$ are given in Table IV. The denominator $\mathcal{D}$ in the RHS of Eq. (37) is obtained as

$$\mathcal{D}(\mathbf{q},\omega) = (\omega + iq^2\nu)[\omega^2 - q^2c^2 + i\omega Lq^2] + iq^4\upsilon^2\nu_1. \tag{B12}$$

The various renormalized transport coefficients appearing on the RHS of Eq. (B12) are expressed in terms of the corresponding response self-energies,

$$Lq^2 = L_0q^2 + i\Sigma_{\hat{g}g}, \tag{B13}$$

$$\nu q^2 = \gamma_0\chi^{-1}_{cc}q^2 + i\Sigma_{\hat{c}c}, \tag{B14}$$

$$\nu' q^2 = \gamma_0\chi^{-1}_{\rho c}q^2 + i\Sigma_{\hat{c}\rho}. \tag{B15}$$

TABLE IV. Elements of matrix $N_{\alpha\hat{\beta}}$ in terms of the renormalized transport coefficients L, ν, and ν', respectively defined in Eqs. (45)–(47). The symbols υ^2 and c^2 are explained in the text. $\gamma_{\hat{c}g}$ is the leading order contribution to the corresponding self-energy $\Sigma_{\hat{c}g} = -iq^3\gamma_{\hat{c}g}$.

	$\hat{\rho}$	$\hat{c}$	$\hat{g}$
ρ	$(\omega + iq^2L)(\omega + iq^2\nu)$ $+iq^4\upsilon^2\gamma_{\hat{c}g}$	$q^2\upsilon^2$	$q(\omega + iq^2\nu)$
c	$-iq^2\{\nu'(\omega + iq^2L)$ $+q^2c^2\gamma_{\hat{c}g}\}$	$\omega(\omega + iq^2L)$ $-q^2c^2$	$-iq^3(\nu' + \omega\gamma_{\hat{c}g})$
g	$-iq^3\upsilon^2\nu' + qc^2(\omega + iq^2\nu)$	$\omega q\upsilon^2$	$\omega(\omega + iq^2\nu)$

The quantity ν_1 in the last term on the RHS of definition (B12) is obtained as $\nu_1 = \nu' + \omega\gamma_{\hat{c}g}$ in terms of the leading order nonzero contributions to the self-energy $\Sigma_{\hat{c}g} = -iq^3\gamma_{\hat{c}g}$ in the small q limit. c^2 and υ^2 respectively represent the renormalized expressions for the sound speeds $c_0^2 = \rho_0\chi_{\rho\rho}^{-1}$ and $\upsilon_0^2 = \rho_0\chi_{\rho c}^{-1}$ obtained in terms of the self-energies

$$c^2 = c_0^2 + q^{-1}\Sigma_{\hat{g}\rho}, \tag{B16}$$

$$\upsilon^2 = \upsilon_0^2 + q^{-1}\Sigma_{\hat{g}c}. \tag{B17}$$

3. Analysis of the FDRs

We begin with the FDR (24) corresponding to $\psi = g$,

$$G_{g_i\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\rho_0\mathrm{Im}G_{\hat{g}_i\varphi}(\mathbf{q},\omega). \tag{B18}$$

Using the definitions (37) and (38) for the correlation and response functions, respectively, in the above fluctuation-dissipation relation we obtain the result

$$\begin{aligned} \sum_{\hat{\alpha},\hat{\gamma}} G_{g_i\hat{\alpha}}\mathcal{C}_{\hat{\alpha}\hat{\gamma}}G_{\hat{\gamma}\varphi} &= 2\beta^{-1}\rho_0\mathrm{Im}G_{\hat{g}_i\varphi}, \\ \sum_{\hat{\alpha}} G_{g_i\hat{\alpha}}\mathcal{C}_{\hat{\alpha}\hat{\gamma}} &= -i\beta^{-1}\rho_0\sum_{\beta}[G_{\hat{g}_i\varphi} - G^*_{\hat{g}_i\varphi}]G^{-1}_{\varphi\hat{\gamma}}, \qquad \text{(B19)} \\ \sum_{\hat{\alpha}} N_{g_i\hat{\alpha}}\mathcal{C}_{\hat{\alpha}\hat{\gamma}} &= -i\beta^{-1}\rho_0\left(\mathcal{D}\delta_{\hat{g}_i\hat{\gamma}} + \sum_{\beta}N^*_{\hat{g}_j\varphi}G^{-1}_{\varphi\hat{\gamma}}\right). \end{aligned}$$

On substituting $\hat{\gamma} = \hat{c}$ and $\hat{g}$ in Eq. (B19), we obtain respectively the following equations:

$$\begin{aligned} &N_{g\hat{c}}\mathcal{C}_{\hat{c}\hat{c}} + N_{g\hat{g}}\mathcal{C}_{\hat{g}\hat{c}} \\ &\quad = -i\beta^{-1}\rho_0(N^*_{\hat{g}\rho}G^{-1}_{\rho\hat{c}} + N^*_{\hat{g}c}G^{-1}_{c\hat{c}} + N^*_{\hat{g}g}G^{-1}_{g\hat{c}}), \end{aligned} \tag{B20}$$

$$\begin{aligned} &N_{g\hat{c}}\mathcal{C}_{\hat{c}\hat{g}} + N_{g\hat{g}}\mathcal{C}_{\hat{g}\hat{g}} \\ &\quad = -i\beta^{-1}\rho_0(\mathcal{D} + N^*_{\hat{g}\rho}G^{-1}_{\rho\hat{g}} + N^*_{\hat{g}c}G^{-1}_{c\hat{g}} + N^*_{\hat{g}g}G^{-1}_{g\hat{g}}). \end{aligned} \tag{B21}$$

Equating the real and imaginary parts from both sides and using the fact that the elements $\mathcal{C}_{\hat{g}\hat{g}}$ and $\mathcal{C}_{\hat{c}\hat{c}}$ are real while $\mathcal{C}_{\hat{g}\hat{c}} = \mathcal{C}^*_{\hat{c}\hat{g}}$ are not, we obtain a set of relations between the correlation and response self-energies. For the self-energy elements $\mathcal{C}_{\hat{g}\hat{g}}$ and $\mathcal{C}_{\hat{c}\hat{c}}$ we respectively obtain the results

$$\begin{aligned} &\mathcal{C}_{\hat{g}\hat{g}} + \frac{|N_{g\hat{c}}|^2}{\mathcal{M}}\mathcal{C}'_{\hat{c}\hat{g}} \\ &\quad = 2\beta^{-1}\rho_0\left[\frac{N'_{g\hat{c}}}{\mathcal{M}}\{N'_{\hat{g}g}G^{-1\prime\prime}_{g\hat{g}} + N'_{\hat{g}c}G^{-1\prime\prime}_{c\hat{g}} + N'_{\hat{g}\rho}G^{-1\prime\prime}_{\rho\hat{g}}\}\right. \\ &\qquad \left. + \frac{N''_{g\hat{c}}}{\mathcal{M}}\{N''_{\hat{g}g}G^{-1\prime\prime}_{g\hat{g}} + N''_{\hat{g}c}G^{-1\prime\prime}_{c\hat{g}} + N''_{\hat{g}\rho}G^{-1\prime\prime}_{\rho\hat{g}}\}\right], \end{aligned} \tag{B22}$$

$$\begin{aligned} \mathcal{C}_{\hat{c}\hat{c}} + \frac{|N_{g\hat{g}}|^2}{\mathcal{M}}\mathcal{C}'_{\hat{c}\hat{g}} &= 2\beta^{-1}\rho_0\left[\frac{N'_{g\hat{g}}}{\mathcal{M}}\{N'_{\hat{g}\rho}G^{-1\prime\prime}_{\rho\hat{c}} - N''_{\hat{g}\rho}G^{-1\prime}_{\rho\hat{c}}\right. \\ &\qquad \left. + N'_{\hat{g}g}G^{-1\prime\prime}_{g\hat{c}} - N''_{\hat{g}g}G^{-1\prime}_{g\hat{c}}\}\right], \end{aligned} \tag{B23}$$

with $\mathcal{M} = N'_{g\hat{g}}N'_{g\hat{c}} + N''_{g\hat{g}}N''_{g\hat{c}}$. The single and double primes above indicate the real and imaginary parts of the corresponding complex quantity, respectively.

Next we consider the FDR (30) to obtain another set of relations between the correlation and response self-energies. Evaluating the functional derivatives ζ_c for a Gaussian free energy, we obtain the result

$$\chi_{\rho c}^{-1}G_{\rho\varphi}(\mathbf{q},\omega) + \chi_{cc}^{-1}G_{c\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\mathrm{Im}G_{\hat{c}\varphi}(\mathbf{q},\omega). \tag{B24}$$

Following the same procedures as in the case of the FDR, Eq. (B24) reduces to the form

$$\begin{aligned} &\sum_{\hat{\alpha}}[\chi_{\rho c}^{-1}N_{\rho\hat{\alpha}}\mathcal{C}_{\hat{\alpha}\hat{\gamma}} + \chi_{cc}^{-1}N_{c\hat{\delta}}\mathcal{C}_{\hat{\delta}\hat{\gamma}}] \\ &\qquad = -i\beta^{-1}\left\{\mathcal{D}\delta_{\hat{c}\hat{\gamma}} + \sum_{\beta}N^*_{\hat{c}\varphi}G^{-1}_{\varphi\hat{\gamma}}\right\}. \end{aligned} \tag{B25}$$

Setting $\hat{\gamma} = \hat{c}$ in Eq. (B25), we obtain

$$\begin{aligned} &\chi_{\rho c}^{-1}(N_{\rho\hat{c}}\mathcal{C}_{\hat{c}\hat{c}} + N_{\rho\hat{g}}\mathcal{C}_{\hat{g}\hat{c}}) + \chi_{cc}^{-1}(N_{c\hat{c}}\mathcal{C}_{\hat{c}\hat{c}} + N_{c\hat{g}}\mathcal{C}_{\hat{g}\hat{c}}) \\ &\quad = -i\beta^{-1}[\mathcal{D} + N^*_{\hat{c}\rho}G^{-1}_{\rho\hat{c}} + N^*_{\hat{c}c}G^{-1}_{c\hat{c}} + N^*_{\hat{c}g}G^{-1}_{g\hat{c}})]. \end{aligned} \tag{B26}$$

Next, substituting $\hat{\gamma} = \hat{g}$ in Eq. (B25), we obtain

$$\begin{aligned} &\chi_{\rho c}^{-1}(N_{\rho\hat{c}}\mathcal{C}_{\hat{c}\hat{g}} + N_{\rho\hat{g}}\mathcal{C}_{\hat{g}\hat{g}}) + \chi_{cc}^{-1}(N_{c\hat{c}}\mathcal{C}_{\hat{c}\hat{g}} + N_{c\hat{g}}\mathcal{C}_{\hat{g}\hat{g}}) \\ &\quad = -i\beta^{-1}[N_{\hat{c}\rho}G^{-1}_{\rho\hat{g}} + N_{\hat{c}c}G^{-1}_{c\hat{g}} + N_{\hat{c}g}G^{-1}_{g\hat{g}})]. \end{aligned} \tag{B27}$$

Comparing real and imaginary parts of Eqs. (B26) and (B27), we obtain the following results, respectively, for $\mathcal{C}_{\hat{c}\hat{c}}$ and $\mathcal{C}_{\hat{g}\hat{g}}$:

$$\begin{aligned} &\mathcal{C}_{\hat{c}\hat{c}} + \frac{|\mathcal{J}|^2}{\mathcal{Q}}\mathcal{C}'_{\hat{c}\hat{g}} \\ &\quad = 2\beta^{-1}\left[\frac{\mathcal{J}'}{\mathcal{Q}}\{N'_{\hat{c}\rho}G^{-1\prime\prime}_{\rho\hat{c}} + N'_{\hat{c}c}G^{-1\prime\prime}_{c\hat{c}} + N'_{\hat{c}g}G^{-1\prime\prime}_{g\hat{c}}\}\right. \\ &\qquad \left. + \frac{\mathcal{J}''}{\mathcal{Q}}\{N''_{\hat{c}\rho}G^{-1\prime\prime}_{\rho\hat{c}} + N''_{\hat{c}c}G^{-1\prime\prime}_{c\hat{c}} + N''_{\hat{c}g}G^{-1\prime\prime}_{g\hat{c}}\}\right], \end{aligned} \tag{B28}$$

$$\begin{aligned} \mathcal{C}_{\hat{g}\hat{g}} + \frac{|\mathcal{K}|^2}{\mathcal{Q}}\mathcal{C}'_{\hat{c}\hat{g}} &= 2\beta^{-1}\left[\frac{\mathcal{K}'}{\mathcal{Q}}\{N'_{\hat{c}\rho}G^{-1\prime\prime}_{\rho\hat{g}} - N''_{\hat{c}\rho}G^{-1\prime}_{\rho\hat{g}}\right. \\ &\qquad \left. + N'_{\hat{c}g}G^{-1\prime\prime}_{g\hat{g}} - N''_{\hat{c}g}G^{-1\prime}_{g\hat{g}}\}\right]. \end{aligned} \tag{B29}$$

In the above equations we have defined the quantities $\mathcal{J}$, $\mathcal{K}$, and $\mathcal{Q}$ in terms of the matrix elements of $N_{\alpha\hat{\beta}}$ and $\chi^{-1}_{\alpha\beta}$ as follows:

$$\mathcal{J} = \chi^{-1}_{\rho c} N_{\rho\hat{g}} + \chi^{-1}_{cc} N_{c\hat{g}}, \tag{B30}$$

$$\mathcal{K} = \chi^{-1}_{\rho c} N_{\rho\hat{c}} + \chi^{-1}_{cc} N_{c\hat{c}}, \tag{B31}$$

$$\mathcal{Q} = \mathcal{J}'\mathcal{K}' + \mathcal{J}''\mathcal{K}''. \tag{B32}$$

In general, for finite wave number (q) and frequency (ω) the FDRs (B22), (B23), (B28), and (B29) between the real and imaginary parts of the correlation and response self-energies are complicated and difficult to resolve. Here we analyze their implications in the hydrodynamic limit of small q and ω by writing the dependence of the various self-energy elements to leading order in the wave number q using simple symmetry arguments.

4. Self-energy relations

We begin by considering the correlation self-energy matrix element $\Sigma_{\hat{g}_i\hat{g}_j}(\mathbf{q},\omega)$. From the MSR action functional for the two component mixture given by Eq. (B7), it follows that the cubic vertices with a $\hat{g}_i(q)$ leg each, contribute an explicit q_i factor. The self-energy $\Sigma_{\hat{g}_i\hat{g}_j}(\mathbf{q},\omega)$ has two vertices with external legs of $\hat{g}_i(q)$ and $\hat{g}_j(q)$. Hence, this self-energy involves an explicit factor of q_iq_j; i.e., $\Sigma_{\hat{g}_i\hat{g}_j} \sim -q_iq_j\gamma_{\hat{g}\hat{g}} \equiv -q^2\gamma_{\hat{g}_i\hat{g}_j}$. Let us now consider the self-energy element $\Sigma_{\hat{c}\hat{c}}(\mathbf{q},\omega)$. The cubic vertex with a $\hat{c}(q)$ leg contributes a q factor. Hence, using similar arguments as above, the self-energy $\Sigma_{\hat{c}\hat{c}}(\mathbf{q},\omega)$ also involves an explicit factor of q^2. However, in this case the vector indices must be contracted to produce a scalar form. Therefore we obtain, $\Sigma_{\hat{c}\hat{c}} \sim -q^2\gamma_{\hat{c}\hat{c}}$. We also verify these results explicitly at the one loop order by considering the corresponding diagrams for $\Sigma_{\hat{g}_i\hat{g}_j}$ and $\Sigma_{\hat{c}\hat{c}}$.

Next, we consider the response self-energy $\Sigma_{\hat{g}_ig_j}(\mathbf{q},\omega)$ which contains only one external $\hat{g}_i$ contributing a factor q_i. The other leg of this self-energy involves the vector field g_j and hence the $O(q)$ level contribution must have the external q_i factor [due to the hatted field $\hat{g}_i(q)$] multiplied to an explicit internal k_j wave vector. The internal wave vector is integrated out. At this $O(q)$, the external q is set equal to zero in the integral for the diagrammatic contribution. However, this integral vanishes for being odd in k, due to the $\mathbf{k} \to -\mathbf{k}$ symmetry. Hence, this response self-energy is at least of the $O(q^2)$, and writing this out explicitly, we obtain $\Sigma_{\hat{g}_i\hat{g}_j} \sim -iq^2\gamma_{\hat{g}_ig_j}$. Using similar arguments for the self-energy $\Sigma_{\hat{c}c}(\mathbf{q},\omega)$, we note that since $\hat{c}$ is a scalar field the $O(q)$ contribution must have the external q factor contracted to an explicit k internal wave vector which is integrated out. Again, since at this order the external q is set equal to zero, the integral vanishes being odd in k. Hence, we define taking into account this factor $\Sigma_{\hat{c}c} \sim -iq^2\gamma_{\hat{c}c}$. We also verify these behaviors explicitly at the one loop order by considering the diagrams for the $\Sigma_{\hat{g}_ig_j}$ and $\Sigma_{\hat{c}c}$.

Next the self-energy $\Sigma_{\hat{g}_i\rho}(\mathbf{q},\omega)$ is considered. Due to the the external leg $\hat{g}_i$ in a vertex an explicit factor of q_i appear. To consider the $O(q)$ contribution of this self-energy, we therefore set $q = 0$ in all the internal integrations and thus this contribution vanishes. We can establish this result at the one loop order by considering the detailed nature of the vertices which contribute to this self-energy. These are of the following two types: (a) those which have one ρ leg [such vertices in the present model have *one other leg* with a vector index g_i or $\hat{g}_i$ (for example, the vertices $V_{\hat{g}_i\rho\rho}$, $V_{\hat{g}_i\rho c}$, and $V_{\hat{c}g_i\rho}$)]; (b) those which have one $\hat{g}_i$ leg [such vertices in the present model have *two other legs* each with a vector index g_j, etc. (for example, the vertices $V_{\hat{g}_ig_jg_i}$), or without *any other leg* having a vector index (for example, $V_{\hat{g}_i\rho c}$, $V_{\hat{g}_i\rho\rho}$, and $V_{\hat{g}_icc}$)]. As a result of this, the internal integration involved in the one loop diagrammatic contribution involves either one or three powers of the internal vector $\mathbf{k}$. For the isotropic liquid, such an integral must vanish, since using the $\mathbf{k} \to -\mathbf{k}$ symmetry the integrand is an odd function. Thus, the $O(q)$ contribution to $\Sigma_{\hat{g}_i\rho}(\mathbf{q},\omega)$ is taken to be zero. The next order surviving contribution must therefore be $O(q_iq^2)$. Hence, we write this self-energy as

TABLE V. q dependence of self-energies $\Sigma_{\hat{\alpha}\beta}$ and $\Sigma_{\hat{\alpha}\hat{\beta}}$.

	ρ	c	g_j	$\hat{c}$	$\hat{g}_j$
$\hat{c}$	$-iq^2\gamma_{\hat{c}\rho}$	$-iq^2\gamma_{\hat{c}c}$	$-iq^3\gamma_{\hat{c}g_j}$	$-q^2\gamma_{\hat{c}\hat{c}}$	$-q^3\gamma_{\hat{c}\hat{g}_j}$
$\hat{g}_i$	$-iq^3\gamma_{\hat{g}_i\rho}$	$-iq^3\gamma_{\hat{g}_ic}$	$-iq^2\gamma_{\hat{g}_ig_j}$	$-q^3\gamma_{\hat{g}_i\hat{c}}$	$-q^2\gamma_{\hat{g}_i\hat{g}_j}$

$$\Sigma_{\hat{g}_i\rho}(\mathbf{q},\omega) = -iq_iq^2\gamma_{\hat{g}\rho}(\mathbf{q},\omega) \equiv -iq^3\gamma_{\hat{g}_i\rho}(\mathbf{q},\omega). \tag{B33}$$

In a similar way we can show that $\Sigma_{\hat{g}_ic}(\mathbf{q},\omega) \sim -iq^3\gamma_{\hat{g}_ic}(\mathbf{q},\omega)$. Finally, the self-energy $\Sigma_{\hat{c}\hat{g}_i}$ has a factor q_i due to the external leg $\hat{g}_i$ and a factor of q_j due to the leg $\hat{c}$. The latter must be contracted with an internal k_j vector. Hence, the $O(q_iq_j)$ contribution involves an integral which is odd in k. The latter vanishes making the lowest order contribution to the self-energy being of $O(q_iq^2)$. Therefore, we write $\Sigma_{\hat{c}\hat{g}_i}(\mathbf{q},\omega) = -iq_iq^2\gamma_{\hat{c}\hat{g}}(\mathbf{q},\omega) \equiv -iq^3\gamma_{\hat{c}\hat{g}_i}(\mathbf{q},\omega)$. The leading order contributions to these self-energies are listed in Table V. Substituting the relevant elements of matrix $G^{-1}_{\alpha\hat{\beta}}$ and matrix $N_{\hat{\alpha}\beta}$ in Eqs. (B22) and (B23), we obtain by comparing leading order terms the following relations between the correlation and response type self-energies,

$$\gamma_{\hat{c}\hat{c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}\rho}}{\chi^{-1}_{\rho c}}, \tag{B34}$$

$$\gamma_{\hat{g}\hat{g}} = 2\beta^{-1}\rho_0\gamma'_{\hat{g}g}, \tag{B35}$$

and the comparison of next higher order terms from these two equations respectively obtain the self-energy relations,

$$\gamma_{\hat{c}\hat{g}} = 2\beta^{-1}\rho_0\gamma'_{\hat{c}g}, \tag{B36}$$

$$\gamma_{\hat{c}\hat{g}} = 2\beta^{-1}\frac{\gamma'_{\hat{g}\rho}}{\chi^{-1}_{\rho c}}. \tag{B37}$$

In a similar way, using elements of matrix $G^{-1}_{\alpha\hat{\beta}}$ and matrix $N_{\alpha\beta}$ in Eqs. (B28) and (B29) and by comparing leading order terms the following relations between the correlation and response type self-energies, we obtain

$$\gamma_{\hat{c}\hat{c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}c}}{\chi^{-1}_{cc}}, \tag{B38}$$

$$\gamma_{\hat{g}\hat{g}} = 2\beta^{-1}\rho_0\gamma'_{\hat{g}g}. \tag{B39}$$

Note that we have now reached the relation between $\gamma_{\hat{g}\hat{g}}$ with the corresponding response, self-energy $\gamma_{\hat{g}g}$ [Eqs. (B35) and (B39)] and either of these self-energies can be used to renormalize the longitudinal viscosity. On the other hand, Eqs. (B34) and (B38) link two response self-energies $\gamma_{\hat{c}\rho}$ and $\gamma_{\hat{c}c}$ to a single self-energy $\gamma_{\hat{c}\hat{c}}$ as

$$\gamma_{\hat{c}\hat{c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}\rho}}{\chi^{-1}_{\rho c}} = 2\beta^{-1}\frac{\gamma'_{\hat{c}c}}{\chi^{-1}_{cc}}. \tag{B40}$$

This proves an important relation by which two different transport coefficients are renormalized in terms of the same self-energy $\gamma_{\hat{c}\hat{c}}$. Finally, comparing the next order terms from Eq. (B29) and making use of the Eq. (B37) linking $\gamma'_{\hat{g}\rho}$ with $\gamma_{\hat{c}\hat{g}}$, we obtain the result

$$\gamma_{\hat{c}\hat{g}} = 2\beta^{-1}\frac{\gamma'_{\hat{g}c}}{\chi^{-1}_{cc}}. \tag{B41}$$

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