Renormalized dynamics of the Dean-Kawasaki model

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We study the model of a supercooled liquid for which the equation of motion for the coarse-grained density $\rho(\mathbf{x},t)$ is the nonlinear diffusion equation originally proposed by Dean and Kawasaki, respectively, for Brownian and Newtonian dynamics of fluid particles. Using a Martin-Siggia-Rose (MSR) field theory we study the renormalization of the dynamics in a self-consistent form in terms of the so-called self-energy matrix Σ. The appropriate model for the renormalized dynamics involves an extended set of field variables $\{\rho,\theta\}$, linked through a nonlinear constraint. The latter incorporates, in a nonperturbative manner, the effects of an infinite number of density nonlinearities in the dynamics. We show that the contributing element of Σ which renormalizes the bare diffusion constant D_0 to D_R is same as that proposed by Kawasaki and Miyazima [Z. Phys. B Condens. Matter **103**, 423 (1997)]. D_R sharply decreases with increasing density. We consider the likelihood of a ergodic-nonergodic (ENE) transition in the model beyond a critical point. The transition is characterized by the long-time limit of the density correlation freezing at a nonzero value. From our analysis we identify an element of Σ which arises from the above-mentioned nonlinear constraint and is key to the viability of the ENE transition. If this self-energy would be zero, then the model supports a sharp ENE transition with $D_R = 0$ as predicted by Kawasaki and Miyazima. With the full model having nonzero value for this self-energy, the density autocorrelation function decays to zero in the long-time limit. Hence the ENE transition is not supported in the model.

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I. INTRODUCTION

In a deeply supercooled liquid [1–3] strong fluctuations in the local density $\rho(\mathbf{x},t)$ play a dominant role in producing its characteristic slow dynamics. Freezing of the supercooled liquid into an amorphous structure starts at short length scales. At short wavelengths the energy and momentum fluctuations in the many-particle system are quickly transferred among the particles while the individual density fluctuations still decay much more slowly. It is therefore plausible that, at a somewhat simplified level, the dynamics for such a system can be described in terms of the density fluctuations only. This formulation has been referred to in the literature as the dynamic density-functional theory [4,5]. The corresponding equilibrium density-functional theory of freezing [6] was developed for the study of the freezing transition of the liquid into a crystal with long-range order. Based on the same principles, models for the amorphous glassy state have also been developed [7–11] treating the inhomogeneous density as an order parameter. In this paper we present the analysis of a model in which the equation of motion for the coarse-grained density is given by a nonlinear Langevin equation [12] with a diffusive kernel and multiplicative noise [13]. The stochastic equation for the dynamics of density fluctuations discussed here has also been used in studying the kinetics of the freezing transition into an ordered phase [14].

Keeping terms involving only the linear fluctuations for the collective density, $\delta\rho(\mathbf{x},t) = \rho(\mathbf{x},t) - \rho_0$, where ρ_0 is the average density in equilibrium, the Dean-Kawasaki equation represents diffusive dynamics with a (bare) diffusion constant D_0. With increase of ρ_0, the dynamics greatly slows down, a behavior generally attributed to strong correlations which develop in the dense liquid state. Theoretically, this effect is understood in terms of nonlinear couplings of density fluctuations in the equations controlling the time evolution of the many-particle system. A basic question to address here is with regard to whether we can express the nonlinear dynamics in terms of a renormalized version of D_0, generalized with wave number and frequency dependence as $D_R(q,z)$. Keeping similarity with methods used in quantum-field-theory models [15], the renormalization of D_0 is obtained in terms of the so-called self-energy contributions [5]. These corrections signify the role nonlinearities in the equations of motion. A primary focus in the first part of the present paper is to compute D_R in the hydrodynamic limit and obtain predictions for its density dependence. The renormalized form of the Dean-Kawasaki model is the starting point of analyzing the role of density nonlinearities on the long-time dynamics of the dense liquid. This is generally referred to as the mode-coupling theory of slow dynamics. Several different procedures exist for calculating the corrections to the linear dynamics in a perturbation theory. Using a simple iteration of the equation of motion [16,17] for $\delta\rho$ allows us to compute the lowest-order perturbative corrections in terms of the correlation functions of the linear theory [18–20]. For studying the feedback mechanism due to the slowly decaying density correlations, a self-consistent formulation of the mode-coupling theory with the Martin-Siggia-Rose field theory is very useful. This has been applied in various levels. For the corresponding field equations, generally the stochastic nonlinear equations resulting from the conservation laws of a selected set of collective modes are used. More recently, the field-theory approach has been applied at the level of the equations of motion for the particles following the Brownian dynamics [21,22] as well as the Newtonian dynamics [23] and the corresponding mode-coupling theory (MCT) has been worked out.

In its simplest form the self-consistent MCT predicts an ergodicity-nonergodicity (ENE) transition [5]. At the ENE transition the long-time limit of the density correlation function $G_{\rho\rho}(k,t)$ becomes nonzero, i.e., the frequency transform $G_{\rho\rho}(k,\omega)$ develops a $\delta(\omega)$ contribution. In the context of glassy dynamics [24–29], this form of MCT has been widely studied. The mode-coupling model follows from the equations of fluctuating nonlinear hydrodynamics [30] formulated in terms of the conserved density ρ and current density $\mathbf{g}$. Considerations of complete set of nonlinearities in the $\mathbf{g}$ equation for a compressible liquid, however, leads to the conclusion that the ENE transition is smoothed off [31]. The absence of the ENE transition has been treated in various subsequent works [32–38] predicting a decay of the time correlations over the longest time. In the second part of the present paper we consider the viability of the ENE transition in the Dean-Kawasaki model.

The paper is organized as follows: In the next section we provide a general introduction to the model studied here and summarize the main achievement of the present work. We construct the Martin-Siggia-Rose- (MSR) type field theory to take into account the role of all the nonlinearities in terms of self-energy matrix elements Σ. In Sec. IV we demonstrate how the renormalization of the nonlinear theory can be worked out in the hydrodynamic limit. In the next section we analyze the feasibility of an ENE transition in the present model. We finish the paper with a short discussion of the main results.

II. THE DEAN-KAWASAKI EQUATION

The collective densities $\tilde{\rho}$ and $\tilde{\mathbf{g}}$ in a many particle system is defined as

$$\tilde{\rho}(\mathbf{x},t) = \sum_{\alpha} \delta[\mathbf{x} - \mathbf{x}_\alpha(t)], \tag{1}$$

$$\tilde{\mathbf{g}}(\mathbf{x},t) = \sum_{\alpha} \mathbf{p}_\alpha \delta[\mathbf{x} - \mathbf{x}_\alpha(t)], \tag{2}$$

where $\{\mathbf{x}_\alpha,\mathbf{p}_\alpha\}$ are, respectively, the position and momentum of its constituent particles, and the sum is implied over all $\alpha = 1,\ldots,N$ particles. The tilde over the fields indicate that these are dependent on the microscopic or phase-space variables. The conservation of the total number of particles in the fluid gives rise to balance equation for the density $\tilde{\rho}(\mathbf{x},t)$. For very different types of microscopic dynamics of the fluid particles this balance equation can be written (with appropriate approximations) in a unique form. For a fluid in which the constituent particles follow Newton's equation, this is the continuity equation with the momentum current $\tilde{\mathbf{g}}$ as the flux and is time reversible. For the Brownian or stochastic dynamics of the particles this is a stochastic equation with noise. We consider these two cases separately.

A. Brownian dynamics

The time evolution of the constituent particles of the fluid is described by the Smoluchowski equations [39] which involve only the particle coordinates $\{\mathbf{x}_\alpha\}$. A colloidal system with heavy particles in a solution is a typical example for such a system. In the overdamped limit the time dependence of the momentum is ignored. The stochastic equation of motion for a particle is obtained as

$$\gamma_0 \frac{d\mathbf{x}_\alpha(t)}{dt} = -\sum_{\delta=1}^{N} \nabla_\alpha U[\mathbf{x}_\alpha(t) - \mathbf{x}_\delta(t)] + \boldsymbol{\xi}_\alpha(t), \tag{3}$$

where $U(\mathbf{x}_\alpha - \mathbf{x}_\delta)$ is the interaction between particles at $\mathbf{x}_\alpha$ and $\mathbf{x}_\delta$. The frictional drag force is γ_0 times the particle velocity. γ_0 is related to the correlation of the thermal noise $\boldsymbol{\xi}_\alpha$ as

$$\left\langle \xi_\alpha^i(t)\xi_\nu^j(t')\right\rangle = 2\gamma_0\beta^{-1}\delta_{\alpha\nu}\delta_{ij}\delta(t-t'), \tag{4}$$

where $\beta = 1/(k_B T)$. In the case of Smoluchowski dynamics the corresponding balance equation (3) for $\tilde{\rho}$ is exact [40,41]. The equation for the time evolution of the fluctuating density $\tilde{\rho}(\mathbf{x},t)$ is

$$\frac{\partial\tilde{\rho}(\mathbf{x},t)}{\partial t} = D_0\bigg[\nabla^2\tilde{\rho}(\mathbf{x},t) + \nabla \cdot \{\tilde{\rho}(\mathbf{x},t)\nabla\int dx'\tilde{U}(\mathbf{x}-\mathbf{x}')\tilde{\rho}(\mathbf{x}',t)\}\bigg] + \tilde{\eta}(\mathbf{x},t). \tag{5}$$

$D_0 = 1/(\beta\gamma_0)$ is the bare diffusion constant and $\tilde{U} = \beta U$ is the dimensionless interaction potential. In Eq. (5) we have defined the random force denoted by $\tilde{\eta}(\mathbf{x},t)$ as

$$\tilde{\eta}(\mathbf{x},t) = \sum_{\alpha=1}^{N} \nabla \cdot \{\delta(\mathbf{x}-\mathbf{x}_\alpha(t))\boldsymbol{\xi}_\alpha(t)\}, \tag{6}$$

with its correlation obtained from that of $\boldsymbol{\xi}_\alpha$ as stated in Eq. (4). The correlation of the noise $\tilde{\eta}(\mathbf{x},t)$ depends on the density $\tilde{\rho}(\mathbf{x},t)$, i.e, indicating that the noise is multiplicative. For a system of particles following Brownian dynamics, Eq. (5) is an exact representation of the evolution of the formally defined stochastic density field $\tilde{\rho}(x,t)$ [40]. The balance equation (5) is dissipative since the corresponding microscopic dynamics is also irreversible. However, since $\tilde{\rho}$ is a sum of delta functions, the equation is hardly of any use for being treated for constructing a field theory.

The collective density field $\rho(\mathbf{x},t)$ is the coarse-grained version of the $\tilde{\rho}$ averaged over the local equilibrium ensemble. The coarse-grained density $\rho(\mathbf{x},t)$ is defined as $\rho(\mathbf{x},t) = \langle\tilde{\rho}(\mathbf{x},t)\rangle_{\text{l.e}}$. The basic equation for the time evolution of the coarse-grained collective density, $\rho(\mathbf{x},t)$, is obtained by averaging this exact equation (5) for $\tilde{\rho}$ over the local equilibrium distribution. The stochastic partial differential equations for the coarse-grained density $\rho(\mathbf{x},t)$, obtained recently in Ref. [42], has smooth spatial and temporal dependence. On thermal averaging the dependence of the functional F on the bare interaction potential U is converted to that on the corresponding thermodynamic direct correlation functions in the coarse-grained equations,

$$\frac{\partial\rho}{\partial t} = D_0\nabla \cdot \left[\rho\nabla\frac{\delta F}{\delta\rho}\right] + \eta. \tag{7}$$

The correlation of the coarse-grained multiplicative noise $\eta(\mathbf{x},t)$ which is the average $\tilde{\eta}(\mathbf{x},t)$ is obtained in the form

$$\langle\eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = 2\beta^{-1}D_0\nabla\rho(\mathbf{x},t)\cdot\nabla'\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{8}$$

The coarse-grained equation involves the free-energy functional $F[\rho]$ which is expressed as a sum of two parts,

$$F = F_{\text{id}}[\rho] + F_{\text{in}}[\rho]. \tag{9}$$

The ideal gas part $F_{\rm id}$ [43] and interaction part $F_{\rm in}$ are respectively obtained as:

$$F_{\rm id}[\rho] = \int d\mathbf{x}\rho(\mathbf{x},t)\left[\ln\frac{\rho(\mathbf{x},t)}{\rho_0} - 1\right], \tag{10}$$

$$F_{\rm in}[\rho] = -\frac{1}{2}\int d\mathbf{x}\int d\mathbf{x}'\delta\rho(\mathbf{x},t)c(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}',t), \tag{11}$$

in terms of the density fluctuations $\delta\rho(\mathbf{x},t) = \rho(\mathbf{x},t) - \rho_0$ around the equilibrium density ρ_0. In the last equation $c(\mathbf{x})$ is the two-point direct correlation function defined in the Ornstein-Zernike relation [44]. Here we have truncated an infinite expansion in density fluctuations with coefficients given by the successive direct correlation functions defined [45] as

$$c^{(i)}(\mathbf{x}_1,\ldots,\mathbf{x}_i;[\rho]) = -\beta\frac{\delta^i F_{\rm in}[\rho]}{\delta\rho(\mathbf{x}_1)\ldots\delta\rho(\mathbf{x}_i)}. \tag{12}$$

B. Newtonian dynamics

For the fluid in which the constituent particles follow the Newtonian dynamics, the corresponding *exact* balance equations for the conserved densities are the reversible Euler equations. In this case the coarse-grained equations motion is obtained for both the mass and the momentum densities $\{\rho,\mathbf{g}\}$. The corresponding equations of fluctuating nonlinear hydrodynamics [46] are treated at a simpler level using the overdamped limit or in the so-called adiabatic approximation. By integrating out the momentum field $\mathbf{g}$ from the field-theoretical model with the set $\{\rho,\mathbf{g}\}$, a stochastic Langevin equation involving only the coarse-grained density $\rho(x,t)$ is reached [47]. The adiabatic approximation assumes that the momentum-density fluctuations relaxes over much shorter time scales than the density fluctuations. As a result of eliminating the momentum density $\mathbf{g}$, the $1/\rho$ nonlinearity in the momentum-density equation disappears. Instead, the equation for ρ has multiplicative noise. The resulting equation for the coarse-grained density $\rho(\mathbf{x},t)$ in the Newtonian dynamics case is identical to Eq. (7). The stochastic partial differential Eq. (7) for the coarse-grained density $\rho(\mathbf{x},t)$ is therefore *approximate* for both kinds of particle dynamics. The coarse-grained equation (7) will be referred to as the Dean-Kawasaki equation.

III. THE MSR FIELD THEORY

In this section we outline the computation of time correlation functions corresponding to a set of field $\{\psi_\alpha\}$ whose dynamics is described by the nonlinear equation of motion

$$\frac{\partial\psi_\alpha}{\partial t} + L^0_{\alpha\beta}\frac{\delta F}{\delta\psi_\beta} = \vartheta_\alpha. \tag{13}$$

F denotes the free-energy functional determining the equilibrium state. We have chosen here a purely dissipative form of the equation of motion involving only the transport matrix $L^0_{\alpha\beta}$. ϑ_α represents the random Gaussian (white) noise whose correlation is the matrix $L^0_{\alpha\beta}$,

$$\langle\vartheta_\alpha(\mathbf{x},t)\vartheta_\beta(\mathbf{x}',t')\rangle = 2\beta^{-1}L^0_{\alpha\beta}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'). \tag{14}$$

We use here the standard MSR field-theoretic approach [48,49] with the functional integral formulation [50–53] to study the renormalized model. We obtain perturbative corrections to the bare transport coefficient D_0 due to the nonlinearities. The matrix $\mathbf{G}$ of two-point correlations between the various fields, respectively, at points denoted as 1 and 2 for both space and time coordinates, includes the correlation functions $G_{\alpha\beta}$ and the response functions $G_{\alpha\hat{\beta}}$. These are respectively defined as

$$G_{\alpha\beta}(12) = \langle\psi_\alpha(1)\psi_\beta(2)\rangle, \tag{15}$$

$$G_{\alpha\hat{\beta}}(12) = \langle\psi_\alpha(1)\hat{\psi}_\beta(2)\rangle. \tag{16}$$

The Greek letter subscripts refer to the set of physical fields $\{\psi_\alpha\}$ and their respective hatted counterparts as $\{\hat{\psi}_\alpha\}$ of MSR theory. The averages are functional integrals over all the fields weighted by $\exp[-\mathcal{A}]$, where the action functional $\mathcal{A}$ is obtained [46] using the equations of motion:

$$\begin{aligned}\mathcal{A}[\psi,\hat{\psi}] &= \int dt\int d\mathbf{x}\Bigg[2k_BT\sum_{\alpha,\beta}\hat{\psi}_\alpha L^0_{\alpha\beta}\hat{\psi}_\beta \\ &\quad + i\sum_\alpha\hat{\psi}_\alpha(\mathbf{x},t)\left[\frac{\partial\psi_\alpha}{\partial t} + L^0_{\alpha\beta}\frac{\delta F}{\delta\psi_\beta}\right]\Bigg] \\ &\equiv \mathcal{A}_0[\psi,\hat{\psi}] + \mathcal{A}_I[\psi,\hat{\psi}].\end{aligned} \tag{17}$$

The parts $\mathcal{A}_0$ and $\mathcal{A}_I$ shown in the last equality respectively denote the Gaussian (quadratic in the fields) and non-Gaussian parts of the MSR-action functional. Averages obtained with only the Gaussian part of the action will be denoted with a subscript 0 to separate them from their respective counterparts representing the fully nonlinear dynamics. Correction to (the inverse of) the zeroth-order Green's function G_0 is obtained in terms of the self-energy matrix Σ through the Dyson equation,

$$\mathbf{G}^{-1}(1,2) = \mathbf{G}_0^{-1}(1,2) - \Sigma(1,2). \tag{18}$$

If there is an external field h_α conjugate to field ψ_α present in the equation of motion, then the correlation function $G_{\mu\hat{\alpha}}$ represents the response of fluctuation of ψ_μ linear to variation of h_α. The MSR field-theoretic model presented above maintains fluctuation dissipation relations (FDR) [46,54,55] between the correlation functions and response functions. Generally, using the time-translational invariance properties of the action, the FDR between correlation and response functions involving the field $\hat{\psi}$ and φ are obtained. These transformations are written in terms of the field $\psi_{\rm i}$ as

$$\psi_i(x,-t) \rightarrow \epsilon_i\psi_i(x,t), \tag{19}$$

$$\hat{\psi}_i(x,-t) \rightarrow -\epsilon_i\left[\hat{\psi}_i(x,t) - i\beta\frac{\delta \mathrm{F}}{\delta\psi_i(x,t)}\right], \tag{20}$$

where ϵ_i gives the transformation of ψ_i under time reversal. In Ref. [56], the general set of transformation rules like (19) and (20) were identified from which the FDR's follow in a very natural way. Both the Gaussian as well as the non-Gaussian parts of the action remain separately invariant under the above set of transformations. The FDR between correlation and response functions involving the field $\hat{\psi}$ and φ are obtained in the form:

$$G_{\psi_f\varphi}(\mathbf{q},\omega) = -2\beta^{-1}\mathrm{Im}G_{\hat{\psi}\varphi}(\mathbf{q},\omega), \tag{21}$$

where we have defined

$$\psi_f(\mathbf{x}) = \frac{\delta F[\psi]}{\delta\psi(\mathbf{x})}. \tag{22}$$

The FDR's in the MSR field theory play an important role in the renormalization of linear theory. As we can see, the general fluctuation-dissipation relations relations are not linear in the fields. If the driving free-energy functional is purely Gaussian [33], then the available FDR's given by Eq. (21) are linear in the fields $\{\psi_\alpha,\hat{\psi}_\alpha\}$. For a non-Gaussian driving free-energy functional F, we need to extend the space of fields beyond $\{\psi,\hat{\psi}\}$ to maintain the linear FDR. In this case the definition (22) gives rise to nonlinear constraint in the model. The fluctuation dissipation relations (21) play a key role in the analysis of the present paper.

IV. BACKGROUND AND MAIN RESULTS: AN OVERVIEW

In 1998 Kawasaki and Miyazima (KM) developed [47] a MSR field-theoretic model involving the coarse-grained density $\rho(\mathbf{x},t)$ and its hatted counterpart $\hat{\rho}(\mathbf{x},t)$. The theory obtained the renormalized correlation functions corresponding to the Dean-Kawasaki nonlinear stochastic equation (7). KM analyzed the possibility of an ENE transition in the supercooled liquid due to mode-coupling effects in their model. At the ENE transition the density correlation function $G_{\rho\rho}(q,t)$ is nonzero for long times t, i.e., the corresponding Fourier transform develops a singular $\delta(\omega)$ part. Due to feedback effects from strongly coupled density fluctuations, the renormalized diffusion constant D_R is zero beyond a critical density leading to the ENE transition. The 2×2 matrix structure of two-point functions $\mathbf{G}$ in the KM model with $\{\rho,\hat{\rho}\}$ fields includes both correlation and response functions. The following linear FDR linking the (Fourier transformed) MSR response function $G_{\rho\hat{\rho}}(k,t)$ and the density correlation function $G_{\rho\rho}(k,t)$ was assumed:

$$G_{\rho\hat{\rho}}(k,t) = i\Theta(t)\tilde{S}^{-1}(k)(k)G_{\rho\rho}(k,t), \tag{23}$$

where $\Theta(t)$ is the Heavside step function [57]. The above FDR requires us to adopt the following relation between the so-called correlation ($\Sigma_{\hat{\rho}\hat{\rho}}$) and response ($\Sigma_{\hat{\rho}\rho}$) elements of the self-energy matrix Σ:

$$\tilde{S}^{-1}(k)\Sigma_{\hat{\rho}\hat{\rho}} = 2\Sigma'_{\hat{\rho}\rho}. \tag{24}$$

The ENE transition scenario in the KM model is illustrated with the Laplace transform of the density correlation function:

$$G_{\rho\rho}(q,z) = \frac{\tilde{S}(q)}{z + iq^2 D_R(q,z)}, \tag{25}$$

where $D_R(q,z)$ is the Laplace transform of the renormalized kinetic coefficient. The FDR's (23) and (24) were used [47] by KM to obtain the renormalized diffusion constant D_R. Due to the mode-coupling effects the renormalized quantity $D_R(q,z) \sim z \to 0$ for small z, leading self-consistently to the result that the long-time limit of the two-point density correlation is nonzero beyond the ENE transition.

In subsequent works it, however, emerged that there are subtle problems with the construction of the renormalized theory of the KM model. The relation (23) between $G_{\rho\rho}$ and $G_{\rho\hat{\rho}}$ presented there does not hold. To study the full implications of the nonlinearities in the Dean-Kawasaki equation, the proper FDR's linking the correlation and response functions in the model have to be considered. The general form of the FDR for the MSR theory given in Eq. (21) indicates that we need to construct the field theory with a larger set of fields. This also gives rise to a nonlinear constraint to be explained in the next section. The simple relation between correlation and response-type self-energies [see Eq. (24)] used in Ref. [47] is also now modified.

The primary results of the present paper are as follows:

(1) The nature of relaxation of the renormalized correlation functions is determined from analysis of the self-energy matrix, introduced in the Dyson equation (18). As explained above instead of the linear FDR relations of (23) and (24) used in Ref. [47], one needs to formulate the MSR field theory with a larger set of variables. Using the full set of FDR's, linear within the extended set of fields [58], we show here that the renormalized diffusion constant D_R is still obtained from the self-energy element $\Sigma_{\hat{\rho}\hat{\rho}}$ as originally proposed by Kawasaki and Miyazima [47]. This is plausible since the nonlinear Dean-Kawasaki equation is unchanged and there is no new density nonlinearity introduced with the new formulation with extended set of slow modes and constraints. The latter helps a better reorganization of the perturbation theory through linear FDR's. Using the so-called Kawasaki rearrangement scheme we obtain the renormalized diffusion constant D_R as a self-consistent functional of the correlation functions. As the decay of the density correlations slow down, D_R is sharply reduced and the corresponding diffusion process is increasingly slowed down, which in the present model can be understood using the feedback mechanism of MCT.

(2) We analyze from a nonperturbative approach if the Dean-Kawasaki dynamics supports an ENE transition. In the present analysis, the extension of the set of field variables brings in a nonlinear constraint which includes an infinite number of density nonlinearities with a single new variable θ. In our analysis we identify a key element of the self-energy matrix, $\Sigma_{\hat{\theta}\rho}$, which arises from this nonlinear constraint. We show that if this self-energy matrix would be zero, then the result of KM is recovered, i.e., the ENE transition is supported in the Dean-Kawasaki model. In the general case in which the crucial self-energy element is nonzero, we obtain from a nonperturbative approach that a $\delta(\omega)$ contribution in the density correlation functions is *not* self-consistent with the renormalized transport coefficient $D_R(q,\omega)$ going to zero in the small-frequency limit. The density correlations therefore decay to zero over long time, thus implying that the Dean-Kawasaki model does not support a sharp ENE transition.

V. THE NONLINEAR CONSTRAINT

Let us first consider the extension of the fields so the FDR's in the Dean-Kawasaki model are linear within the bigger set. The available FDR relations in this model involve the functional derivative of the free energy $F[\rho]$ with respect to density ρ,

$$\begin{aligned}\frac{\delta F[\rho]}{\delta\rho(\mathbf{x},t)} &= \frac{\delta F_{\rm id}(\rho)}{\delta\rho(\mathbf{x},t)} + \frac{\delta F_{\rm in}(\rho)}{\delta\rho(\mathbf{x},t)} \\ &= \ln\frac{\rho(\mathbf{x},t)}{\rho_0} - \int d\mathbf{x}' c(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}',t) \\ &\equiv \int \mathcal{S}(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}')d\mathbf{x}' + \theta(\mathbf{x},t). \end{aligned} \tag{26}$$

The first term on right-hand side of Eq. (26) denotes the convolution of the function $\mathcal{S}(\mathbf{x})$ and the density fluctuation $\delta\rho$. The Fourier transform of $\mathcal{S}(\mathbf{x})$ is $[1-\rho_0 c(q)]/\rho_0$, whose inverse is $\rho_0 S(q)$, where $S(q)$ is the static structure factor of the fluid. In the following we denote $\rho_0 S(q) \equiv \tilde{S}(q)$, which is the equal time correlation of density fluctuations or $G_{\rho\rho}(q,t=0)$. The new variable $\theta(\mathbf{x},t)$ is defined as the nonlinear terms of the expansion of $\ln[\rho(\mathbf{x},t)/\rho_0]$ in Eq. (10),

$$\theta(\mathbf{x},t) = -\sum_{n=2}^{\infty}\frac{1}{n}\left[-\frac{\delta\rho(\mathbf{x},t)}{\rho_0}\right]^n \equiv f[\delta\rho(\mathbf{x},t)]. \tag{27}$$

Equation (7) now reduces to the following nonlinear diffusion equation for the coarse-grained density fluctuations with couplings to the field $\theta(\mathbf{x},t)$:

$$\frac{\partial\rho}{\partial t} = D_0\nabla\left[\rho(\mathbf{x},t)\left\{\nabla\int \mathcal{S}(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}')d\mathbf{x}' + \nabla\theta(\mathbf{x},t)\right\}\right] + \eta(\mathbf{x},t). \tag{28}$$

Equations (28) and the constraint (27) together constitute a close set of equations for the density fluctuations. The introduction of the variable $\theta(\mathbf{x},t)$ with respect to the nonlinear part of the functional derivative of $F_{\rm id}[\tilde{\rho}]$ does not change the Dean-Kawasaki equation. The contribution from the ideal gas part remains $-D_0\nabla^2\rho(\mathbf{x},t)$. However, with the extended set $\{\rho,\theta\}$ the available FDR's in the model are linear in terms of the fields. We now construct the corresponding MSR field theory to compute self-consistently the correction to the linear diffusion coefficient D_0 as a result of the nonlinear coupling of fields in Eq. (28).

The set of physical fields and their respective hatted counterparts are, respectively, denoted here as $\{\psi,\hat{\psi}\} \equiv \{\rho,\theta,\hat{\rho},\hat{\theta}\}$. The averages are functional integrals over all the fields weighted by $\exp[-\mathcal{A}]$ where the action functional $\mathcal{A}$ is obtained [46] using the equations of motion for the ρ, as well as the nonlinear constraint of (27). The MSR action functional with the non-Gaussian free energy F in Eq. (9) is obtained as:

$$\begin{aligned}\mathcal{A}[\psi,\hat{\psi}] = \int dt \int d\mathbf{x}\Big[&2D_0\rho(\mathbf{x},t)[\nabla\hat{\rho}(\mathbf{x},t)]\cdot\nabla\hat{\rho}(\mathbf{x},t) \\ &+ i\hat{\theta}(\mathbf{x},t)\{\theta(\mathbf{x},t) + f[\delta\rho(\mathbf{x},t)]\} \\ &+ i\hat{\rho}(\mathbf{x},t)\Big[\frac{\partial\rho}{\partial t} - D_0\nabla\cdot\Big(\rho(\mathbf{x},t)\nabla \\ &\times\Big\{\int d\mathbf{x}'\mathcal{S}(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}') + \theta(\mathbf{x},t)\Big\}\Big)\Big]\Big]. \end{aligned} \tag{29}$$

A. Linear fluctuation-dissipation relations

In the present case the set of linear transformations which keeps the MSR action functional $\mathcal{A}[\psi,\hat{\psi}]$ given in Eq. (29) invariant is given by

$$\rho(\mathbf{x},-t) = \rho(\mathbf{x},t), \tag{30}$$

$$\hat{\rho}(\mathbf{x},-t) = -\hat{\rho}(\mathbf{x},t) + i\int \mathcal{S}(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}',t) + i\theta(\mathbf{x},t), \tag{31}$$

$$\theta(\mathbf{x},-t) = \theta(\mathbf{x},t), \tag{32}$$

$$\hat{\theta}(\mathbf{x},-t) = \hat{\theta}(\mathbf{x},t) + i\frac{\partial}{\partial t}\rho(\mathbf{x},t). \tag{33}$$

The following set of FDR's holds in the nonlinear theory are obtained from the above invariant properties of the MSR action. We write them in terms of the spatial Fourier transform as

$$G_{\rho\hat{\rho}}(\mathbf{q},t) = i\Theta(t)[\tilde{S}^{-1}(\mathbf{q})G_{\rho\rho}(\mathbf{q},t) + G_{\rho\theta}(\mathbf{q},t)], \tag{34}$$

$$G_{\theta\hat{\rho}}(\mathbf{q},t) = i\Theta(t)[\tilde{S}^{-1}(\mathbf{q})G_{\theta\rho}(\mathbf{q},t) + G_{\theta\theta}(\mathbf{q},t)], \tag{35}$$

$$G_{\rho\hat{\theta}}(\mathbf{q},t) = i\Theta(t)\frac{\partial}{\partial t}G_{\rho\rho}(\mathbf{q},t), \tag{36}$$

$$G_{\theta\hat{\theta}}(\mathbf{q},t) = i\Theta(t)\frac{\partial}{\partial t}G_{\theta\rho}(\mathbf{q},t) + i\delta(t)[\tilde{S}^{-1}(\mathbf{q})G_{\rho\rho}(\mathbf{q},t) + G_{\theta\rho}(\mathbf{q},t)]. \tag{37}$$

Equations (34)–(37) are used below to analyze the renormalization of the linear theory due to the nonlinear couplings of density fluctuations in the Dean-Kawasaki equation. The above-listed FDR's were reported earlier in Ref. [58] except for the second term on the right-hand side of the relation (37) involving the $\delta(t)$. This term was missed in Ref. [58]. This correction, as we will see below, has severe implications on the renormalizability of the theory.

VI. RENORMALIZED DYNAMICS

We develop, using the MSR approach, outlined in the earlier section, the field theory to compute the time correlation functions of fluctuations of the density ρ. This is done in two parts in the following. First, we compute the zeroth-order correlation functions by keeping only the Gaussian part $\mathcal{A}_0$ of the MSR action functional $\mathcal{A}$ introduced in Eq. (29). Next, the corrections due to the non-Gaussian part of the action $\mathcal{A}_I$ is obtained in a perturbation theory using a diagrammatic approach.

A. Correlations in the Gaussian model

The Gaussian part $\mathcal{A}_0$ of the action functional (29) is obtained by keeping only quadratic terms in the fluctuations of the fields,

$$\begin{aligned}\mathcal{A}_0[\psi,\hat{\psi}] = \int dt\int d\mathbf{x}\Big[&2D_0\rho_0\{\nabla\hat{\rho}(\mathbf{x},t)\}\cdot\nabla\hat{\rho}(\mathbf{x},t) \\ &+ i\hat{\theta}(\mathbf{x},t)\theta(\mathbf{x},t) + i\hat{\rho}(1)\Big\{\frac{\partial\rho(\mathbf{x},t)}{\partial t} \\ &- D_0\Big(\nabla^2\int \mathcal{S}(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x}')d\mathbf{x}' \\ &+ \rho_0\nabla^2\theta(\mathbf{x},t)\Big)\Big\}\Big]. \end{aligned} \tag{38}$$

TABLE I. Elements of matrix $\mathcal{B}_0$ used in defining the response parts of $\mathbf{G}_0^{-1}$.

	ρ	θ
$\hat{\rho}$	$(\omega + i\tilde{D}_0 q^2)$	$i\bar{D}_0 q^2$
$\hat{\theta}$	0	i

The above form is written in a matrix form using the matrix notation for the field $\psi(\mathbf{x},t)$ as $\Psi(1)$ as

$$\mathcal{A}_0 = \int d1 \int d2 \Psi(1) G_0^{-1}(12)\Psi(2), \tag{39}$$

where 1 stands for both time and space coordinates as well as the different fields like ρ and θ and their hatted counterparts in the MSR theory. The inverse matrix $\mathbf{G}_0^{-1}$ is obtained in the block diagonal form

$$\mathbf{G}_0^{-1} = \begin{bmatrix} \bigcirc & \mathcal{B}_0^\dagger \\ \mathcal{B}_0 & \mathcal{C}_0 \end{bmatrix}. \tag{40}$$

The matrix $\mathcal{B}_0$ corresponding to the response parts of the zeroth-order matrix $\mathbf{G}_0$ is listed in Table I. $\mathcal{B}_0^\dagger$ is the Hermitian conjugate of $\mathcal{B}_0$. In writing the elements of the $\mathcal{B}_0$ matrix we used two scaled forms of the bare diffusion constant D_0. First, $\tilde{D}_0 = D_0/S(q)$ and, second, $\bar{D}_0 = \rho_0 D_0$. The correlation part $\mathcal{C}_0$ of $\mathbf{G}_0^{-1}$ involving the hatted fields for both the indices is obtained as

$$[\mathcal{C}_0]_{\hat{\alpha}\hat{\beta}} = 2\bar{D}_0 q^2 \delta_{\alpha\beta}\delta_{\alpha\rho}. \tag{41}$$

The only nonzero correlation function at the zeroth order with Gaussian level action is obtained as

$$G^0_{\rho\rho}(q,\omega) = \frac{2\bar{D}_0 q^2}{\omega^2 + \tilde{D}_0^2 q^4}. \tag{42}$$

The pole of the corresponding Laplace transformed quantity $G^0_{\rho\rho}(q,z)$ represents a diffusive mode [59].

B. Renormalization

The field-theoretic method outlined in the previous section is now applied toward understanding the full implications of the nonlinearities in the Dean-Kawasaki equation. Analysis of the available FDR's in the model proves to be useful in obtaining the renormalization of the linear theory in a nonperturbative manner. We add a correction to (the inverse of) the zeroth-order Green's function G_0 in terms of the self-energy matrix Σ defined in Dyson's equation (18). The inverse of the full correlation matrix G is obtained in the form

$$G^{-1} = \begin{bmatrix} \bigcirc & \mathcal{B}^\dagger \\ \mathcal{B} & \mathcal{C} \end{bmatrix}. \tag{43}$$

We identify the key elements of the self-energy matrix Σ in terms of which the full implications of the vertices arising from the non-Gaussian part $\mathcal{A}_I$ of the MSR action functional $\mathcal{A}$ are obtained. After writing the appropriate elements of the Σ matrix in the leading order in wave number, the $\mathbf{G}^{-1}$ is obtained in the block form in terms of the 2×2 matrices $\mathcal{B}$ and $\mathcal{C}$. The latter are as listed respectively in Tables II and III. Corrections to the renormalized diffusion coefficients $\tilde{D}_R$ and $\bar{D}_R$ are, respectively, obtained as

TABLE II. Response elements $\mathcal{B}$ of matrix $G^{-1}_{\hat{\alpha}\beta}$ in terms of renormalized coefficients.

	ρ	θ
$\hat{\rho}$	$\omega + iq^2\tilde{D}_R$	$i\bar{D}_R q^2$
$\hat{\theta}$	$-\Sigma_{\hat{\theta}\rho}$	$(i - \Sigma_{\hat{\theta}\theta})$

$$\tilde{D}_R = \tilde{D}_0 + iq^{-2}\Sigma_{\hat{\rho}\rho}, \tag{44}$$

$$\bar{D}_R = \bar{D}_0 + iq^{-2}\Sigma_{\hat{\rho}\theta}, \tag{45}$$

Taking the inverse of the matrix in Eq. (43), the full matrix $\mathbf{G}$ consisting of the correlation and response functions are obtained. The elements of the full-response function matrix are expressed in terms of $N_{\alpha\hat{\beta}}(\mathbf{q},\omega)$ as

$$G_{\alpha\hat{\varphi}} = \frac{N_{\alpha\hat{\varphi}}}{\mathcal{D}}, \tag{46}$$

with $\mathcal{D}$ being the determinant of the matrix $\mathcal{B}$. For the latter we obtain

$$\mathcal{D}(\mathbf{q},\omega) = (\omega + iq^2\tilde{D}_R)\{i - \Sigma_{\hat{\theta}\theta}\} + i\bar{D}_R q^2 \Sigma_{\hat{\theta}\rho}. \tag{47}$$

The matrix $N_{\alpha\hat{\beta}}$ is provided in Table IV. The correlation functions between the unhatted fields are obtained from the relation

$$G_{\alpha\beta}(q,\omega) = -G_{\alpha\hat{\gamma}}(q,\omega)\mathcal{C}_{\hat{\gamma}\hat{\delta}}(q,\omega)G_{\hat{\delta}\beta}(q,\omega), \tag{48}$$

with the matrix $\mathcal{C}_{\hat{\gamma}\hat{\delta}}$ being provided in Table III.

C. Analysis of the FDR's

We consider the FDR's (34) and (35) involving the $\hat{\rho}$ field in the following compact form:

$$\tilde{S}^{-1}(\mathbf{q})G_{\rho\alpha}(\mathbf{q},\omega) + G_{\theta\alpha}(\mathbf{q},\omega) = -2\mathrm{Im}[G_{\hat{\rho}\alpha}(\mathbf{q},\omega)], \tag{49}$$

where $\alpha \in \{\rho,\theta\}$. Using the definitions (46)–(48), respectively, for the response and correlation functions, we obtain

$$\sum_{\hat{\gamma}} Z_{\rho\hat{\gamma}} C_{\hat{\gamma}\hat{\delta}} = -i\left(\mathcal{D}\delta_{\hat{\rho}\hat{\delta}} + \sum_{\alpha} N_{\hat{\rho}\alpha} G^{-1}_{\alpha\hat{\delta}}\right)$$

with the $Z_{\rho\hat{\gamma}}$'s being defined as

$$Z_{\rho\hat{\gamma}} = \tilde{S}^{-1}(\mathbf{q})N_{\rho\hat{\gamma}} + N_{\theta\hat{\gamma}}. \tag{50}$$

Substituting the results for the various elements of the self-energy matrix Σ in the above relations involving the Z's, we obtain in Appendix A a set of useful relations. These relations connect the elements of the response part of the self-energy $(\Sigma_{\psi\hat{\psi}})$ with the corresponding elements from the correlation

TABLE III. Correlation elements $\mathcal{C}$ of matrix $G^{-1}_{\hat{\alpha}\hat{\beta}}$.

	$\hat{\rho}$	$\hat{\theta}$
$\hat{\rho}$	$2\bar{D}_0 q^2 - \Sigma_{\hat{\rho}\hat{\rho}}$	$-\Sigma_{\hat{\rho}\hat{\theta}}$
$\hat{\theta}$	$-\Sigma_{\hat{\theta}\hat{\rho}}$	$-\Sigma_{\hat{\theta}\hat{\theta}}$

TABLE IV. Elements of matrix $N_{\alpha\hat{\beta}}$ in terms of the renormalized transport coefficient $\tilde{D}_R$ and $\bar{D}_R$.

	$\hat{\rho}$	$\hat{\theta}$
ρ	$(i - \Sigma_{\hat{\theta}\theta})$	$-i\bar{D}_R q^2$
θ	$\Sigma_{\hat{\theta}\rho}$	$(\omega + iq^2\tilde{D}_R)$

part of the self-energy ($\Sigma_{\hat{\psi}\hat{\psi}}$). These relations further simplify in the hydrodynamic limit of small q and z, justifying the renormalization of the model. In Appendix A we simplify the Eqs. (50) by substituting the limiting forms for the Z's in the hydrodynamic limit. After some tedious algebra we obtain the following relations to leading order in wave numbers:

$$2\gamma'_{\hat{\rho}\theta} = -\gamma_{\hat{\rho}\hat{\rho}} + \Delta_R, \tag{51}$$

$$2\tilde{S}\gamma'_{\hat{\rho}\rho} = -\gamma_{\hat{\rho}\hat{\rho}} - \Delta_R \tilde{S}\gamma'_{\hat{\theta}\rho}, \tag{52}$$

where we denote as $\Delta_R = \gamma'_{\hat{\theta}\hat{\rho}}/\tilde{\gamma}_{\theta\rho}$. Using these relations we simplify the expression (47) for the denominator $\mathcal{D}$ which defines the response functions in Eq. (46). In Appendix B we obtain the result

$$\mathcal{D}(\mathbf{q},z) = i[z + iq^2 D_R(q,z)\zeta(q,z)]. \tag{53}$$

The renormalized diffusion coefficient D_R is to be obtained from the correlation self-energy $\gamma_{\hat{\rho}\hat{\rho}}$ through the relation:

$$D_R(q,\omega) = D_0 - (2\rho_0)^{-1}\gamma_{\hat{\rho}\hat{\rho}}(q,\omega). \tag{54}$$

The quantity $\zeta(q,\omega)$ appearing in the expression for $\mathcal{D}$ is a sum of two parts, $\zeta_0(q)$ and $\zeta_{\rm mc}(q,\omega)$ respectively representing the bare and mode-coupling contributions:

$$\zeta(q,\omega) = \zeta_0(q) + \zeta_{\rm mc}(q,\omega). \tag{55}$$

The bare part is obtained as $\zeta_0(q) = \tilde{S}^{-1}(q)$ and is present even in the absence of any nonlinearity in the model. For the linear dynamics this makes the effective diffusion constant D_0 become renormalized by the structure factor $S(q)$, $\tilde{D}_0 = D_0/S(q)$ as already shown in the previous subsection. The mode-coupling part $\zeta_{\rm mc}(q,z)$ in the defining relation (55) is expressed in terms of the self-energy $\gamma_{\hat{\theta}\rho}$ involving the field $\hat{\theta}$.

$$\zeta_{\rm mc}(q,z) = -i\rho_0\gamma_{\hat{\theta}\rho}(q,z). \tag{56}$$

Therefore $\zeta_{\rm mc}$ is a direct consequence of the nonlinear constraint (27) in the model. If the constraint is absent, it reduces $\zeta_{\rm mc}$ to zero.

D. Renormalized correlation functions

The self-energy $\gamma_{\hat{\rho}\hat{\rho}}$ in $D_R(q,z)$ is expressed in a series representing the order-by-order expansion in terms of the vertex functions which appear in the non-Gaussian part $\mathcal{A}_I$ of the MSR action functional (29). This series can be organized in terms of the one-particle irreducible contributions using what is termed the Kawasaki rearrangement [47,60] in the form

$$D_R(q,z) = \equiv \frac{D_0}{1 + \Delta_{\rm mc}(q,z)}. \tag{57}$$

The quantity $\Delta_{\rm mc}$ represents the mode-coupling effects which at the lowest order are expressed in terms of one-loop diagrams. The diagrams are constructed using standard techniques [15]. In the Appendix we provide the evaluation of the one-loop diagrams for the relevant self-energies. With the cubic vertices in $\mathcal{A}_I$, we obtain that at the one-loop order $\Delta_{\rm mc}$ contains the bubble-shaped diagrams with a convolution of two correlation functions. Detailed calculation with the various one-loop diagrams is provided in Appendix B. The relevant diagrams for the self-energy $\Sigma_{\hat{\rho}\hat{\rho}}$ is displayed in Fig. 1. The one-loop expression involves products of the correlations functions $\{G_{\rho\rho}, G_{\rho\theta}, G_{\theta\theta}\}$,

$$\begin{aligned}\Delta_{\rm mc}(q,t) = \frac{D_0}{2\rho_0}\int \frac{d\mathbf{k}}{(2\pi)^3}&[{\rm V}_1(\mathbf{q},\mathbf{k})G_{\theta\rho}(k,t)G_{\rho\theta}(q-k,t)\\ &+ G_{\rho\rho}(q-k,t)\{{\rm V}_2(\mathbf{q},\mathbf{k})G_{\rho\rho}(k,t)\\ &+ {\rm V}_3(\mathbf{q},\mathbf{k})(G_{\rho\theta}(k,t) + G_{\theta\rho}(k,t))\\ &+ {\rm V}_4(\mathbf{q},\mathbf{k})G_{\theta\theta}(k,t)\}].\end{aligned} \tag{58}$$

The expressions for the four vertex functions ${\rm V}_i(\mathbf{q},\mathbf{k})$ for $i = 1,\ldots,4$, obtained from the analysis of the one-loop diagrams, are listed in Appendix B. If the correlation functions develop a nondecaying part at the ENE transition, then the denominator of the right-hand side of (57) has the singular behavior $\Delta_{\rm mc}(z) \sim 1/z$ and hence $D_R(z) \sim z$ in the low-frequency limit. In the next section we use this result to analyze the viability of an ENE transition in the liquid at metastable densities.

Substituting in the right-hand side of (46) the results for the matrix elements $N_{\alpha\hat{\beta}}$ and the determinant $\mathcal{D}$ respectively from Table IV and Eq. (47), we obtain the following renormalized

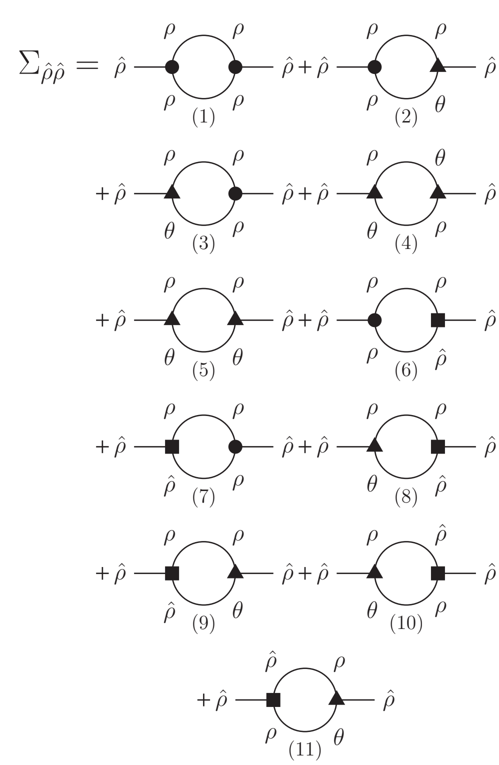

FIG. 1. One-loop diagrams for the self-energy matrix element $\Sigma_{\hat{\rho}\hat{\rho}}$. The different types of vertices $V_{\hat{\theta}\rho\rho}$, $V_{\hat{\rho}\rho\rho}$, and $V_{\hat{\rho}\theta\rho}$ appearing in the MSR action (29) are marked in the diagrams as filled squares, circles, and triangles, respectively.

expressions for the two relevant response functions:

$$G_{\rho\hat{\rho}}(q,z) = \frac{1}{z + iq^2 D_R(q,z)\zeta(q,z)}, \quad (59)$$

$$G_{\theta\hat{\rho}}(q,z) = \frac{i\zeta_{\rm mc}(q,z)}{z + iq^2 D_R(q,z)\zeta(q,z)}. \quad (60)$$

The above results for the response functions together with the expression (48) are useful in analyzing the long-distance long-time properties of the correlation functions in terms of the renormalized transport coefficient $D_R(q,z)$. The quantity ζ introduced above also plays a crucial role. The pole in the Laplace transform of the correlation functions determined from the zero of $\mathcal{D}$ at

$$z = -iq^2 D_R(q,z)\zeta(q,z). \quad (61)$$

VII. THE DYNAMIC TRANSITION OF MCT

In this section we analyze, using a non perturbative approach [31], whether the ENE transition can occur in the model for which dynamics of the density fluctuations is described by the nonlinear Dean-Kawasaki equation. The transition is characterized by the corresponding long-time limit of the density correlation function changing to a nonzero value. We focus primarily on the viability of the basic feedback mechanism of MCT [25] in the present model. More specifically, we address the question whether it is self-consistent to have a singular $\delta(\omega)$ contribution in the Fourier transforms of the correlation functions $G_{\rho\rho}$ and $G_{\rho\theta}$ while the corresponding Laplace transformed renormalized transport coefficient $D_R(q,\omega)\to 0$ for small frequencies. We begin by noting the following two links between correlation functions and generalized transport coefficients obtained in the previous sections.

Link A: The mode-coupling contributions in the renormalized transport coefficient $D_R(q,z)$ are expressed in terms of convolutions of the correlation functions $G_{\rho\rho}$ or $G_{\rho\theta}$. One-loop expressions are provided in Eq. (58). At the ENE transition the time-dependent correlation functions remain nonzero at long times and hence their corresponding Laplace transforms develop a $1/z$ pole. It follows from the relations (57) that at the transition, the renormalized diffusion constant $D_R(q,z) \sim z$ vanishes for small frequencies.

Link B: The defining relations of the correlation functions $G_{\rho\rho}$ or $G_{\rho\theta}$ are in terms of the renormalized transport coefficient $D_R(q,z)$ as stated in Eqs. (46)–(48). Equation (48) gives the Fourier transform of the respective correlation functions $G_{\rho\rho}(q,\omega)$ and $G_{\rho\theta}(q,\omega)$ in terms of the corresponding set of (conjugate) response functions $G_{\alpha\hat{\beta}}$ and the double hatted self-energy elements $\mathcal{C}_{\hat{\alpha}\hat{\beta}}$. The response functions are obtained from Eq. (46). The relevant quantities for $G_{\rho\rho}$ and $G_{\rho\theta}$ (linking with matrix elements $\mathcal{C}_{\hat{\rho}\hat{\rho}}$) are the ones with the $\hat{\rho}$ index, i.e., $G_{\rho\hat{\rho}}$ and $G_{\theta\hat{\rho}}$. Among the double hatted self-energies $\mathcal{C}_{\hat{\alpha}\hat{\beta}}$, of particular interest is the quantity $\mathcal{C}_{\hat{\rho}\hat{\rho}}$ which is the renormalized diffusion coefficient $D_R(q,\omega)$ given in Eqs. (57) and (58). The response function $G_{\rho\hat{\theta}}$ and $G_{\theta\hat{\theta}}$, both associated with a $\hat{\theta}$ field, vanish in the zero-frequency limit, since the renormalized $D_R(q,\omega)\to 0$ at the ENE transition.

According to Link B stated above, the singular contributions to the correlation functions $G_{\rho\rho}$ and $G_{\rho\theta}$ are respectively written as

$$G_{\rho\rho} \sim -G_{\rho\hat{\rho}}\mathcal{C}_{\hat{\rho}\hat{\rho}}G_{\hat{\rho}\rho}. \quad (62)$$

$$G_{\rho\theta} \sim -\mathrm{Re}[G_{\rho\hat{\rho}}\mathcal{C}_{\hat{\rho}\hat{\rho}}G_{\hat{\rho}\theta}]. \quad (63)$$

For $G_{\rho\rho}$ to have a $\delta(\omega)$ contribution at the ENE transition we must have for the corresponding Laplace transform $G_{\rho\hat{\rho}} \sim 1/z$. To show this, we consider the right-hand side of relation (62). Taking the leading-order singular behaviors of the response functions $G_{\rho\hat{\rho}} \sim 1/z$, we obtain

$$2\mathrm{Re}\left[\frac{\epsilon}{(\omega + i\epsilon)(\omega - i\epsilon)}\right] = 2\pi\delta(\omega), \quad (64)$$

since $\mathcal{C}_{\hat{\rho}\hat{\rho}} \sim z$ is vanishing at small frequencies. Hence if $G_{\rho\rho}(q,t)$ is to be nonzero for long time t, the corresponding response function $G_{\rho\hat{\rho}}(q,z)$ must have a pole at $z = 0$, i.e., $G_{\rho\hat{\rho}} \sim 1/z$. This conclusion will be referred to below as C1. Using similar arguments it is straightforward to show from Eq. (63) that to have a $\delta(\omega)$ contribution in $G_{\rho\theta}$, *both* $G_{\rho\hat{\rho}}$ and $G_{\hat{\rho}\theta}$ should have a singular part as $\sim 1/z$. This conclusion will be referred to below as C2.

We have obtained above that the response functions $G_{\rho\hat{\rho}}$ and $G_{\theta\hat{\rho}}$ are, respectively, given by Eqs. (59) and (60). The small frequency limit of $\zeta(q,\omega)$ is therefore crucial in determining the behavior of these response functions in the corresponding limit. If $G_{\rho\rho}$ or $G_{\rho\theta}$ have $1/z$ pole, then the same divergence occurs in the mode-coupling contribution to $\zeta_{\rm mc}$. This also follows from the one-loop expression (56) given in Appendix B for the mode-coupling part $\zeta_{\rm mc}$ of ζ. With a diverging $\zeta(q,\omega) \sim 1/z$ and since $D_R \sim z$ the quantity ζD_R is finite. Hence it follows from (59) that the pole of the response function $G_{\rho\hat{\rho}}$ is at finite z. On the other hand, from (60) we get that in this case the response function $G_{\theta\hat{\rho}}(q,z)$ has a $1/z$ pole. Thus with a diverging $\zeta(q,z)$, the $1/z$ behavior is only in the response function $G_{\theta\hat{\rho}}(q,z)$ but not $G_{\rho\hat{\rho}}(q,z)$. Using the conclusions C1 and C2 presented in the previous paragraph, it follows that we cannot have self-consistently either $G_{\rho\rho}$ or $G_{\theta\rho}$ develop a $\delta(\omega)$ contribution. Therefore both the correlations $G_{\rho\rho}$ and $G_{\rho\theta}$ decay in the long-time limit and D_R remains finite in the small-frequency limit. The quantity $\zeta(q,z)$ arising from the self-energy $\Sigma_{\hat{\theta}\rho}$ is also finite but nonzero. Following the same relations it is easy to show that in this case the correlation $G_{\theta\theta}$ also decays in long time. The ENE is not supported in the model.

The linear FDR's outlined in the previous section are also in agreement with this long-time (ergodic) behavior of the two correlation functions. Consider the two FDR (34) and (35) which, in the frequency transformed form, is obtained as

$$\tilde{S}^{-1}(q)G_{\rho\rho}(q,\omega) + G_{\rho\theta}(q,\omega) = -2\mathrm{Im}[G_{\rho\hat{\rho}}(q,\omega)], \quad (65)$$

$$\tilde{S}^{-1}(q)G_{\theta\rho}(q,\omega) + G_{\theta\theta}(q,\omega) = -2\mathrm{Im}[G_{\theta\hat{\rho}}(q,\omega)]. \quad (66)$$

Since neither $G_{\theta\hat{\rho}}(q,z)$ nor $G_{\rho\hat{\rho}}(q,z)$ has a $1/z$ pole it implies that $G_{\rho\rho}(q,t)$, $G_{\rho\theta}(q,t)$, and $G_{\theta\theta}(q,t)$ all become zero in the long-time limit. For the Dean-Kawasaki model, decaying of the correlation functions to zero in the long-time limit was also observed in recent works [61].

To summarize, we have demonstrated here through a nonperturbative analysis that the self-energy matrix element $\gamma_{\hat{\theta}\rho}$ plays a key role in determining the nature of the asymptotic dynamics. The self-energy arises from the nonlinear constraint (27) which takes into account an infinite number of vertices involving density fluctuations. If this quantity is zero, then the sharp ENE transition originally predicted by Kawasaki and Miyazima is supported. In the general case, however, the small-frequency limit of the renormalized diffusion constant $D_R(z)$, expressed with the Kawasaki rearrangement stated in Eq. (57), remains finite. The correlation functions $G_{\rho\rho}$ and $G_{\rho\theta}$ decay to zero in the long-time limit and the ENE is not supported in the model.

VIII. DISCUSSION

In this paper we have studied the renormalized dynamics for the nonlinear Dean-Kawasaki equation. We work out the MSR field theory corresponding to the stochastic equation for the coarse-grained density $\rho(\mathbf{x},t)$ of a dense liquid. We study the effects of nonlinearities on the nature of relaxation of density fluctuations in the system from a nonperturbative approach. The renormalization of the linear transport coefficient (D_0) is obtained here in a self-consistent form. The perturbation series for the renormalized transport coefficient D_R is organized with the Kawasaki rearrangement [62]. Having identified the proper renormalization of the transport coefficient, we analyze if an ENE transition is supported in the renormalized Dean-Kawasaki model.

The expression (58) of the cutoff function is for small wave numbers. This represents the long-wavelength behavior of the correlation function and how it decays to zero. The present work shows, using a nonperturbative approach, that the ENE transition is finally cut off since the effects of freezing finally extend to hydrodynamic length scales. For quantitative estimates of relaxation, evaluation of the mode-coupling integrals would require the correlation functions at all wave numbers. For large wave vectors, the present analysis then would have to be extended to finite-k values. Here one can adopt a phenomenological approach of introducing a short wavelength kernel [63] to regularize the model for large k. In a strict sense, however, this confronts us with the key problem of developing theory of a dense liquid (or glassy systems) over intermediate length and time scales. For dense systems, unlike a crystal, there is no basic ordered structure around which displacements of the particles can be treated as a small parameter. On the other hand,, we cannot formulate the theory in terms of a density expansion like that of a low-density gas using the inverse mean free path as the small parameter.

The first MSR field-theoretic treatment [47] of the Dean-Kawasaki equation had predicted an ENE transition in the model. At the transition the frequency transform of the density correlation function $G_{\rho\rho}(q,\omega)$ develops a singular $\delta(\omega)$ contribution. The mode-coupling contributions to the renormalized diffusion constant D_R depends on the density correlation $G_{\rho\rho}$ in such a way that former goes to zero beyond the ENE transition. Subsequent work [58] considered the renormalized theory of Ref. [47] with the extended set of variables $\{\rho,\theta\}$ and using a set of linear FDR. A careful consideration [64] of Ref. [58] shows that equations presented in this work erroneously leads to the same linear FDR (23) originally proposed by Kawasaki and Miyazima in Ref. [47]. For example, if Eq. (3.113) of Ref. [58] is substituted in the second Eq. of section (3.6.1) there, the linear FDR (23) between $G_{\rho\hat{\rho}}(k,t)$ and the density correlation $G_{\rho\rho}(k,t)$ follows and is an incorrect result. The relation (23) holds only in the case of Gaussian free energy, i.e., with linear dynamics. It does *not* hold for the Dean-Kawasaki equation which represents nonlinear dynamics. In fact, the analysis of Sec. (3.9) of Ref. [58] (in particular, the linear relationships (3.113)–(3.115) of Ref. [58]) does not hold for the nonlinear dynamics of the Dean-Kawasaki equation. Obviously, the relation (23) modifies to (35) in the nonlinear case. In a more recent work [61], it has been claimed that with the corrected form of the FDR, at the one-loop level, the two-point density correlation decays to zero in long time. Results from our nonperturbative analysis presented here is in agreement with this observation. The role of the nonlinear constraint (27) in the renormalization of the dynamics is important in this respect. The corrections to the linear model can be properly constructed with the help of the Dyson equation and available FDR's instead of restoring to an analysis specific to one-loop diagrams only. However, the Kawasaki rearrangement is needed for calculation of the renormalized diffusion constants in this case as well.

It is instructive to compare the results of the Dean-Kawasaki model formulated in terms of only the density field ρ with that of the fluctuating hydrodynamic model. In the latter the dynamics is described in terms of both the density and momentum variables $\{\rho,\mathbf{g}\}$. It has been shown in earlier works [31,46] that the ENE transition is not supported in the fluctuating hydrodynamics model. We have obtained here a similar result for case of Dean-Kawasaki dynamics. We, first, note that for each of the above two models, the feedback mechanism of MCT links the density correlation $G_{\rho\rho}$ having a singular $\delta(\omega)$ contribution to a critical behavior of a corresponding transport coefficient. In the fluctuating nonlinear hydrodynamics model $G_{\rho\rho} \sim \delta(\omega)$ is linked to the corresponding transport coefficient of *longitudinal viscosity* $L(q,\omega) \to \infty$ in the low-frequency limit. This also conforms to the divergence of viscosity. On the other hand, for the dynamics described by the Dean-Kawasaki equation, the ENE transition will imply that the renormalized *diffusion constant* $D_R(\omega)$ obtained through the Kawasaki rearrangement approaches zero in the small-frequency limit. Both models involve a corresponding nonlinear constraint arising from similar considerations. Thus, in order to have linear FDR's (21) of two-point correlations, we need to treat ψ_f as a new field. For the Dean-Kawasaki model this requires that the nonlinear part of the functional derivative $\delta F_{\mathrm{id}}/\delta\rho$ is defined as a new variable θ and hence imposing the constraint (27). Similarly, for the fluctuating hydrodynamics model with $\{\rho,\mathbf{g}\}$ fields, the functional derivative $(\delta F/\delta\mathbf{g})$ or $\mathbf{g}/\rho$ is nonlinear and hence the new field $\mathbf{v}$ is introduced through the constraint $\mathbf{g} = \rho\mathbf{v}$. For both the fluctuating hydrodynamics model and the Dean-Kawasaki model, the ergodicity restoring mechanism is linked to the corresponding nonlinear constraint. In each case, the constraint replaces *an infinite number of nonlinear terms of density fluctuations* through introduction of a corresponding new field, which is the current $\mathbf{v}$ and θ, respectively, for the two models. There is, however, a subtle difference between the two situations. In the fluctuating hydrodynamic case, the constraint renders a *nonlinear* term, involving the ρ^{-1} term in the equation of motion for **g** [46] to be a linear one. For the Dean-Kawasaki model it is the other way around, i.e., the nonlinear terms appear in the field equation for ρ as a result of introducing the θ field. In each of these two models, a respective self-energy element linked to the corresponding constraint plays the key role for the removal of the sharp transition. For the fluctuating hydrodynamics model, it is $\Sigma_{\hat{v}\rho}$. If this self-energy is zero, then there is a sharp transition and we obtain the simple MCT which has contributed much in the development of glass physics over past 30 years. In the Dean-Kawasaki model the self-energy $\Sigma_{\hat{\theta}\rho}$ arising from the constraint (27) plays the same role. If this self-energy is zero, then there is a sharp transition and analysis of Ref. [47] holds. To illustrate this, we note that if $\gamma_{\hat{\theta}\rho} = 0$, in Eq. (55) we have $\zeta = \tilde{S}^{-1}(q)$ and the response function $G_{\rho\hat{\rho}}$ has a $1/z$ pole in this case. Hence $G_{\rho\rho}(\omega) \sim \delta(\omega)$ and there will be a sharp ENE transition. Thus a nonzero self-energy $\gamma_{\hat{\theta}\rho}$ arising from the nonlinear constraint (27) is key to absence of the sharp transition.

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APPENDIX A: FLUCTUATION-DISSIPATION RELATION: ANALYSIS

We consider the FDR's (34) and (35) involving the $\hat{\rho}$ field in the following compact form:

$$\tilde{S}^{-1}(\mathbf{q})G_{\rho\alpha}(\mathbf{q},\omega) + G_{\theta\alpha}(\mathbf{q},\omega) = -2\text{Im}[G_{\hat{\rho}\alpha}(\mathbf{q},\omega)].$$

Now using the definitions (46)–(48) respectively for the response and correlation functions, we obtain the useful relation

$$\sum_{\hat{\gamma}} Z_{\rho\hat{\gamma}}\mathcal{C}_{\hat{\gamma}\hat{\delta}} = -i\left(\mathcal{D}\delta_{\hat{\rho}\hat{\delta}} + \sum_{\alpha} N_{\hat{\rho}\alpha}G^{-1}_{\alpha\hat{\delta}}\right),$$

where we have defined

$$Z_{\rho\hat{\gamma}} = \tilde{S}^{-1}(\mathbf{q})N_{\rho\hat{\gamma}} + N_{\theta\hat{\gamma}}. \tag{A1}$$

Now we obtain a set of equations by setting the index $\hat{\delta} = \hat{\theta}$ and $\hat{\rho}$ as follows:

$$Z_{\rho\hat{\rho}}\mathcal{C}_{\hat{\rho}\hat{\theta}} + Z_{\rho\hat{\theta}}\mathcal{C}_{\hat{\theta}\hat{\theta}} = -2[N'_{\rho\hat{\rho}}N''_{\theta\hat{\rho}} - N''_{\rho\hat{\rho}}N'_{\theta\hat{\rho}}], \tag{A2}$$

$$Z_{\rho\hat{\rho}}\mathcal{C}_{\hat{\rho}\hat{\rho}} + Z_{\rho\hat{\theta}}\mathcal{C}_{\hat{\theta}\hat{\rho}} = -2[(N'_{\rho\hat{\rho}}N''_{\theta\hat{\theta}} - N'_{\theta\hat{\rho}}N''_{\rho\hat{\theta}}) + i(N''_{\rho\hat{\rho}}N''_{\theta\hat{\theta}} - N''_{\rho\hat{\theta}}N''_{\theta\hat{\rho}})], \tag{A3}$$

where we have followed the standard notation of single and double primed quantities respectively denoting the real and imaginary parts of the complex entity denoted by the unprimed symbol. Since the elements of the self-energy Σ satisfies $\Sigma_{\hat{\alpha}\hat{\beta}} = \Sigma^*_{\hat{\beta}\hat{\alpha}}$, using the results of matrix III, we obtain that

TABLE V. q dependence of self-energies.

	ρ	θ	$\hat{\rho}$	$\hat{\theta}$
$\hat{\rho}$	$iq^2\gamma_{\hat{\rho}\rho}$	$iq^2\gamma_{\hat{\rho}\theta}$	$-q^2\gamma_{\hat{\rho}\hat{\rho}}$	$-q^2\gamma_{\hat{\rho}\hat{\theta}}$
$\hat{\theta}$	$i\gamma_{\hat{\theta}\rho}$	$iq^2\gamma_{\hat{\theta}\theta}$	$-q^2\gamma_{\hat{\theta}\hat{\rho}}$	$-\gamma_{\hat{\theta}\hat{\theta}}$

$\mathcal{C}_{\hat{\rho}\hat{\rho}}$ and $\mathcal{C}_{\hat{\theta}\hat{\theta}}$ are real and $\mathcal{C}_{\hat{\theta}\hat{\rho}} = \mathcal{C}^*_{\hat{\rho}\hat{\theta}}$. Using this in Eqs. (A3) and (A2) we obtain by equating real and imaginary parts the following four equations:

$$Z'_{\rho\hat{\theta}}\mathcal{C}'_{\hat{\theta}\hat{\theta}} + Z'_{\rho\hat{\rho}}\mathcal{C}'_{\hat{\theta}\hat{\rho}} + Z''_{\rho\hat{\rho}}\mathcal{C}''_{\hat{\theta}\hat{\rho}} = -2[N'_{\rho\hat{\rho}}N''_{\theta\hat{\rho}} - N''_{\rho\hat{\rho}}N'_{\theta\hat{\rho}}], \tag{A4}$$

$$Z''_{\rho\hat{\theta}}\mathcal{C}'_{\hat{\theta}\hat{\theta}} + Z''_{\rho\hat{\rho}}\mathcal{C}'_{\hat{\theta}\hat{\rho}} - Z'_{\rho\hat{\rho}}\mathcal{C}''_{\hat{\theta}\hat{\rho}} = 0, \tag{A5}$$

$$Z'_{\rho\hat{\rho}}\mathcal{C}'_{\hat{\rho}\hat{\rho}} + Z'_{\rho\hat{\theta}}\mathcal{C}'_{\hat{\theta}\hat{\rho}} - Z''_{\rho\hat{\theta}}\mathcal{C}''_{\hat{\theta}\hat{\rho}} = 2[N'_{\rho\hat{\rho}}N''_{\theta\hat{\theta}} - N'_{\theta\hat{\rho}}N''_{\rho\hat{\theta}}], \tag{A6}$$

$$Z''_{\rho\hat{\rho}}\mathcal{C}'_{\hat{\rho}\hat{\rho}} + Z''_{\rho\hat{\theta}}\mathcal{C}'_{\hat{\theta}\hat{\rho}} + Z'_{\rho\hat{\theta}}\mathcal{C}''_{\hat{\theta}\hat{\rho}} = 2[N''_{\rho\hat{\rho}}N''_{\theta\hat{\theta}} - N''_{\rho\hat{\theta}}N''_{\theta\hat{\rho}}]. \tag{A7}$$

The matrix elements $Z_{\rho\hat{\mu}}$ for $\mu \in \{\rho,\theta\}$ are given in Eq. (A1) while the elements of the $N_{\alpha\hat{\beta}}$ matrix are given in matrix IV. Using these results for the various elements of the self-energy matrix we obtain a set of relations between the element response self-energy ($\Sigma_{\psi\hat{\psi}}$) part and correlation type self-energies ($\Sigma_{\hat{\psi}\hat{\psi}}$).

1. Hydrodynamic limit

We begin by considering the self-energy matrix elements. There are primarily two types, as pointed out above, response self-energy and correlation self-energy elements, respectively denoted as $\Sigma_{\psi\hat{\psi}}(\mathbf{q},\omega)$ and $\Sigma_{\hat{\psi}\hat{\psi}}(\mathbf{q},\omega)$. From the MSR action functional for the system given by Eq. (29), it follows that the cubic vertices with a $\hat{\rho}$ leg contribute an explicit iq factor.

First, we consider the response self-energy $\Sigma_{\hat{\rho}\theta}(\mathbf{q},\omega)$ which contains only one external $\hat{\rho}(q)$ contributing a factor iq. The other leg of this self-energy involves θ. In order to maintain the scaler nature of the self-energy in this case the external vector index in **q** will be contracted (summed over) with that of the internal (integrated over) wave vector **k** with the same Cartesian index. In the small-wave-number limit ($q \to 0$) the $O(q)$ contribution for this response self-energy vanishes due to the **k** to $-$**k** symmetry in the integrand. Therefore in the hydrodynamic limit we obtain $\Sigma_{\hat{\rho}\theta}(\mathbf{q},\omega) \sim iq^2\gamma_{\hat{\rho}\theta}(\mathbf{0},0)$. Similarly, we obtain $\Sigma_{\hat{\rho}\rho} \sim iq^2\gamma_{\hat{\rho}\rho}$. The vertex with an external $\hat{\theta}$-leg does not have a derivative. We obtain $\Sigma_{\hat{\theta}\rho} \sim i\gamma_{\hat{\theta}\rho}$, and $\Sigma_{\hat{\theta}\theta} \sim iq^2\gamma_{\hat{\theta}\theta}$ as their respective lowest order contributions. The behavior of the different self-energy matrix elements, in the hydrodynamic limit, is shown in Table V.

TABLE VI. Real parts of matrix $N_{\alpha\hat{\beta}}$.

	$\hat{\rho}$	$\hat{\theta}$
ρ	$q^2\omega\tilde{\gamma}_{\hat{\theta}\theta}$	$-q^2\omega\tilde{\gamma}_{\hat{\rho}\theta}$
θ	$-\omega\tilde{\gamma}_{\hat{\theta}\rho}$	$\omega(1+q^2\tilde{\gamma}_{\hat{\rho}\rho})$

TABLE VII. Imaginary parts of matrix $N_{\alpha\hat{\beta}}$.

	$\hat{\rho}$	$\hat{\theta}$
ρ	$1-q^2\gamma'_{\hat{\theta}\theta}$	$-q^2(\bar{D}_0-\gamma'_{\hat{\rho}\theta})$
θ	$\gamma'_{\hat{\theta}\rho}$	$q^2(\tilde{D}_0-\gamma'_{\hat{\rho}\rho})$

Next, we consider the correlation elements (two hatted indices) of the self-energy matrix. $\Sigma_{\hat{\rho}\hat{\rho}}(q,\omega)$, with two vertices each having an external leg $\hat{\rho}$, should be at least of the $O(q^2)$. Writing this out explicitly we obtain $\Sigma_{\hat{\rho}\hat{\rho}} \sim -q^2\gamma_{\hat{\rho}\hat{\rho}}$. For the self-energy element $\Sigma_{\hat{\theta}\hat{\theta}}(q,\omega)$, and since the field $\hat{\theta}$ does not associate with an external derivative, this self-energy has a zeroth-order (in q) contribution. Thus the self-energy with two $\hat{\theta}$ legs is of $O(1)$, i.e., $\Sigma_{\hat{\theta}\hat{\theta}}(q,\omega) \sim -\gamma_{\hat{\theta}\hat{\theta}}$. On the other hand, the self-energy $\Sigma_{\hat{\theta}\hat{\rho}}$ has an explicit external wave vector q and hence due to the $\mathbf{k}$ to $-\mathbf{k}$ symmetry in the integral with respect to the internal wave-vector, this self-energy is of $O(q^2)$. Hence the self energy $\Sigma_{\hat{\theta}\hat{\rho}}(q,\omega) \sim -q^2\gamma_{\hat{\theta}\hat{\rho}}$. We list the real and imaginary parts of the elements of the $N_{\alpha\hat{\beta}}$ matrix in the hydrodynamic limit respectively in Tables VI and VII. Now putting the leading order dependence on the wave numbers for the various response and correlation self energies, we obtain the constants $Z_{\rho\hat{\gamma}}$ appearing in Eqs. (A4)–(A7) in terms of the corresponding γ's as:

$$Z'_{\rho\hat{\rho}} = \omega[q^2\tilde{\gamma}_{\hat{\theta}\theta}\tilde{S}^{-1}(\mathbf{q}) - \tilde{\gamma}_{\hat{\theta}\rho}], \quad \text{(A8)}$$

$$Z''_{\rho\hat{\rho}} = [\{1-q^2\gamma'_{\hat{\theta}\theta}\}\tilde{S}^{-1}(\mathbf{q}) + \gamma'_{\hat{\theta}\rho}], \quad \text{(A9)}$$

$$Z'_{\rho\hat{\theta}} = \omega[-q^2\tilde{\gamma}_{\hat{\rho}\theta}\tilde{S}^{-1}(\mathbf{q}) + \{1+q^2\tilde{\gamma}_{\hat{\rho}\rho}\}], \quad \text{(A10)}$$

$$Z''_{\rho\hat{\theta}} = q^2[\gamma'_{\hat{\rho}\theta}\tilde{S}^{-1}(\mathbf{q}) - \gamma'_{\hat{\rho}\rho}]. \quad \text{(A11)}$$

In the hydrodynamic limit using the above four equations, we obtain the given relations. From Eq. (A4),

$$\gamma_{\hat{\theta}\hat{\theta}} = -2\tilde{\gamma}_{\hat{\theta}\rho}. \quad \text{(A12)}$$

From Eq. (A5),

$$\gamma_{\hat{\theta}\hat{\theta}}(\gamma'_{\hat{\rho}\theta}\tilde{S}^{-1}(\mathbf{q}) - \gamma'_{\hat{\rho}\rho}) + \gamma'_{\hat{\theta}\hat{\rho}}(\tilde{S}^{-1}(\mathbf{q}) + \gamma'_{\hat{\theta}\rho}) = 0. \quad \text{(A13)}$$

From Eq. (A6),

$$-\tilde{\gamma}_{\hat{\theta}\rho}(\gamma_{\hat{\rho}\hat{\rho}} + 2\gamma'_{\hat{\rho}\theta}) + \gamma'_{\hat{\theta}\hat{\rho}} = 0. \quad \text{(A14)}$$

From Eq. (A7),

$$[\gamma_{\hat{\rho}\hat{\rho}}\tilde{S}^{-1}(\mathbf{q}) + 2\gamma'_{\hat{\rho}\rho}] + \gamma'_{\hat{\theta}\rho}(\gamma_{\hat{\rho}\hat{\rho}} + 2\gamma'_{\hat{\rho}\theta}) = 0. \quad \text{(A15)}$$

Using Eqs. (A6) and (A7) we obtain

$$2\gamma'_{\hat{\rho}\theta} = -\gamma_{\hat{\rho}\hat{\rho}} + \Delta_R, \quad \text{(A16)}$$

$$2\tilde{S}\gamma'_{\hat{\rho}\rho} = -\gamma_{\hat{\rho}\hat{\rho}} - \Delta_R\tilde{S}\gamma'_{\hat{\theta}\rho}. \quad \text{(A17)}$$

We define $\Delta_R = \gamma'_{\hat{\theta}\hat{\rho}}/\tilde{\gamma}_{\hat{\theta}\rho}$ above. Using Eqs. (A12), (A16), and (A17) we can obtain Eq. (A13). Equation (A13) is an independent equation,

$$\begin{aligned}\mathcal{D}(\mathbf{q},\omega) &= (\omega + iq^2\tilde{D}_R)(i - \Sigma_{\hat{\theta}\theta}) + i\bar{D}_R q^2\Sigma_{\hat{\theta}\rho}\\ &= i(\omega + iq^2\tilde{D}_R) - \bar{D}_R q^2\gamma_{\hat{\theta}\rho}\\ &= i[\omega + iq^2(\tilde{D}_R + \bar{D}_R\gamma_{\hat{\theta}\rho})],\end{aligned}$$

where the two diffusion constants are respectively obtained as stated in Eqs. (44) and (45). This further simplifies the denominator,

$$\begin{aligned}\mathcal{D}(\mathbf{q},\omega) &= i[\omega + iq^2\tilde{D}_R + iq^2\bar{D}_R\gamma_{\hat{\theta}\rho}]\\ &= i[\omega + iq^2 D_R(q,\omega)\zeta(q,\omega)],\end{aligned} \quad \text{(A18)}$$

where D_R is the renormalized diffusion coefficient to be obtained from the correlation self-energy $\gamma_{\hat{\rho}\hat{\rho}}$ through the relation

$$D_R(q,\omega) = D_0 - (2\rho_0)^{-1}\gamma_{\hat{\rho}\hat{\rho}}(q,\omega). \quad \text{(A19)}$$

The quantity $\zeta(q,\omega)$ is obtained as

$$\zeta(q,\omega) = \tilde{S}^{-1}(q) + \rho_0\gamma_{\hat{\theta}\rho}(q,\omega). \quad \text{(A20)}$$

APPENDIX B: SELF-ENERGY: ONE-LOOP RESULTS

In this Appendix we list the one-loop contributions to the relevant self-energy matrix elements. The non-Gaussian part of the MSR action functional (29) in the text gives rise to the following three-point vertices:

$$\tilde{V}_{\hat{\rho}\rho\rho}(123) = -iD_0\nabla_1\cdot[\delta(12)\nabla_1\tilde{S}^{-1}(13)]. \quad \text{(B1)}$$

The symmetrized forms of these vertices are obtained as

$$V_{\hat{\rho}\rho\rho}(123) = -i\frac{D_0}{2}\nabla_1[\delta(12)\nabla_1\tilde{S}^{-1}(13) + \delta(13)\nabla_1\tilde{S}^{-1}(12)]. \quad \text{(B2)}$$

The corresponding one-loop contribution to the self-energy is obtained as

$$\Sigma(12) = 2V(1\bar{2}\bar{3})G(\bar{2}\bar{4})G(\bar{3}\bar{5})V(\bar{4}\bar{5}2), \quad \text{(B3)}$$

where the barred index represents the coordinate which is integrated and the field index which is summed over. $V(123)$ is the cubic vertex in the MSR action functional symmetrized with respect to exchange of the filed indices and coordinates. Using this formulation we evaluate all the diagrams to obtain the following results.

1. The self-energy $\Sigma_{\hat{\rho}\hat{\rho}}$

The one-loop expression for self-energy $\Sigma_{\hat{\rho}\hat{\rho}}$ is obtained from the sum of the diagrams shown in Fig. 1. Each of the Feynman-diagrams appearing here (as well as in the Fig. 2) are marked with an attached number label. In the discussion below we refer to this number label directly in text or as

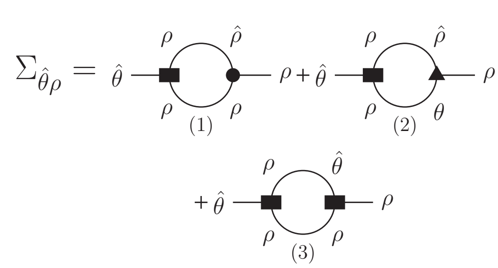

FIG. 2. One-loop diagrams for the self-energy matrix element $\Sigma_{\hat{\theta}\rho}$. The notations for the vertices are the same as described in the caption to Fig. 1.

a superscript in the LHS (see Eqs. B4 to B14 below) to indicate the contribution from the corresponding diagram. In writing the equations below we absorb a factor of D_0^2 in the definition of the corresponding diagrammatic contribution. To avoid cluttering we define two quantities $u_{\mathbf{k}} = \hat{\mathbf{q}}.\mathbf{k}$ and $J_{\mathbf{k}} = u_{\mathbf{k}}\tilde{S}^{-1}(\mathbf{k})$. The sum of the first five diagrams involving only correlation functions is obtained respectively as

$$\gamma^{(1)}_{\hat{\rho}\hat{\rho}}(q,t) = \int \frac{d\mathbf{k}}{(2\pi)^3} J_{\mathbf{k}}[J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}]G_{\rho\rho}(k,t)G_{\rho\rho}(q-k,t) \tag{B4}$$

$$\gamma^{(2)}_{\hat{\rho}\hat{\rho}}(q,t) = \int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}[J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}]G_{\rho\theta}(k,t)G_{\rho\rho}(q-k,t) \tag{B5}$$

$$\gamma^{(3)}_{\hat{\rho}\hat{\rho}}(q,t) = \int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}[J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}]G_{\theta\rho}(k,t)G_{\rho\rho}(q-k,t) \tag{B6}$$

$$\gamma^{(4)}_{\hat{\rho}\hat{\rho}}(q,t) = \int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}u_{\mathbf{q}-\mathbf{k}}G_{\theta\rho}(k,t)G_{\rho\theta}(q-k,t) \tag{B7}$$

$$\gamma^{(5)}_{\hat{\rho}\hat{\rho}}(q,t) = \int \frac{d\mathbf{k}}{(2\pi)^3} u^2_{\mathbf{k}}G_{\theta\theta}(k,t)G_{\rho\rho}(q-k,t). \tag{B8}$$

Next we consider the one-loop self-energy diagrams for $\Sigma_{\hat{\rho}\hat{\rho}}$ involving one response function. The contribution from 6 and 7 respectively involving response functions $G_{\rho\hat{\rho}}$ and $G_{\hat{\rho}\rho}$ are simplified using the FDR (34) to obtain

$$\gamma^{(6)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3}\{J_{\mathbf{k}}G_{\rho\rho}(k,t) + u_{\mathbf{k}}G_{\rho\theta}(k,t)\} \times [J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}]G_{\rho\rho}(q-k,t), \tag{B9}$$

$$\gamma^{(7)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3}\{J_{\mathbf{k}}G_{\rho\rho}(k,t) + u_{\mathbf{k}}G_{\theta\rho}(k,t)\} \times [J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}]G_{\rho\rho}(q-k,t). \tag{B10}$$

The contribution from 8 and 9 respectively involving response functions $G_{\theta\hat{\rho}}$ and $G_{\hat{\rho}\theta}$ are simplified using the FDR (35) to obtain

$$\gamma^{(8)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}[J_{\mathbf{k}}G_{\theta\rho}(k,t) + u_{\mathbf{k}}G_{\theta\theta}(k,t)] \times G_{\rho\rho}(q-k,t), \tag{B11}$$

$$\gamma^{(9)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}[J_{\mathbf{k}}G_{\rho\theta}(k,t) + u_{\mathbf{k}}G_{\theta\theta}(k,t)] \times G_{\rho\rho}(q-k,t). \tag{B12}$$

Finally contributions from 10 and 11 respectively involving response functions $G_{\rho\hat{\rho}}$ and $G_{\hat{\rho}\rho}$ are simplified using the FDR (34) to obtain

$$\gamma^{(10)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}G_{\theta\rho}(k,t)[J_{\mathbf{q}-\mathbf{k}}G_{\rho\rho}(q-k,t) + u_{\mathbf{q}-\mathbf{k}}G_{\rho\theta}(q-k,t)], \tag{B13}$$

$$\gamma^{(11)}_{\hat{\rho}\hat{\rho}}(q,t) = -\int \frac{d\mathbf{k}}{(2\pi)^3} u_{\mathbf{k}}G_{\rho\theta}(k,t)[J_{\mathbf{q}-\mathbf{k}}G_{\rho\rho}(q-k,t) + u_{\mathbf{q}-\mathbf{k}}G_{\theta\rho}(q-k,t)]. \tag{B14}$$

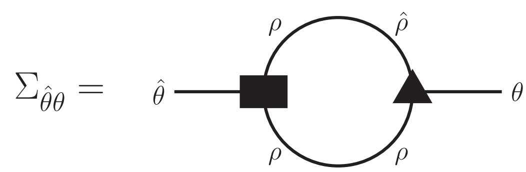

FIG. 3. One-loop diagrams for the self-energy matrix element $\Sigma_{\hat{\theta}\theta}$. The notations for the vertices are the same as described in the caption to Fig. 1.

Hence adding up the contributions from all 11 diagrams we obtain, including the D_0^2 factor, the result for the self-energy as

$$\begin{aligned}\gamma_{\hat{\rho}\hat{\rho}}(q,t) &= \sum_{i=1}^{11}\gamma^{(i)}_{\hat{\rho}\hat{\rho}}(q,t) = -D_0^2\int\frac{d\mathbf{k}}{(2\pi)^3}\bigg[G_{\rho\rho}(q-k,t) \\ &\quad\times\left[\frac{1}{2}\{J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}\}^2 G_{\rho\rho}(k,t) + u_{\mathbf{k}}\{J_{\mathbf{k}} + J_{\mathbf{q}-\mathbf{k}}\}\right. \\ &\quad\left.\times\{G_{\rho\theta}(k,t) + G_{\theta\rho}(k,t)\}\right] + u^2_{\mathbf{k}}G_{\theta\theta}(k,t)G_{\rho\rho} \\ &\quad\times(q-k,t) + u_{\mathbf{k}}u_{\mathbf{q}-\mathbf{k}}G_{\theta\rho}(k,t)G_{\rho\theta}(q-k,t)\bigg] \\ &= -D_0^2\Delta_{\mathrm{mc}}.\end{aligned} \tag{B15}$$

2. Self-energy $\Sigma_{\hat{\theta}\rho}$ and $\Sigma_{\hat{\theta}\theta}$

We display one-loop diagrams for the self-energies $\Sigma_{\hat{\theta}\rho}$, and $\Sigma_{\hat{\theta}\theta}$ respectively in Figs. 2 and 3. The one-loop expression for self-energy $\Sigma_{\hat{\theta}\rho}(q,t)$ is obtained as

$$\Sigma_{\hat{\theta}\rho}(q,t) = -\frac{D_0}{\rho_0^2}\int\frac{d\mathbf{k}}{(2\pi)^3}[(\mathbf{q}-\mathbf{k})^2G_{\rho\rho}(k,t)\tilde{S}^{-1}(k) + \mathbf{k}.(\mathbf{q}-\mathbf{k})G_{\rho\theta}(k,t)]G_{\rho\hat{\rho}}(q-k,t). \tag{B16}$$

Using the FDR for $G_{\rho\hat{\rho}}(k,t)$ we obtain

$$\begin{aligned}&= -i\Theta(t)\frac{D_0}{\rho_0^2}\int\frac{d\mathbf{k}}{(2\pi)^3} \\ &\quad\times[(\mathbf{q}-\mathbf{k})^2G_{\rho\rho}(k,t)\tilde{S}^{-1}(k) + \mathbf{k}.(\mathbf{q}-\mathbf{k})G_{\rho\theta}(k,t)] \\ &\quad\times[G_{\rho\rho}(q-k,t)\tilde{S}^{-1}(q-k) + G_{\rho\theta}(q-k,t)].\end{aligned}$$

The one-loop expression for $\Sigma_{\hat{\theta}\theta}(q,t)$ is

$$\begin{aligned}\Sigma_{\hat{\theta}\theta}(q,t) &= \frac{D_0}{\rho_0^2}\int\frac{d\mathbf{k}}{(2\pi)^3}\mathbf{q}.(\mathbf{q}-\mathbf{k})G_{\rho\rho}(k,t)G_{\rho\hat{\rho}}(q-k,t) \\ &= i\Theta(t)\frac{D_0}{\rho_0^2}\int\frac{d\mathbf{k}}{(2\pi)^3}\mathbf{q}.(\mathbf{q}-\mathbf{k})G_{\rho\rho}(k,t) \\ &\quad\times[G_{\rho\rho}(q-k,t)\tilde{S}^{-1}(q-k) + G_{\rho\theta}(q-k,t)],\end{aligned}$$

which, using the isotropic form of the correlation function, is of $O(q^2)$.

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