Equilibrium dynamics of the Dean-Kawasaki equation: MCT and beyond

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We extend a previously proposed field-theoretic self-consistent perturbation approach for the equilibrium dynamics of the Dean-Kawasaki equation presented in [J. Stat. Mech. 2008 P02004]. By taking terms missing in the latter analysis into account we arrive at a set of three new equations for correlation functions of the system. These correlations involve the density and its logarithm as local observables. Our new one-loop equations, which must carefully deal with the noninteracting Brownian gas theory, are more general than the historic Mode-Coupling one in that a further and well-defined approximation leads back to the original mode-coupling equation for the density correlations alone. However, without performing any further approximation step, our set of three equations does not feature any ergodic-non ergodic transition, as opposed to the historical mode-coupling approach.

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I. INTRODUCTION

Supercooled liquids approaching the glass transition exhibit fascinating dynamic phenomena such as tremendous slowing down and kinetic heterogeneity [1, 2]. These dynamic properties are intimately connected to the nature of the transition from liquid to glass. First-principle theoretical understanding of the dynamics of supercooled liquids and the nature of their glass formation still remains one of the greatest challenges in condensed matter science.

Applications of the mode coupling theory (MCT) to supercooled liquids [3] started in 1984, and there are still certain aspects of it that remain controversial. The best known result is the derivation of the standard MCT (SMCT) equation for the dynamical structure factor. This equation predicts the existence of a sharp kinetic transition between fluid-like ergodic and glass-like non-ergodic states. However, it is a widespread belief that this sharp transition to full dynamical arrest is spurious, and that a sharp cross-over must instead take place [4, 32]. The latter could result from thermally activated processes left out from the SMCT equation. The absence of a sharp transition described by SMCT is now well established numerically [5, 6], but the theoretical extensions of SMCT devised in order to take the so-called “activated events” [7, 33] into account is even more controversial than SMCT itself [8, 9]. These extensions attempt to include thermally activated processes which are expected to round off the sharp ergodic-nonergodic (ENE) transitions, by including the current variables in the description. But if the current variables relax in time much faster than the density variables as in deeply supercooled liquids, the former variables can adiabatically
be eliminated to result in a closed stochastic equation for the density modes. This is called the dynamical density functional theory where thermally activated processes are also taken into account [10, 11]. This is evident in appearance of the second functional derivative with respect to the density variable in the stochastic equation for the probability density functional (see Eq. (5) below). This guarantees the approach to equilibrium of the density probability distribution functional as the time goes to infinity. In any case, the search for a proper theoretical description that incorporates density fluctuations alone is needed, at least in order to treat Brownian dynamics, in which no energy or momenta currents exist.

With these issues in the background, in a recent work [12] (referred to as [KK08] throughout the present work), two of the present authors investigated the dynamical density functional equation for colloidal suspension, that we denote here by the Dean-Kawasaki (DK) equation [10, 13]. In one loop order [KK08] claimed derivation of SMCT which predicts a sharp ENE transition. [KK08] anticipated at that time that smearing effects for this transition should come from higher loop, which, however, is not the case. As we first show in this paper, there is a missing term in [KK08], and recovering it gives rise to a new set of equations that bear on several correlation functions instead of only one. We will show that these equations with one-loop self energies do not support the existence of a sharp ENE transition. We then demonstrate a striking property of these equations: that SMCT can be recovered from them by a further, well defined (but again not justified) approximation.

II. THE DEAN-KAWASAKI EQUATION

The DK equation is the following stochastic time evolution equation for the density field \(\rho(r,t)\)

\[
\frac{\partial \rho(r,t)}{\partial t} = \nabla \cdot \left( \rho(r,t) \nabla \frac{\delta F[\rho]}{\delta \rho(r,t)} \right) + \nabla \left( \sqrt{2T \rho(r,t) \eta(r,t)} \right),
\]

where \(T\) is the temperature of the system (we set the Boltzmann constant \(k_B\) unity throughout). In Eq. (1), the Gaussian thermal noise \(\eta_{\alpha}(r,t)\) has zero mean and unit variance

\[
\langle \eta_{\alpha}(r,t) \eta_{\beta}(r',t') \rangle = \delta_{\alpha \beta} \delta(r-r') \delta(t-t').
\]

The free energy density functional \(F[\rho]\) reads:

\[
F[\rho] = F_{id}[\rho] + F_{int}[\rho],
\]

\[
F_{id}[\rho] = T \int dr \rho(r) \left( \ln \left( \frac{\rho(r)}{\rho_0} \right) - 1 \right),
\]

\[
F_{int}[\rho] = \frac{1}{2} \int dr \int dr' U(|r-r'|) \delta \rho(r) \delta \rho(r'),
\]

where \(\delta \rho(r,t) \equiv \rho(r,t) - \rho_0\) is the density fluctuation around the equilibrium density \(\rho_0\). In Eq. (3) \(F_{id}[\rho]\) is the entropic contribution to the free-energy, and \(F_{int}[\rho]\) the interaction one with \(U(|r-r'|)\) representing the particle interaction. This equation, obtained by Dean [13] is the exact evolution equation for the microscopic density of a collection of interacting Brownian particles under overdamped dynamics. It thus contains all the complexity of the systems that undergo a glass transition in numerical simulations, or colloidal experiments. Surprisingly, Eqs. (1–3) were proposed by one of the present authors [10] as a mesoscopic kinetic equation for the coarse-grained density with \(U(r) = -Tc(r)\) (\(c(r)\) being the direct correlation function of the liquid). In particular, Eq. (1) was obtained via adiabatic elimination of the much faster-decaying momentum field in the fluctuating hydrodynamic equations of dense liquids [7]. We wish to emphasize that in this work, we do not use this substitution at any place, and the appearance of the renormalized static correlation function \(c(r)\) occurs naturally, due to the compatibility between equilibrium and dynamic correlations in the formalism we use, and the correct treatment of non-perturbative sum rules enforced by this compatibility.

A prominent feature of the DK equation in Eq. (1–3) is that the diffusion part has an extra factor of density, and hence the corresponding thermal noise has to be of multiplicative form in order for the density modes to relax towards their expected Gibbs distribution. The multiplicative nature of the noise greatly complicates the theoretical treatment of the dynamics of the system. It is also crucial to give a diffusion equation for the density fluctuations in the absence of interaction due to the non-polynomial density dependence of \(F_{id}[\rho]\);

\[
\frac{\delta F_{id}}{\delta \rho(r)} = T \ln \left( \frac{\rho(r)}{\rho_0} \right) \text{hence} \nabla \cdot \left( \rho \nabla \frac{\delta F_{id}}{\delta \rho} \right) = T \nabla^2 \rho.
\]

This feature is physically expected (and analytically proved in [15]) for the non-interacting Brownian particles. The second element is essential for the system to evolve toward the equilibrium stationary state due to the presence of the extra factor of density in the diffusion part. Specifically, the Fokker-Planck (FP) equation corresponding to Eqs. (1) and (2) can be written as

\[
\frac{\partial P(\{\rho\},t)}{\partial t} = -\int dr \frac{\delta}{\delta \rho(r)} \nabla \cdot \rho \left[ T \frac{\delta}{\delta \rho(r)} + \frac{\delta F[\rho]}{\delta \rho(r)} \right] P(\{\rho\},t),
\]

where \(P(\{\rho\},t)\) is the probability distribution of the density configuration \(\{\rho(r)\}\) at time \(t\). The equilibrium distribution \(P_{eq}[\rho] \propto \exp(-F[\rho]/T)\) is a stationary solution of the FP equation Eq. (5). As stated in the Introduction, in view of the presence of the second functional derivative with respect to the density variable in Eq. (5), which comes from the multiplicative thermal noise in (1), thermally activated processes are included in the equation. Of course this is a weak analogy to Kramer’s theory for
the escape across a one-dimensional potential energy barrier, where the second derivative of the free-energy represents the local curvature, which governs the escape rate. But at present we do not know an analytic way of handling the above Fokker-Planck-type equation to produce activated processes. We here just quote Langer’s theory [16] where the Fokker-Planck-type equation is used to calculate nucleation rate of first order transition using instanton-type non-perturbative calculation, which is therefore outside any renormalized perturbation theory (RPT) including the present one.

A systematic way of analyzing the DK equation is to perform a RPT (i.e. the loop expansion) on the dynamic action $S[\rho, \dot{\rho}]$ obtained from the original DK equation, which enables one to treat dynamics of the correlation and response functions on equal footing. Via the Martin-Siggia-Rose-Janssen-De Dominicis method [17, 18, 20] one can obtain the dynamic action $S[\rho, \dot{\rho}]$ for (1) and (2)

\[
S[\rho, \dot{\rho}] = \int_{r,t} \left\{ i\dot{\rho} \left[ \partial_t \rho - \nabla \cdot \left( \rho \nabla \frac{\delta F[\rho]}{\delta \rho} \right) \right] - T \rho (\nabla \rho)^2 \right\}, \tag{6}
\]

where $\int_{r,t} \equiv \int dr \int dt$, the auxiliary field $\dot{\rho}$ is a real field, and the last cubic term involving the quadratic $\dot{\rho}$ comes from the average over the multiplicative thermal noise $\eta$. The dynamic action of this form with the Ramakrishnan-Yousouff (RY) free energy functional, i.e., the free energy functional in Eq. (3) with $U(r) = -TC(r)$, was first written down in [21]. In deriving Eq. (6), employing the Itô calculus makes the Jacobian of the transformation constant [18, 19]. In principle, studying time-reversibility in such dynamic actions must be carried out within the Stratonovich discretization scheme, but remarkably the action in the latter scheme does not pick up any additional contributions.

However, it was explicitly shown [22] that the direct application of the loop expansion for the dynamic action in Eq. (6) turns out to be incompatible with the fluctuation-dissipation relation (FDR) between the density correlation function $G_{\rho\rho}(r, t)$ and the corresponding physical response function $R(r, t)$ (see (9) below), which is the central element of the equilibrium dynamics. Closely related to this, while in the additive Langevin equations the noise-response $iG_{\rho\dot{\rho}}(r, t)$ is actually proportional to the physical response, that was shown to be not the case for the DK equation [22] where the physical response is given by an unusual composite two-point function, written down in Eq. (9) below. ABL [14] further elucidated the origin of this inconsistency between the FDR and the RPT, and thereby provided an elegant way of resolving it by focusing on the time-reversal (TR) symmetry of the dynamic action. ABL then proposed the introduction of the conjugate pair of auxiliary fields $\{\theta, \dot{\theta}\}$ (defined below) to linearize the one of the TR transformations, which restores preservation of the FDR order by order in the loop expansion for the new form of the dynamic action incorporating $\{\theta, \dot{\theta}\}$. [KK08] developed a modified version of ABL’s auxiliary field method.

Yet another different scheme was recently proposed by two of the present authors in [23], exploiting the reversibility of the dynamics to map the problem onto an effective quantum system. This approach yields a reversible formulation, in which the case of noninteracting Brownian gas (NBG) ($U = 0$ in (1) is a Gaussian theory, but the correlation function in this approach is a four-point object, and thus new difficulties arise when attempting to perform a loop expansion. The formulation proposed in [14], in [KK08], and in the present paper are more straightforward to analyze, since they are formulated in terms of two-body objects, however, it will be shown that the case of NBG is a strongly non-Gaussian theory, giving rise to new difficulties. More precisely, the new form of the action, when $U = 0$, contains an infinite number of polynomial terms, which should be taken into account non-perturbatively. Instead, performing a loop-expansion amounts to developing in perturbation theory around a Gaussian ground state, whereas the NBG case is strongly non-Gaussian [15] due to the multiplicative nature of the thermal noise acting on the density field.

A. Time-reversal symmetries

The dynamic action in Eq. (6) is invariant under the following two types of TR transformation for $\rho$ and $\dot{\rho}$.

The U-transformation involves the free energy functional:

\[
U: \begin{cases} 
\rho(r, -t) \rightarrow \rho(r, t) \\
\dot{\rho}(r, -t) \rightarrow -\dot{\rho}(r, t) + \frac{i}{T} \frac{\delta F[\rho]}{\delta \rho(r, t)}.
\end{cases} \tag{7}
\]

Another transformation explicitly involves the time derivative:

\[
T: \begin{cases} 
\rho(r, -t) \rightarrow \rho(r, t) \\
\nabla \cdot \left( \rho(r, -t) \nabla \dot{\rho}(r, -t) \right) \rightarrow \\
\nabla \cdot \left( \rho(r, t) \nabla \dot{\rho}(r, t) \right) + \frac{i}{T} \partial_t \rho(r, t).
\end{cases} \tag{8}
\]

The U-transformation was first written down by Janssen [18], and the T-transformation appears first in [14]. Important feature of these TR transformations is that both are nonlinear: the former is nonlinear owing to the form of the entropic contribution $F_{\mu}[\rho]$, whereas the latter due to the multiplicative nature of the thermal noise acting on the density field.

\[
\frac{1}{T} \partial_t G_{\rho\rho}(r - r', t - t') = R(r - r', t - t') - R(r - r', t - t') \tag{9}
\]

\[
G_{\rho\rho}(r - r'; t - t') \equiv \langle \delta \rho(r, t) \delta \rho(r', t') \rangle, \tag{9}
\]

\[
R(r - r', t - t') \equiv - \left\langle \rho(r, t) \nabla \cdot \left( \rho(r', t') \nabla \dot{\rho}(r', t') \right) \right\rangle. \tag{9}
\]
The problem is that the decomposed Gaussian and non-Gaussian components of the original dynamic action in Eq. (6) are separately invariant under neither transformations, which makes the direct loop expansion incompatible with the FDR.

Each TR transformation offers its own perturbation scheme preserving the FDR. First of all, both transformations can be made linear by introducing extra sets of field variables. In this regard, it is much more convenient to linearize the U-transformation since introducing a single set of extra fields (denoted by \( \theta \) and \( \bar{\theta} \)) would suffice. One then can apply the standard loop-expansion method for the new dynamic action incorporating the new fields \( \theta \) and \( \bar{\theta} \) (see (12) below).

One can follow another perturbation scheme, a potential expansion method, which preserves the T-transformation. Here although the interaction-\( T \) transformation as
\[
\hat{\theta} = 0.
\]
Thus \( \theta \) field represents all the non-Gaussian density fluctuations coming from the entropic part of the free energy.

The resulting new dynamic action \( S[\rho, \hat{\rho}, \theta, \bar{\theta}] \) is then decomposed into a Gaussian part \( S_g \) and a non-Gaussian part \( S_{ng} \), defined as:
\[
S[\rho, \hat{\rho}, \theta, \bar{\theta}] = S_g[\rho, \hat{\rho}, \theta, \bar{\theta}] + S_{ng}[\rho, \hat{\rho}, \theta, \bar{\theta}],
\]
\[
S_g \equiv \int_{r,t} \left\{ i\hat{\rho} \left[ \partial_\rho - T \nabla^2 \rho - \rho_0 T \nabla^2 \theta - \rho_0 \nabla^2 U + \delta \rho \right] \right\},
\]
\[
S_{ng} \equiv \int_{r,t} \left\{ i\hat{\rho} \left[ -T \nabla \cdot \left( \delta \rho \nabla \left[ \hat{K} \delta \rho \right] \right) - T \nabla \cdot (\delta \rho \nabla \theta) \right] \right\},
\]
where \( \delta \) denotes the space convolution, i.e.,
\[
\left( \hat{K} \delta \rho \right) (r,t) \equiv \int_{r'} K(|r-r'|) \delta \rho(r',t),
\]
and \( K \) is defined by:
\[
K(|r-r'|) \equiv \frac{1}{\rho_0} \delta(r-r') + \frac{1}{T} U(|r-r'|).
\]

In Eq. (12), the field \( \bar{\theta}(r,t) \) appears from the integral representation of the delta-functional constraint \( \delta[\theta(r,t) - f(\rho(r,t))] \) from Eq. (11), which inevitably generates the nonpolynomial (logarithmic) nonlinearity \( -i\bar{\theta}f(\delta \rho) \), while the original action in Eq. (6) contains polynomial nonlinearities only.

Although Andreanov, Biroli, and Lefèvre (ABL) [14] uses the same formal setting as the present work, the results they obtain have a rather different structure. In particular, ABL uses a different definition of \( \theta \) variable. ABL’s definition \( \theta_{ABL} = \delta F/\delta \rho \) includes even the linear part of density fluctuation, whereas \( \theta \) in Eq. (11) explicitly separate the linear density fluctuation from \( \delta F_{id}/\delta \rho \). Because of this difference, distinction between ABL and the present work persists through the forms of the new dynamic action, some of the FDRs, and the dynamic equations. It is thus useful to keep in mind that their (Fourier transformed) correlation functions \( C_{\rho \rho}(k,t) \) and \( C_{\theta \theta}(k,t) \) correspond respectively to \( T(K(k)G_{\rho \rho}(k,t) + G_{\rho \theta}(k,t)) \) and \( T(K(k)G_{\theta \rho}(k,t) + G_{\theta \theta}(k,t)) \) in our case. Even if this correspondence is taken into account, the resulting dynamic equations exhibit a subtle difference in both cases, particularly for the equations for \( G_{\rho \theta} \) and \( G_{\theta \theta} \). At any rate, we tend to believe that ABL’s analysis concerning the decay at long times of correlation functions of the theory is erroneous, and in particular, the equation for the nonergodicity parameter of \( G_{\rho \theta}(k,t) \) (Eq. (97) of [14]), which is based on several hypotheses and on the neglect of the first memory integral in the long-time limit (Eq. (88) of [14]), which would not be zero. In this work we do not make such hypotheses, and our results are obtained without any assumptions other than the starting models.

B. Linearization of the U-transformation

In this work, we focus on the loop expansion method via the linearization of the U-transformation. As stated earlier, it can be achieved by letting a new auxiliary field \( \theta(r,t) \) take up only the nonlinear part of the U-transformation as
\[
\theta(r,t) = f(\delta \rho(r,t)) \equiv \frac{1}{T} \frac{\delta F_{id}(\rho)}{\delta \rho(r,t)} - \frac{\delta \rho(r,t)}{\rho_0}.
\]
\[
\ln \left( 1 + \frac{\delta \rho(r,t)}{\rho_0} \right) - \frac{\delta \rho(r,t)}{\rho_0}
\]
\[
= - \sum_{n=2}^{\infty} \frac{(-1)^n}{n} \left( \frac{\delta \rho(r,t)}{\rho_0} \right)^n.
\]
\[
\]
Now $S_0$ and $S_{np}$ become separately invariant under the linearized U-transformation

$$U_L:\begin{cases}
\rho(r,-t) = \rho(r,t) , \\
\dot{\rho}(r,-t) = -\dot{\rho}(r,t) + i(\dot{K} \ast \delta \rho)(r,t) + i\theta(r,t) , \\
\theta(r,-t) = \theta(r,t) , \\
\dot{\theta}(r,-t) = \dot{\theta}(r,t) + i\partial_t \rho(r,t) .
\end{cases}$$

Here the modulus of the associated transformation matrix $U$ is unity (det $U = -1$). That the transformation of $\rho$ field involves not only $\rho$ but also $\theta$ fields reflects non-Gaussian contributions of the entropic free energy. The response field $\dot{\theta}$ acts as the time derivative of density fluctuation for time-reversal invariance since it couples with a function of density fluctuation.

Now due to the separate invariance of the new dynamic action under the $U_L$ transformation, the RPT for the new dynamic action is guaranteed to preserve the FDR order by order.

However, it is obvious from the form of the action (12) that the NBG case is not a Gaussian theory, since setting $U = 0$ in the action does not cancel its non-Gaussian part. An order by order loop expansion is thus not guaranteed to treat correctly the NBG.

Until now, we have recalled the preliminary steps of the work presented in [KK08], for the sake of clarity, but in the following some results taken from this previous work will be recalled without proof, for the sake of compactness.

III. DYNAMIC EQUATIONS

The linear transformation under time reversal in Eq. (15) dictates the following linear FDRs between the correlation and response functions:

$$\begin{align*}
G_{\rho\rho}(k,t) & = i \Theta(t) (K(k)G_{\rho\rho}(k,t) + G_{\rho\theta}(k,t)) , \\
G_{\rho\theta}(k,t) & = i \Theta(t) \partial_t G_{\rho\rho}(k,t) , \\
G_{\theta\rho}(k,t) & = i \Theta(t) (K(k)G_{\rho\theta}(k,t) + G_{\theta\theta}(k,t)) , \\
G_{\theta\theta}(k,t) & = i \Theta(t) \partial_t G_{\theta\theta}(k,t) + i\delta(t) ,
\end{align*}$$

where $K(k) \equiv 1/\rho_0 + U(k)/T$ is the Fourier transform of $K(r)$ defined in Eq. (14). Due to the nature of $\theta$ field, $G_{\rho\theta}(k,t)$ in the first FDR is a non-Gaussian contribution (from the entropic part of the free energy) to the density correlation. Since $\dot{\theta}$ field acts as time derivative of density fluctuation, the second FDR in (16) is nothing but the standard FDR, the first line of (9) with the physical response now reduced to $R(r,t) = iG_{\rho\theta}(r,t)/T$.

The additional Dirac delta term in $G_{\theta\theta}$ in (16) does not come from the FDR but is a contribution coming directly from the bare correlator given by the action in Eq. (12), which is not renormalized by any loop diagrams as can be seen by direct inspection. This term reflects the fact that the Lagrange multiplier $\dot{\theta}$ acts also at equal times, and thus affects the equilibrium correlations in addition to dynamic ones. [KK08] missed out this last term, which significantly affects the analysis given in this previous work. In particular, the overlooked delta function generates additional terms in the equations of motion for the correlation and response functions, and thereby stymies the nonperturbative analysis given in Section 3.9 of [KK08]. The new analysis given below remedies these erroneous results. We note that such a term was correctly taken into account in the previous work of ABL [14].

The self-energies are defined as the functional inverse of the propagators via the Schwinger-Dyson (SD) equation:

$$\Gamma(t) = (G_0^{-1} \otimes G)(k,t) - (\Sigma \otimes G)(k,t) ,$$

where $G_0^{-1}$ is the inverse of the bare propagator, which is simply read off from the dynamical action, $G$ and $\Sigma$ are $4 \times 4$ matrices containing all correlators and self-energies respectively, and $\Gamma$ is a $4 \times 4$ identity matrix. The symbol $\otimes$ stands for time convolution. The FDRs holds also for the self-energies, and read:

$$\begin{align*}
\Sigma_{\rho\rho}(k,t) & = i \Theta(t) \left( -K(k)\Sigma_{\rho\rho}(k,t) + \partial_t \Sigma_{\rho\theta}(k,t) \right) , \\
\Sigma_{\rho\theta}(k,t) & = -i \Theta(t) \Sigma_{\rho\rho}(k,t) , \\
\Sigma_{\theta\rho}(k,t) & = i \Theta(t) \left( -K(k)\Sigma_{\theta\rho}(k,t) + \partial_t \Sigma_{\theta\theta}(k,t) \right) , \\
\Sigma_{\theta\theta}(k,t) & = -i \Theta(t) \Sigma_{\theta\rho}(k,t) , \\
\Sigma_{\theta\rho}(k,t) & = -\Sigma_{\rho\theta}(k,t) .
\end{align*}$$

The FDRs in Eqs. (16) and (18) are employed to derive the dynamic equations for the correlation functions given in the next section.

### A. Equations for the correlation functions

The dynamic equations for the correlation and response functions are formally given by the matrix SD equation in Eq.(17).

We first write down the dynamic equations for the correlation functions:

$$\begin{align*}
\partial_t G_{\rho\rho}(k,t) & = -\rho_0 Tk^2 \left[ K(k)G_{\rho\rho}(k,t) + G_{\rho\theta}(k,t) \right] \\
+ & \int_0^t ds \left[ \Sigma_{\rho\rho}(k,t-s) \left[ K(k)G_{\rho\rho}(k,s) + G_{\rho\theta}(k,s) \right] - \Sigma_{\rho\theta}(k,t-s) \partial_s G_{\rho\rho}(k,s) \right] ,
\end{align*}$$

$$G_{\theta\rho}(k,t) = \Sigma_{\theta\rho}(k,0)G_{\rho\rho}(k,t)$$

$$+ \int_0^t ds \left[ \Sigma_{\theta\rho}(k,t-s) \left[ K(k)G_{\rho\rho}(k,s) + G_{\rho\theta}(k,s) \right] - \Sigma_{\rho\theta}(k,t-s) \partial_s G_{\rho\rho}(k,s) \right] .$$

(19)
$\partial_t G_{\rho \theta}(k, t) = -\rho_0 T k^2 \left[ K(k) G_{\rho \rho}(k, t) + G_{\theta \theta}(k, t) \right]$

$- \Sigma_{\rho \theta}(k, t) \left[ K(k) G_{\rho \rho}(k, 0) + G_{\theta \rho}(k, 0) \right]$

$+ \int_0^t ds \left( \Sigma_{\rho \theta}(k, t-s) \left[ K(k) G_{\rho \rho}(k, s) + G_{\theta \theta}(k, s) \right] - \Sigma_{\theta \rho}(k, t-s) \partial_s G_{\rho \rho}(k, s) \right)$. \hspace{1cm} (21)

$G_{\theta \theta}(k, t) = \Sigma_{\theta \theta}(k, 0) G_{\theta \theta}(k, t)$

$- \Sigma_{\theta \theta}(k, t) \left[ K(k) G_{\rho \rho}(k, 0) + G_{\theta \rho}(k, 0) \right]$

$+ \int_0^t ds \left( \Sigma_{\theta \theta}(k, t-s) \left[ K(k) G_{\rho \rho}(k, s) + G_{\theta \theta}(k, s) \right] - \Sigma_{\theta \rho}(k, t-s) \partial_s G_{\rho \rho}(k, s) \right)$. \hspace{1cm} (22)

The second terms on the right hand sides of Eqs. (21) and (22) are the ones generated by the presence of the delta function in the last member of Eq. (16). These new terms turn out to invalidate the nonperturbative results given in Sec. 3.9 of [KK08]. As a result, one cannot obtain a single closed equation for $G_{\rho \rho}(k, t)$ alone. Instead, one should face a set of coupled equations for the correlation functions involving $\theta$ variables. The very same situation is observed in the formulation of Jacquin and van Wijland [23], and it is a difficulty that seem to naturally occur when pairs of extra fields are introduced.

### B. Static input

Since we are dealing with the dynamics of fluctuations around the equilibrium state, the static information prescribed by the free energy $F[\rho]$ in Eq. (3) should be consistent with the initial conditions for the dynamic equations of motion for the correlation functions. Injecting the causal forms of the correlators in the SD equation (17), time derivatives contained in the inverse propagator will act on the Heaviside functions and produce Dirac delta functions. Now assuming that only $G_{\theta \theta}$ contains a delta function, and that no self-energy gets such a term, we can readily equate the delta functions in Eq. (17) we obtain directly:

$\begin{pmatrix}
\mathcal{X}(k, 0) & \mathcal{Y}(k, 0) & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & \mathcal{X}(k, 0) & \hat{G}_{\rho \rho}(k, 0) + \rho_0 T k^2 \\
0 & 0 & 0 & 1
\end{pmatrix}$

$= \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}$ \hspace{1cm} (23)

where the dot stands for time derivative. In Eq. (23), we have defined:

$\begin{pmatrix}
\mathcal{X}(k, 0) = K(k) G_{\rho \rho}(k, t) + G_{\rho \theta}(k, t) \\
\mathcal{Y}(k, 0) = K(k) G_{\rho \theta}(k, t) + G_{\theta \theta}(k, t)
\end{pmatrix}$. \hspace{1cm} (24)

We thus obtain three constraints, which will be shown to be fully consistent with the rest of the dynamical equations:

$\mathcal{X}(k, 0) = 1$, \hspace{1cm} (25)

$\mathcal{Y}(k, 0) = 0$, \hspace{1cm} (25)

$\hat{G}_{\rho \rho}(k, 0) = -\rho_0 T k^2$. \hspace{1cm} (26)

Since the system is assumed to be in equilibrium, we feed the equilibrium correlation, which is the static structure factor of the liquid:

$G_{\rho \rho}(k, 0) = \rho_0 S(k)$ \hspace{1cm} (27)

Injecting this and Eq. (25) directly into Eq. (26), we obtain the important, non-perturbative relation:

$\Sigma_{\theta \theta}(k, 0) = \left( \frac{1}{\rho_0 S(k)} - K(k) \right) = - \left( c(k) + \frac{U(k)}{T} \right)$, \hspace{1cm} (28)

$G_{\rho \theta}(k, 0) = 1 - \rho_0 S(k) K(k)$, \hspace{1cm} (26)

$G_{\theta \theta}(k, 0) = \Sigma_{\theta \theta}(k, 0) K(k)$. \hspace{1cm} (28)

where we expressed in the first line the static structure factor $S(k)$ in terms of the direct correlation function $c(k)$ using

$\frac{1}{S(k)} = 1 - \rho_0 c(k)$.

### C. Analysis of Dynamic equations

In [KK08], overlooking the extra delta function contribution to $G_{\theta \theta}(k, t)$ in Eq. (16), the authors falsely obtained the relation

$\frac{G_{\rho \rho}(k, t)}{G_{\rho \rho}(k, 0)} = \frac{G_{\rho \rho}(k, t)}{\rho_0 S(k)}$. \hspace{1cm} (29)

This incorrect relation turns the first FDR in Eq. (16) into the (false) linear FDR between $G_{\rho \rho}(k, t)$ and $G_{\rho \rho}(k, t)$

$G_{\rho \rho}(k, t) = i \Theta(t) \frac{1}{\rho_0 S(k)} G_{\rho \rho}(k, t)$.
This erroneous FDR led to a closed equation for \( G_{\rho\rho}(k, t) \) alone. This is not so. One really has to face a coupled set of dynamic equations for the correlation functions \( G_{\rho\rho}(k, t), G_{\rho\theta}(k, t) \) (or \( G_{\theta\rho}(k, t) \)), and \( G_{\theta\theta}(k, t) \). We write them down again incorporating the static input;

\[
\partial_t G_{\rho\rho}(k, t) = -\rho_0 T k^2 \mathcal{X}(k, t)
\]

\[
+ \int_0^t ds \left[ \Sigma_{\rho\rho}(k, t-s) \mathcal{X}(k, s) - \Sigma_{\rho\rho}(k, t-s) \partial_s G_{\rho\rho}(k, s) \right],
\]

(29)

\[
\mathcal{X}(k, t) = \frac{1}{\rho_0 S(k)} G_{\rho\rho}(k, t)
\]

\[
+ \int_0^t ds \left[ \Sigma_{\rho\rho}(k, t-s) \mathcal{Y}(k, s) - \Sigma_{\rho\rho}(k, t-s) \partial_s G_{\rho\rho}(k, s) \right],
\]

(30)

\[
\partial_t G_{\rho\theta}(k, t) = -\rho_0 T k^2 \mathcal{Y}(k, t) - \Sigma_{\rho\theta}(k, t)
\]

\[
+ \int_0^t ds \left[ \Sigma_{\rho\theta}(k, t-s) \mathcal{Y}(k, s) - \Sigma_{\rho\theta}(k, t-s) \partial_s G_{\rho\theta}(k, s) \right],
\]

(31)

\[
\mathcal{Y}(k, t) = \frac{1}{\rho_0 S(k)} G_{\rho\theta}(k, t)
\]

\[
+ \int_0^t ds \left[ \Sigma_{\rho\theta}(k, t-s) \mathcal{Y}(k, s) - \Sigma_{\rho\theta}(k, t-s) \partial_s G_{\rho\theta}(k, s) \right].
\]

(32)

The above equations (29)-(32) form a coupled set of FDR-preserving dynamic equations for the correlation functions \( G_{\rho\rho}(k, t), G_{\rho\theta}(k, t) \) (or \( G_{\theta\rho}(k, t) \)), and \( G_{\theta\theta}(k, t) \) since the response functions appearing in the self-energies can be replaced by the corresponding correlation functions via the linear FDRs in Eq. (16). The second terms in the right hand sides of Eqs. (31) and (32) are those generated by the overlooked delta function contribution in the last member of Eq. (16). Due to these terms, the relation \( G_{\rho\theta} = i\Theta(t)G_{\rho\rho}(k, t)/\rho_0 S(k) \) is invalid and consequently one cannot obtain the single closed equation for \( G_{\rho\rho}(k, t) \). These equations with the explicit expressions for the self-energies, could be solved numerically with the given initial conditions.

Let us rearrange the dynamic equations (29)-(32) for analysis. Subtracting Eq. (30) multiplied by \( \rho_0 T k^2 \) from Eq. (29), we obtain

\[
\partial_t G_{\rho\rho}(k, t) = -\frac{T k^2}{S(k)} G_{\rho\rho}(k, t)
\]

\[
+ \int_0^t ds \left[ \Sigma_1(k, t-s) \mathcal{X}(k, s) - \Sigma_2(k, t-s) \partial_s G_{\rho\rho}(k, s) \right],
\]

where \( \Sigma_1(k, t) \) and \( \Sigma_2(k, t) \) are defined as

\[
\Sigma_1(k, t) \equiv \Sigma_{\rho\rho}(k, t) - \rho_0 T k^2 \Sigma_{\rho\theta}(k, t),
\]

(33)

\[
\Sigma_2(k, t) \equiv \Sigma_{\rho\rho}(k, t) - \rho_0 T k^2 \Sigma_{\theta\theta}(k, t).
\]

Note that the first memory integral in Eq. (33) is folded onto \( \mathcal{X}(k, t) \) instead of the time derivative of \( G_{\rho\rho}(k, t) \) as in the second one. One recognizes from the form of Eq. (29) that one can make the first integral folded into the time derivative of \( G_{\rho\rho}(k, t) \) with the new memory function \( \mathcal{M}(k, t) \);

\[
\int_0^t ds \, \Sigma_1(k, t-s) \mathcal{X}(k, s) \equiv -\int_0^t ds \, \mathcal{M}(k, t-s) \partial_s G_{\rho\rho}(k, s)
\]

(36)

where the memory kernel \( \mathcal{M}(k, t) \) obeys the following exact equation (see Appendix A):

\[
\rho_0 T k^2 \mathcal{M}(k, t) = \Sigma_1(k, t) - \int_0^t ds \, \Sigma_2(k, t-s) \mathcal{M}(k, s)
\]

\[
+ \int_0^t ds \, \Sigma_{\rho\theta}(k, t-s) \mathcal{M}(k, s).
\]

(37)

This rearrangement is superficially analogous to the one employed in the irreducible memory function formulation [28, 29] in the projection operator approach. In our case, it is not an operation that simplifies the diagrammatic structure of the memory kernel, but it is rather a projection onto the subspace of density, factoring out the \( \theta \) correlations, which is a necessary step to make contact with the SMCT. We note that the folding onto the time derivative of \( G_{\rho\rho}(k, t) \) causes a fundamental change in the structure of the loop expansion by inevitably bringing higher order contributions to the new memory function \( \mathcal{M}(k, t) \). That is, in addition to the one-loop term, \( \Sigma_1(k, t)/\rho_0 T k^2 \), higher order terms are generated by iterating Eq. (37) for \( \mathcal{M}(k, t) \). As shown below, this lowest order term \( \Sigma_1(k, t)/\rho_0 T k^2 \) gives SMCT, but the full one loop calculation contains these higher order contributions beyond SMCT (which gives rise to ENE transition) such that this ‘extra’ part of one loop gives rise to a smearing out of SMCT.

Substituting Eq. (36) into Eq. (33), one can rewrite Eq. (33) as the following exact equation

\[
\partial_t G_{\rho\rho}(k, t) = -\frac{T k^2}{S(k)} G_{\rho\rho}(k, t)
\]

\[
- \int_0^t ds \left( \mathcal{M} + \Sigma_2 \right)(k, t-s) \partial_s G_{\rho\rho}(k, s).
\]

(38)

We must stress that Eq. (38) is not a closed equation for \( G_{\rho\rho}(k, t) \) alone since the memory kernels \( \mathcal{M}(k, t) \) and \( \Sigma_2(k, t) \) will involve both \( G_{\rho\rho}(k, t) \) and \( G_{\theta\theta}(k, t) \) as well.

In the same manner, one can also rearrange Eq. (30) as

\[
\mathcal{X}(k, t) = \frac{1}{\rho_0 S(k)} G_{\rho\rho}(k, t)
\]

\[
- \int_0^t ds \left( N + \Sigma_{\theta\theta} \right)(k, t-s) \partial_s G_{\rho\rho}(k, s).
\]

(39)
Here the new memory kernel \( N(k,t) \), being defined as

\[
\int_0^t ds \, \Sigma_\delta(k,t-s) \chi(k,s) \equiv -\int_0^t ds \, N(k,t-s) \partial_s G_{\rho\rho}(k,s),
\]

obeys (see Appendix A)

\[
\rho_0 T k^2 \mathcal{N}(k,t) = \Sigma_\delta(k,t) - \int_0^t ds \left( \Sigma_\delta(k,t-s) \Sigma_\delta(k,s) - \Sigma_\delta(k,t-s) \Sigma_\delta(k,s) \right) \mathcal{N}(k,s).
\]

Eqs. (31) and (32) can be rewritten as:

\[
\partial_t G_{\rho\rho}(k,t) = -\frac{T k^2}{S(k)} G_{\rho\rho}(k,t) - \Sigma_2(k,t)
\]

and

\[
\gamma(k,t) = \frac{1}{\rho_0 S(k)} G_{\rho\rho}(k,t) - \Sigma_\delta(k,t)
\]

\[
+ \int_0^t ds \left( \Sigma_1(k,t-s) \gamma(k,s) - \Sigma_2(k,t-s) \partial_s G_{\rho\rho}(k,s) \right).
\]

Finally the one-loop expression of \( \Sigma_1(k,t) \) reads:

\[
\Sigma_1(k,t) = \frac{T^2 k^2}{\rho_0} \int_q k \cdot q G_{\theta\rho}(q,t) G_{\rho\rho}(|k - q|,t)
\]

\[
+ T^2 \int_q k \cdot q \int q k \cdot (k - q) G_{\theta\rho}(q,t) G_{\rho\rho}(|k - q|,t)
\]

\[
+ T^2 \int_q \left( k \cdot q \right)^2 G_{\theta\rho}(q,t) G_{\rho\rho}(|k - q|,t)
\]

\[
+ \frac{1}{2} \int_q \left[ k \cdot q U(q) + k \cdot (k - q) U(|k - q|) \right] \left( G_{\rho\rho} G_{\rho\rho} \right)
\]

\[
+ \frac{T k^2}{2\rho_0} \int_q \left[ k \cdot q U(q) + k \cdot (k - q) U(|k - q|) \right] \left( G_{\rho\rho} G_{\rho\rho} \right)
\]

\[
+ 2T \int_q k \cdot q \left[ k \cdot q U(q) + k \cdot (k - q) U(|k - q|) \right] G_{\theta\rho}(q,t) G_{\rho\rho}(|k - q|,t).
\]

Thus, the FDR-preserving one-loop theory is represented by a coupled set of dynamic equations: Eqs. (38), Eq. (37), Eq. (39) with Eq. (41), and Eqs. (42)-(43), with the one-loop self-energies given by Eqs. (44), (45), and (46). In the following we show how the SMCT equation can be retrieved within the one-loop theory. We emphasize, however, that the one-loop theory is not tantamount to the SMCT: we will show that the full one-loop theory does not support the dynamic transition into nonergodic phase predicted by the SMCT, i.e., the equilibrium dynamics remains always ergodic within the full one-loop theory.

A. Contact with the SMCT

We have now completed the description of the dynamical theory, and shown that a simple one-loop approximation is not sufficient to obtain the SMCT equation. In order to do so, one must force the dynamical equations to be expressed in function only of the density-density correlator \( G_{\rho\rho}(k,t) \).

We thus introduce a book-keeping parameter \( \lambda \) in front of each self-energy i.e. we make the replacement:

\[
S_{ng} \rightarrow \sqrt{\lambda} S_{ng} ,
\]

and use \( \lambda \) as an organizing device for the diagrammatic expansion. We will expand the equations to lowest order in \( \lambda \), and evaluate the result at \( \lambda = 1 \) at the end. This is of course not an expansion in powers of a small parameter, but neither is the loop expansion! This procedure of introducing a fictitious expansion parameter in the calculation, then equating it to one originates from quantum field theory, where an expansion in powers of \( \hbar \) produces a loop expansion. In statistical field theory, such a small parameter is usually absent (with the exception of \( \mathcal{O}(N) \) models for example, where expansions in powers of \( 1/N \)))
can be performed), and the loop expansion is produced by placing a $1/\lambda$ factor in front of the action, then organizing the diagrammatic expansion in powers of $\lambda$, before setting $\lambda$ to 1 at the end of the calculation.

From their expressions in Eqs. (34) and (35) we are thus bound to find:

$$
\Sigma_1 = \lambda \Sigma_1^{(1)} + \lambda^2 \Sigma_1^{(2)} + O(\lambda^3),
$$

$$
\Sigma_2 = \lambda \Sigma_2^{(1)} + \lambda^2 \Sigma_2^{(2)} + O(\lambda^3),
$$

and thus we find from Eq. (37)

$$
\mathcal{M}(k,t) = \frac{1}{\rho_0 T k^2} \Sigma_1^{(1)}(k,t) \lambda + O(\lambda^2). \quad (48)
$$

Thus to order one in $\lambda$, we obtain from (38) the following equation:

$$
\partial_t G_{\rho\rho}(k,t) = -\frac{T k^2}{S(k)} G_{\rho\rho}(k,t) - \lambda \int_0^t ds \left( \Sigma_1^{(1)} \right)(k,t-s) \partial_s G_{\rho\rho}(k,s) + O(\lambda^2). \quad (49)
$$

This equation is still not a closed equation for $G_{\rho\rho}(k,t)$ since the self-energies $\Sigma_1^{(1)}$ and $\Sigma_2^{(1)}$ depend on $G_{\rho\rho}(k,t)$ and $G_{\theta\theta}(k,t)$ as well. But with the same reasonings, Eqs. (39–43) show that:

$$
G_{\rho\theta}(k,t) = \left( \frac{1}{\rho_0 S(k)} - K(k) \right) G_{\rho\rho}(k,t) + O(\lambda),
$$

$$
G_{\theta\theta}(k,t) = \left( \frac{1}{\rho_0 S(k)} - K(k) \right)^2 G_{\rho\rho}(k,t) + O(\lambda).
$$

Thus at order one in $\lambda$, the self-energies involve only the density correlation function $G_{\rho\rho}(k,t)$, and we recover the false FDR obtained in [KK08]. Within this level of approximation, all results in [KK08] thus hold.

Eq. (45) shows that:

$$
\Sigma_2^{(1)}(k,t) = \frac{T}{2\rho_0 k^2} \int_q \left[ k \cdot q c(q) + k \cdot (k-q) c(|k-q|) \right] G_{\rho\rho} G_{\rho\rho}. \quad (50)
$$

Note that in Eq. (50) the interaction potential $U$ cancels out, and the direct correlation function of the liquid, $c(q)$, naturally emerges through the static structure factor $1/S(q) = 1 - \rho_0 c(q)$.

Likewise, Eq. (46) reduces to

$$
\frac{1}{\rho_0 T k^2} \Sigma_1^{(1)}(k,t) = \frac{T}{2\rho_0 k^2} \int_q \left[ k \cdot q c(q) + k \cdot (k-q) c(|k-q|) \right] G_{\rho\rho} G_{\rho\rho},
$$

$$
- \frac{T}{2\rho_0} \int_q \left[ k \cdot q c(q) + k \cdot (k-q) c(|k-q|) \right] G_{\rho\rho} G_{\rho\rho}. \quad (51)
$$

With Eqs. (50) and (51), Eq. (49) yields the SMCT equation when evaluated to first order in $\lambda$ then at $\lambda = 1$:

$$
\partial_t G_{\rho\rho}(k,t) = -\frac{T k^2}{S(k)} G_{\rho\rho}(k,t)
$$

$$
- \int_0^t ds M_{\text{MCT}}(k, t - s) \partial_s G_{\rho\rho}(k,s),
$$

$$
M_{\text{MCT}}(k,t) = \frac{T}{2\rho_0 k^2} \int_q \left[ k \cdot q c(q) + k \cdot (k-q) c(|k-q|) \right]^2 G_{\rho\rho} G_{\rho\rho}. \quad (52)
$$

It is remarkable that the inter-particle potential $U(q)$, appearing in the original DK equation, completely cancels out, and in the final result the direct correlation function $c(q)$ emerges, which is due to the consistency between the dynamics and the statics.

In addition, we point out that if from the outset one takes a coarse-graining point of view and uses for the form of the free energy functional the Ramakrishnan-Yousouff form, i.e., $U(r) = -Tc(r)$ in Eq. (3), we would obtain the SMCT result in much simpler way. With $U(k) = -Tc(k)$, we have from the static input that $\Sigma_{\rho\rho}(k,0) = -(c(k) + U(k)/T) = 0$, and hence $G_{\rho\rho}(k,0) = 0$ and $G_{\rho\theta}(k,0) = 0$. Thus, in order to obtain the SMCT result, when we plug $U(q) = -Tc(q)$ and $G_{\rho\rho}(q,t) \approx 0$ into (45), we immediately obtain Eq. (50). In the same manner, when we substitute $U(q) = -Tc(q)$, $G_{\rho\rho}(q,t) \approx 0$ and $G_{\rho\theta}(q,t) \approx 0$ into Eq. (46), we obtain Eq. (51). We would thus have the SMCT result much more easily.

The straight one-loop expansion arising from the FDR-preserving one-loop expansion requires to be further worked out to yield the SMCT equation: coming back to the full dynamical equations (37–43), we can see that the SMCT result is retrieved when

- the memory (convolution) contributions to $\mathcal{M}(k,t)$ are ignored, and
- the memory contributions to $\mathcal{X}(k,t)$ and $\mathcal{Y}(k,t)$ are also ignored.

Both operations can easily be seen to be equivalent to the expansion in powers of $\lambda$ that we performed. Taking into account the full set of the dynamic equations with the one-loop self-energies is actually found to smear out the sharp transition predicted by the SMCT, making the equilibrium dynamics always ergodic, which is shown in the next subsection.

B. Absence of ENE transition

Here we examine the long-time-limit behavior of the dynamic equations to show that the full dynamic equations with the one-loop self-energies do not support dynamic transition into the nonergodic phase predicted by the SMCT.
We first introduce the notations for the long-time limit of the various quantities

\[ f_{\rho\rho}(k) \equiv \lim_{t \to \infty} G_{\rho\rho}(k, t), \]
\[ \sigma_{\rho\rho}(k) \equiv \lim_{t \to \infty} \Sigma_{\rho\rho}(k, t), \]
\[ x(k) \equiv \lim_{t \to \infty} \mathcal{X}(k, t), \text{etc} \] (53)

Extension to other quantities should be evident.

Let us begin with the time derivative of Eq. (30), which is given by

\[ \partial_t G_{\theta\rho}(k, t) = \Sigma_{\theta\rho}(k, t) \mathcal{X}(k, t) 
   + \int_0^t ds \Sigma_{\theta\rho}(k, t - s) \partial_s \mathcal{X}(k, s) 
   - \int_0^t ds [\partial_s \Sigma_{\theta\rho}(k, t - s)] \partial_s G_{\rho\rho}(k, s) \] (54)

Its long time limit is given by

\[ 0 = \sigma_{\theta\rho}(k) \mathcal{X}(k, 0) + \sigma_{\theta\rho}(k) \left( x(k) - \mathcal{X}(k, 0) \right) 
   - \bar{\sigma}_{\theta\rho}(k) \left( f_{\rho\rho}(k) - \rho_0 S(k) \right) 
   = \sigma_{\theta\rho}(k) x(k) - \bar{\sigma}_{\theta\rho}(k) \left( f_{\rho\rho}(k) - \rho_0 S(k) \right). \] (55)

Here \( \bar{\sigma}_{\theta\rho}(k) \equiv \lim_{t \to \infty} [\partial_t \Sigma_{\theta\rho}(k, t)] \) vanishes since it involves the time derivative of \( G_{\rho\rho}(k, t) \) (see Eq. (44)). Thus Eq. (55) reduces to

\[ \sigma_{\theta\rho}(k) x(k) = \sigma_{\theta\rho}(k) \left( K(k) f_{\rho\rho}(k) + f_{\theta\rho}(k) \right) = 0 \] (56)

The one-loop expression of the self-energy \( \Sigma_{\theta\rho}(k, t) = \Sigma_{2} + \rho_0 T k^2 \Sigma_{\theta\rho}(k, t) \) is easily obtained from Eq. (44) and Eq. (45) as

\[ \Sigma_{\theta\rho}(k, t) = \frac{T k^2}{2 \rho_0} \int_q G_{\rho\rho}(q, t) G_{\rho\rho}([k - q], t) 
   + \frac{T}{\rho_0^3} \int_q \mathbf{k} \cdot \mathbf{q} \left[ \frac{U(q)}{T} G_{\rho\rho}(q, t) + G_{\theta\rho}(q, t) \right] G_{\rho\rho}([k - q], t) 
   = \frac{T}{\rho_0^3} \int_q \mathbf{k} \cdot \mathbf{q} \left[ K(q) G_{\rho\rho}(q, t) + G_{\theta\rho}(q, t) \right] G_{\rho\rho}([k - q], t) \] (57)

where we used \( K(q) = U(q)/T + 1/\rho_0 \). Taking \( t \to \infty \) in (57) gives

\[ \sigma_{\theta\rho}(k) = \frac{T}{\rho_0^3} \int_q \mathbf{k} \cdot \mathbf{q} \left[ K(q) f_{\rho\rho}(q) + f_{\theta\rho}(q) \right] f_{\rho\rho}([k - q]). \] (58)

Therefore, Eq. (56) and Eq. (58) are incompatible with and thereby rule out the following nonergodic behaviors

(a) both \( f_{\rho\rho}(k) \neq 0 \) and \( f_{\theta\rho}(k) \neq 0 \), and
(b) both \( f_{\rho\rho}(k) \neq 0 \) and \( f_{\theta\rho}(k) = 0 \),

leaving out the other two possibilities:

(c) both \( f_{\rho\rho}(k) = 0 \) and \( f_{\theta\rho}(k) \neq 0 \), and

(d) both \( f_{\rho\rho}(k) = 0 \) and \( f_{\theta\rho}(k) = 0 \). We now show that the nonergodic behavior (c) is not supported by the dynamics. Taking the long-time limit of Eq. (39), we obtain

\[ f_{\theta\rho}(k) = \left( \frac{1}{\rho_0 S(k)} - K(k) \right) f_{\rho\rho}(k) 
   - \left( n(k) + \sigma_{\theta\rho}(k) \right) \left( f_{\rho\rho}(k) - \rho_0 S(k) \right) \] (59)

where \( n(k) \equiv \lim_{t \to \infty} N(k, t) \). On the other hand, the equation of motion for \( N(k, t) \) is easily obtained from Eq. (40) as

\[ \rho_0 T k^2 \partial_t N(k, t) = \partial_t \Sigma_{\theta\rho}(k, t) 
   - \int_0^t ds \Sigma_{\theta\rho}(k, t - s) \partial_s \Sigma_{\theta\rho}(k, s) 
   + \int_0^t ds \Sigma_{\theta\rho}(k, t - s) \partial_s N(k, s). \] (60)

where \( \Sigma_{\theta\rho}(k, 0) = 0 \) and \( N(k, 0) = 0 \) are used. The long-time limit of this equation is then given by

\[ 0 = -\left( \sigma_{\theta\rho}(k) \right)^2 + \sigma_{\theta\rho}(k) n(k) \] (61)

Now substituting \( f_{\rho\rho}(k) = 0 \) into both Eq. (59) and Eq. (61), we respectively get

\[ f_{\theta\rho}(k) = \rho_0 S(k) n(k) \] (62)

and

\[ 0 = \sigma_{\theta\rho}(k) n(k). \] (63)

The former equation follows since \( \sigma_{\theta\rho}(k) = 0 \) for \( f_{\rho\rho}(k) = 0 \) from (44). The latter one follows since \( \sigma_{\theta\rho}(k) \) is vanishing when \( f_{\rho\rho}(k) = 0 \).

If the possibility (c) were to hold, then we would have

\[ \sigma_{\theta\rho}(k) = \sigma_{1}(k) + \rho_0 T k^2 \Sigma_{\theta\rho}(k, t) \]

\[ = \sigma_{1}(k) = T^2 \int_q \mathbf{k} \cdot \mathbf{q} \mathbf{k} \cdot (\mathbf{k} - \mathbf{q}) f_{\theta\rho}(k) f_{\rho\rho}(k) \neq 0 \] (64)

where the last line results from the long-time limit of Eq. (46) with \( f_{\rho\rho}(k) = 0 \). The equation (63) and (64) would then imply that \( n(k) = 0 \), which in turn would mean \( \theta_{\rho}(k) = 0 \) from (62), which contradicts the initial assumption \( f_{\rho\rho}(k) \neq 0 \). Thus the possibility (c) cannot simultaneously satisfy Eqs. (62) and (63). We are thus only left with the ergodic behavior \( f_{\rho\rho}(k) = 0 \) and \( f_{\theta\rho}(k) = 0 \), which is the only possibility compatible with both Eq. (62) and Eq. (63).

Thus we find that the ENE phase transition predicted by the SMCT is absent when the full coupled dynamic equations with the one-loop self-energies are considered; the equilibrium dynamics within one-loop theory always remains ergodic. This dynamic aspect is a very much desired result since the FDR-preserving one-loop theory
presented here incorporates the SMCT, but at the same time goes beyond the SMCT, leading to the ergodic equilibrium dynamics. In order to see how the full dynamic equations round off the sharp transition of the SMCT, one needs to perform numerical integration of the full coupled dynamic equations with one-loop self-energies.

V. DISCUSSION AND CONCLUSION

A. Discussion

In this work, we reanalyzed the equilibrium dynamics of the DK equation for colloids via the FDR-preserving loop expansion for the new dynamic action incorporating an extra set of auxiliary fields \( \{\theta, \theta\} \), which is invariant under the linear field transformations. In doing so, we rectified the incorrect point made in [KK08]. The authors missed a delta function contribution to the response function \( G_{\rho\rho}(k, t) \), which led to the false linear dependence of \( G_{\theta\theta}(k, t) \) and \( G_{\theta\theta}(k, t) \) on \( G_{\rho\rho}(k, t) \), and equivalently to the invalid FDR \( G_{\rho\rho}(k, t) = i\delta(t)G_{\rho\rho}(k, t)/\rho_0 S(k) \). These invalid linear relations led the authors to obtain a nonperturbative closed equation for \( G_{\rho\rho}(k, t) \) alone in [KK08]. When the aforementioned missing contribution is reinstated, the full dynamic equations do not allow these simple relations between the correlation functions, and in turn do not lead to a single closed equation for \( G_{\rho\rho}(k, t) \). Instead, one really has to confront a coupled set of dynamic equations for the correlation functions \( G_{\rho\rho}(k, t), G_{\theta\rho}(k, t), \) and \( G_{\theta\theta}(k, t) \), which is ultimately subject to numerical integrations.

In the present work, we elucidated how one can retrieve the SMCT equation for \( G_{\rho\rho}(k, t) \) from the full set of dynamic equations with the one-loop self-energies. In this process, we witnessed an amazing cancellation of the bare interaction potential, leaving the resulting equation involving the direct correlation function only, which results from a consistency between statics and dynamics. We also find that the SMCT is obtained in a much simpler fashion if from the outset the coarse-graining viewpoint is taken using the Ramakrishnan-Yousouff form of the free energy, i.e., Eq. (3) with \( U(r) = -Te(r) \).

The most important recognition in the present study is that the one-loop theory is not tantamount to the SMCT, and goes beyond the SMCT; while the SMCT can be retrieved from the full dynamic equations with a well-defined approximation (and hence systematic corrections can be given) within the one-loop theory, the full one-loop dynamic equations themselves are found to remove the sharp ENE transition predicted by the SMCT. How exactly the full one-loop dynamic equations smear out this sharp transition is a remaining key question, which is left for further study. It would be most fascinating to see, upon solving numerically the full one-loop dynamic equations, an initial SMCT-like dynamical regime, followed by a relaxation towards equilibrium.

In the formulation of Jacquin and van Wijland [23], the same situation was found, i.e. that one-loop theory was not enough to obtain a closed equation on the density sector, and further simplifications (one-loop consistent substitutions) were needed, that led to the usual SMCT equation. Even more striking, upon application of this method to a very simple kinetically constrained model, which is not supposed to present an ENE transition of the MCT type, a SMCT-like equation was again recovered that led to a spurious ENE transition [30]. However in this model it is easy to see, with the same line of thoughts as that followed in the previous subsection, that the transition cannot exist in the full theory without projection on the density sector, in accordance with independent calculations [31].

Another line of work has been followed in order to incorporate momentum currents in the formalism, a process which was postulated to be at the origin of the cutoff of the ENE transition [7, 33], and later challenged [9]. In the present field theory setting extended to nonlinear fluctuating hydrodynamics, the situation has been instead shown [8, 34, 35] to be similar to the simple DK case, and thus we expect our conclusions to hold also in that case, even though a direct proof would be of course quite cumbersome.

The present findings seem to pose a challenge to our understanding on the nature of the SMCT. The DK equation describes the relaxation dynamics towards the well-defined equilibrium state, and therefore it is natural that the ergodic-nonergodic phase transition is absent in the DK equation. Then it would be interesting to ask in what approximation or in which limit one can get the SMCT. It would be interesting to show that the SMCT can be recovered from a more physically grounded limit such as \( N \to \infty \) in a \( N \)-component generalized DK equation.

The SMCT does not seem to correspond to a usual simple mean field theory, but rather to an unusual one with subtle aspects. It would thus be desirable to investigate the high dimensional version of the present set of equations, in order to see whether coherent results are obtained in this limit, which is usually associated to mean-field, since in infinite dimensions, every particle has an infinite number of nearest neighbors.

The SMCT equation in high dimensions was recently studied [36–39], and the equation was shown in [37, 38] to break down in this limit. Whether this is still the case in the hereby proposed dynamical equations remains to be investigated.

A major concern with the present formulation is that it is an expansion around a Gaussian theory, and not around the interaction-free case. Thus at each loop order, one is likely to break the exact evolution equation of the density in the NBG. For example one can see that the one-loop self energies do not cancel straightforwardly when evaluated at \( U = 0 \), and even though the solution of the full set of equations with \( U = 0 \) can well be given by the NBG result, it is not guaranteed by the formalism (although the full action treated exactly of course correctly treats the \( U = 0 \) limit, as was already shown in
obtained before further analyses of our set of equations. The NBG case is a strongly non-Gaussian theory, as one can already see at the static level, where all the cumulants of the density, and their associated vertex functions are non-zero in the NBG. This has also been checked in the dynamical framework in [15]. Thus nonperturbative relations like the first line of Eq. (28) (required by the consistency between statics and dynamics) are likely to hold, and to be broken in a naive loop expansion as usually performed. As an explicit example, for $U = 0$ case, the first line of Eq. (28) is not fulfilled by Eq.(43), the one-loop expression for $\Sigma_{\hat{q}\hat{p}}(k,t)$. As we indicated in the introduction, a possible remedy would be to directly work with the conjugate fields $\{\rho, \hat{\rho}\}$, and expand around the non-Gaussian NBG theory, utilizing the T-transformation. Another type of approach, chosen in [25] and [24], leave the potential to overcome these difficulties, but due to the specificities of the theories developed in these works, progress is likely to be difficult.

B. Conclusion

We have amended a previous work aiming at rederiving the SMCT equation from a field-theoretic treatment of the Brownian dynamics of an equilibrium glass. We found that the SMCT equation is not obtained from a simple one-loop self-consistent approximation, but rather from a further step of approximation, that we identified in a clear way. We have shown that taking from the outset a coarse-grained point of view by replacing $U$ by $-Tc(r)$ leads back again to the SMCT equation, thus reconciling our approach with previous works [10] based on this coarse-grained point of view. We have shown that the full one-loop dynamical equations does not support the existence of an ENE transition, which is not without reminding the results obtained in [14], and that the forceful projection of the dynamics on the density-density sector leads to this spurious ENE transition, in accordance with the results obtained in [23].

However, we emphasize the fact that the present loop expansion, as opposed to that in [23], is not an expansion around the NBG limit, and is thus likely to produce spurious results if taken as is. A deeper understanding of the high-order sum rules constraining the theory should be obtained before further analyses of our set of equations.

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Appendix A: Derivation of Eqs. (37) and (41)

We first define the Laplace transform

$$A^L(z) \equiv \mathcal{L}(A)(z) = \int_0^\infty dt \ e^{-zt} A(t) \quad (A1)$$

and the function $a(k)$ as:

$$a(k) = \rho_0 Tk^2 \quad (A2)$$

Dropping the wave-vector dependance, we rewrite (29) as

$$a\hat{\chi}(t) = -\dot{G}_{\rho\rho}(t) + \int_0^t ds \left[ \Sigma_{\hat{q}\hat{p}}(t-s)\chi(s) - \Sigma_{\hat{q}\hat{p}}(t-s)\dot{G}_{\rho\rho}(s) \right]. \quad (A3)$$

Its Laplace transform is given by

$$a\left(1 - \frac{1}{a} \Sigma_{\rho\rho}^L(z)\right)A^L(z) = -\left(1 + \Sigma_{\rho\rho}^L(z)\right)\mathcal{L}(\dot{G}_{\rho\rho})(z) \quad (A4)$$

A new memory function $\mathcal{M}(t)$ was defined in (36) as

$$\int_0^t ds \Sigma_1(t-s)\chi'(s) = -\int_0^t ds \mathcal{M}(t-s)\dot{G}_{\rho\rho}(s) \quad (A5)$$

Its Laplace transform is given by

$$\Sigma_1^L(z)\chi^L(z) = -\mathcal{M}^L(z)\mathcal{L}(\dot{G}_{\rho\rho})(z) \quad (A6)$$

Multiplying both sides of (A4) by $a^{-1}\Sigma_1^L(z)$ and using (A6), we obtain

$$\left(1 - \frac{1}{a} \Sigma_{\rho\rho}^L(z)\right)\mathcal{M}^L(z) = \frac{1}{a} \left(1 + \Sigma_{\rho\rho}^L(z)\right)\Sigma_1^L(z) \quad (A7)$$

Rearranging this equation gives

$$\mathcal{M}^L(z) = \frac{1}{a} \left[ \Sigma_1^L(z) + \Sigma_{\rho\rho}^L(z)\Sigma_1^L(z) + \Sigma_{\rho\rho}^L(z)\mathcal{M}^L(z) \right]. \quad (A8)$$

Putting this equation back in the time domain, we get

$$a\mathcal{M}(t) = \Sigma_1(t) - \int_0^t ds \Sigma_1(t-s)\dot{G}_{\rho\rho}(s) + \int_0^t ds \Sigma_{\hat{q}\hat{p}}(t-s)\mathcal{M}(s) \quad (A9)$$

This is Eq. (37) when the wavenumber is restored.

Another memory function $\mathcal{N}(t)$ was defined in (40) as

$$\int_0^t ds \Sigma_{\hat{q}\hat{p}}(t-s)\chi'(s) = -\int_0^t ds \mathcal{N}(t-s)\dot{G}_{\rho\rho}(s). \quad (A10)$$
Its Laplace transform is given by
\[ \Sigma_{\hat{\rho}p}(z)\mathcal{L}(z) = -\mathcal{N}^{L}(z)\mathcal{L}(\hat{G}_{\hat{\rho}p})(z). \tag{A11} \]
Let us multiply both sides of (A4) by \( a^{-1}\Sigma_{\hat{\rho}p}(z) \). Using (A11), we get
\[ \left(1 - \frac{1}{a} \Sigma_{\hat{\rho}p}(z)\right)\mathcal{N}^{L}(z) = \frac{1}{a} \left(1 + \Sigma_{\hat{\rho}p}(z)\right)\Sigma_{\hat{\rho}p}(z). \tag{A12} \]
Rearranging this equation gives
\[ \mathcal{N}^{L}(z) = \frac{1}{a} \left[\Sigma_{\hat{\rho}p}(z) + \Sigma_{\hat{\rho}p}(z)\Sigma_{\hat{\rho}p}(z) + \Sigma_{\hat{\rho}p}(z)\right]. \tag{A13} \]

This is transformed back in the time domain as:
\[ a\mathcal{N}(t) = \Sigma_{\hat{\rho}p}(t) - \int_{0}^{t} ds \Sigma_{\hat{\rho}p}(t - s)\Sigma_{\hat{\rho}p}(s) + \int_{0}^{t} ds \Sigma_{\hat{\rho}p}(t - s)\mathcal{N}(s). \tag{A14} \]
where the identity \( \Sigma_{\hat{\rho}p}(t) = -\Sigma_{\hat{\rho}p}(t) \) is used. This is (41) when the wavenumber is restored.

[1] S. P. Das, *Statistical Physics of Liquids at Freezing and Beyond*, (Cambridge University Press, Cambridge, 2011).
[2] L. Berthier and G. Biroli, Rev. Mod. Phys. **83**, 587 (2011).
[3] W. Götze, *Complex dynamics of glass-forming liquids: A mode-coupling theory* (Oxford University Press, Oxford, 2008).
[4] J.-P. Bouchaud, *The mode-coupling theory of supercooled liquids: Does it wear any clothes?*, http://www.condmatjournalclub.org/?p=1022 (2010).
[5] G. Brambilla et al, Phys. Rev. Lett. **102**, 085703 (2009).
[6] El Masri, D. et al, J. Stat. Mech. **2009** P07015.
[7] S. P. Das and G. F. Mazenko, Phys. Rev. A **34**, 2265 (1986).
[8] S. P. Das and G. F. Mazenko, Phys. Rev. E **79**, 021504 (2009).
[9] M. E. Cates and S. Ramaswamy, Phys. Rev. Lett. **96**, 135701 (2006).
[10] K. Kawasaki, Physica A **208**, 35 (1994).
[11] G. F. Mazenko and B. Kim, Adv. Chem. Phys. **78**, 129 (1990).
[12] B. Kim and K. Kawasaki, J. Stat. Mech. **2008** P02004.
[13] D. Dean, J. Phys. A: Math. Gen. **29**, 613 (1996).
[14] A. Andraeanov, G. Biroli, and A. Lefèvre, J. Stat. Mech. **2006** P07008.
[15] A. Velenich, C. Chamon, L. F. Cugliandolo and D. Kreimer, J. Phys. A: Math. Theor. **41**, 235002 (2008).
[16] J. Langer, Ann. Phys. **54**, 258 (1969).
[17] P. C. Martin, E. D. Siggia, and H. A. Rose, *Statistical dynamics of classical systems*, Phys. Rev. A **8**, 423 (1973).
[18] H. K. Janssen, *Field-theoretic method applied to critical dynamics*, in Dynamical Critical Phenomena and Related Topics vol. 104 (Charles P. Enz, ed.), Springer Berlin Heidelberg, Berlin, 1979 pp. 25–47.
[19] H. K. Janssen, Topics in modern statistical physics, ch. *On the renormalized field-theory of nonlinear critical relaxation*, World Scientific, Singapore, 1992.
[20] C. De Dominicis, J. Phys. Colloques **37**, C1–247 (1976).
[21] K. Kawasaki and S. Miyazima, Z. Phys. B **103**, 423 (1997).
[22] K. Miyazaki and D. R. Reichman, J. Phys. A: Math. Gen. **38**, L343 (2005).
[23] H. Jacquin and F. van Wijland, Phys. Rev. Lett. **106**, 210602 (2011).
[24] G. F. Mazenko, Phys. Rev. E **81**, 061102 (2010); Phys. Rev. E **83**, 041125 (2011); G. F. Mazenko, D. D. McCowan, and P. Spyridis, Phys. Rev. E **85**, 051105 (2012).
[25] G. Szamel, J. Chem. Phys. **127**, 084515 (2007); Prog. Theor. Exp. Phys. **2013** 021201.
[26] Andersen, H. C. J. Phys. Chem. B **107**, 10226 (2003).
[27] J. P. Hansen and I. R. McDonald, *Theory of simple liquids*, Academic Press, London, 1986.
[28] B. Cichocki and W. Hess, Physica A **141**, 475 (1987).
[29] K. Kawasaki, Physica A **215**, 61 (1995).
[30] H. Jacquin, PhD. Thesis (2012) http://tel.archives-ouvertes.fr/tel-00734605
[31] S. Whitelam, L. Berthier, and J. P. Garrahan, Phys. Rev. E **71**, 026128 (2005).
[32] P. Mayer, K. Miyazaki, and D. R. Reichman, Phys. Rev. Lett. **97**, 095702 (2006).
[33] W. Götze and L. Sjögren, Z. Phys. B **65**, 415 (1987).
[34] T. H. Nishino and H. Hayakawa, Phys. Rev. E **78**, 061502 (2008).
[35] J. Yeo, Phys. Rev. E **80**, 051501 (2009).
[36] B. Schmid and R. Schilling, Phys. Rev. E **81**, 041502 (2010).
[37] A. Ikeda and K. Miyazaki, Phys. Rev. Lett. **104**, 255704 (2010).
[38] P. Charbonneau, A. Ikeda, G. Parisi, and F. Zamponi, Phys. Rev. Lett. **107**, 185702 (2011).
[39] P. Charbonneau, A. Ikeda, G. Parisi, and F. Zamponi, Proc. Nat. Acad. Sci. **109**, 13939 (2012).