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LETTER TO THE EDITOR

Mode-coupling theory and the fluctuation–dissipation theorem for nonlinear Langevin equations with multiplicative noise

Kunimasa Miyazaki and David R Reichman

Department of Chemistry, Columbia University, 3000 Broadway, New York, NY 10027, USA

E-mail: km2239@columbia.edu

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Abstract

In this letter, we develop a mode-coupling theory for a class of nonlinear Langevin equations with multiplicative noise using a field-theoretic formalism. These equations are simplified models of realistic colloidal suspensions. We prove that the derived equations are consistent with the fluctuation–dissipation theorem. We also discuss the generalization of the result given here to real fluids, and the possible description of supercooled fluids in the ageing regime. We demonstrate that the standard idealized mode-coupling theory is not consistent with the FDT in a strict field-theoretic sense.

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1. Introduction

Mode-coupling theory (MCT) has been a useful first-principles approach for studying the dynamics of supercooled liquids (see, for example, [1]). MCT was originally derived using projection operator methods together with several uncontrolled approximations. The theory has been successful in providing a quantitative account of many experimental and numerical observations, such as nonergodic parameters and structural relaxation exponents [2]. The approximate nature of the conventional MCT, however, has restricted its validity to description of only: (i) two-point correlation functions[1], (ii) for systems at equilibrium and (iii) at relatively high temperatures below which the theory predicts a spurious glass transition.

[1] See, however, Biroli and Bouchaud [3]. Note that, strictly speaking, the mode-coupling approach taken here is of the field-theoretic, diagrammatic variety. This is what allows the authors to compute multi-point correlation functions, something that is difficult to do in a useful manner from the projection operator approach. The field theory used in the work of Biroli and Bouchaud is that of Das and Mazenko [4] which assumes the FDT from the start. The work presented here is a first step beyond such a simplified treatment.

Experimental and numerical studies, on the other hand, have provided us with a rich dynamics, none of which conventional MCT can explain. For example, simulations and experiments of supercooled fluids have revealed the existence of the correlated *local* dynamical heterogeneities (see, for example, [5]), and dynamical scaling [6]. Another example is supercooled liquids brought out of equilibrium by quenching the system to low temperature or by adding shear flow. Here many experiments and simulations have shown violation of the fluctuation–dissipation theorem (FDT) and the existence of the effective temperatures [7–9]. A microscopic theory which goes beyond MCT is desirable to account these phenomena. For such purposes, a systematic field-theoretic approach is a good candidate. Such a field theoretical perturbation scheme for dynamical processes has been developed by Martin, Siggia and Rose (MSR) [10]. This approach is advantageous over the projection operator technique in that: (i) it is conceptually straightforward to extend MCT-type equation to incorporate higher order moments of fluctuations, (ii) it is useful for calculating multipoint correlation functions which are essential observables to monitor the dynamical heterogeneities in the supercooled fluids and (iii) it enables one to treat the correlation function and the response function (which are related by the FDT if equilibrium holds) on an equal footing and, therefore, it is a powerful tool for the treatment of nonequilibrium systems.

Conventional MCT used for glassy or disordered systems is believed to be equivalent with renormalized perturbation theory [11] without vertex corrections within the standard loop expansion of the MSR formalism [10]. This is true for a certain class of disordered systems such as the p-spin spin glass models [12]. For supercooled fluids, however, this field theory has never been systematically used to derive the MCT equation *even at equilibrium*. The primary obstacle is that, although the original equations of motion satisfy the FDT at equilibrium, this does not imply that an arbitrary perturbation scheme also preserves the constraints of the FDT at each order of the expansion. Derivations of MCT for supercooled fluids from the field-theoretic point of view have been discussed by several authors [4, 13, 14] but either the FDT has been assumed (rather than consistently derived) [4, 14] or a certain model has to be introduced for the derived equation to guarantee the FDT [13]. Difficulties in deriving the MCT equations and extending them to higher order by a systematic loop expansion for supercooled fluids are due to certain properties of the nonlinearities in the microscopic Langevin equation. These difficulties do not exist in the schematic p-spin models. To illustrate the difficulties, let us consider a Langevin equation which describes dynamics of the density field, $\rho(\mathbf{r}, t)$, of the dense colloidal suspension, as an example [15];

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} = D\nabla \cdot \left\{ \nabla \rho(\mathbf{r}, t) - \rho(\mathbf{r}, t)\nabla \int \mathrm{d}\mathbf{r}' c(\mathbf{r} - \mathbf{r}')\delta\rho(\mathbf{r}', t) \right\} + f_\rho(\mathbf{r}, t), \tag{1}$$

where D is the diffusion coefficient which is assumed to be a constant, $\delta\rho(\mathbf{r}, t) = \rho(\mathbf{r}, t) - \rho_0$ with $\rho_0 = \langle \rho(\mathbf{r}, t) \rangle$ is a density fluctuation, $f_\rho(\mathbf{r}, t)$ is a random noise and $c(\mathbf{r})$ is the direct correlation function. The second term in the brackets accounts for the interaction between the particles and is the source of the nonlinearity of the Langevin equation[2]. Equation (1) is known to exhibit the glassy properties at high densities. It has also been shown that, in equilibrium, this equation can be reduced to a standard MCT equation in the overdamped limit, using projection operators [18–20]. In order to see the difficulty in applying the field-theoretic MSR, let us rewrite equation (1) in a following form:

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} = \int \mathrm{d}\mathbf{r}' L_{\rho(\mathbf{r})\rho(\mathbf{r}')} \frac{\delta S}{\delta \rho(\mathbf{r}')} + f_\rho(\mathbf{r}, t), \tag{2}$$

[2] Equation (1) is already coarse-grained in a sense that the bare interaction potential is replaced with the effective potential $-c(\mathbf{r})/k_\mathrm{B}T$. See [16, 17].

where

$$L_{\rho(\mathbf{r})\rho(\mathbf{r}')} = {k_\mathrm{B}}^{-1} D\nabla \cdot \nabla' \rho(\mathbf{r}, t)\delta(\mathbf{r} - \mathbf{r}') \tag{3}$$

is the Onsager coefficient and S is the entropy of the whole system which is given as a functional of the density by

$$S = k_\mathrm{B}\left\{-\int \mathrm{d}\mathbf{r}\, \rho(\mathbf{r})[\ln\{\rho(\mathbf{r})/\rho_0\} - 1] + \frac{1}{2}\int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' c(|\mathbf{r} - \mathbf{r}'|)\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')\right\}. \tag{4}$$

The random field $f_\rho(\mathbf{r}, t)$ satisfies

$$\langle f_\rho(\mathbf{r}, t) f_\rho(\mathbf{r}', t')\rangle = 2k_\mathrm{B} L_{\rho(\mathbf{r})\rho(\mathbf{r}')}\delta(t - t'). \tag{5}$$

One sees that there are two types of nonlinearities entangled in equation (2). One is due to the thermodynamic force $\delta S/\delta\rho(\mathbf{r})$, which is nonlinear in $\delta\rho$. Another is the density-dependent Onsager coefficient which, through equation (5), makes the random force a nonlinear function of the density, producing *multiplicative noise*. These properties are quite general for Langevin equations for realistic fluids. As elucidated in the next section, these two nonlinearities are the origin of difficulties which hamper the field theoretical approach for realistic fluids.

In this letter, we develop the tools to treat some of the difficult nonlinearities discussed above, including multiplicative noise. Our goal is to derive MCT-type equations which satisfy the FDT at the lowest level of the loop expansion if the system is at equilibrium. This goal is an important prerequisite condition when we set out to explore nonequilibrium systems. Our theory will serve as a first step, however incremental, to prepare for the development of a field theory for supercooled fluids which goes beyond the conventional MCT and is capable of exploring, for example, nonequilibrium systems, the effect of higher order loops and multipoint correlation functions. In particular, we will show that:

(a) Care must be exercised in field theoretical derivations of the response of a system described by equation (1) to an external field since the response function is not trivially connected to the propagator in general. This fact has been ignored in past works [21].
(b) The multiplicative noise term is essential for a proper treatment of the memory term. This point has been overlooked in past works [14], as the proper field theoretic treatment of this term is subtle.
(c) A standard one-loop treatment can lead to the usual MCT of Götze and coworkers [1, 2] at the expense of satisfying the FDT. This will lead us to consider a slightly simpler model for which the associated self-consistent one-loop theory presents the FDT. This model will allow us to make connection with the work of Schmitz *et al* [13].

In the next section, we give more detailed accounts of the background and motivation of the present work. Section 3 is devoted to derivation of MCT for multiplicative noise using the MSR method. Consistency with the FDT is discussed in sections 4 and 5. Perspectives for developing MCT for the nonequilibrium case are discussed in section 6.

2. Background

In order to make the argument general, let us consider a classical stochastic dynamical process of a field variable $x_i(t)$, where i is an index that denotes the type of field (such as the density) and coordinates (such as positions or wave vectors) which may be either discrete or continuous. If the system is macroscopically at equilibrium, $x_i(t)$ obeys a nonlinear Langevin equation of a general form:

$$\dot{x}_i = M_{i\alpha}\frac{\partial S}{\partial x_\alpha} + L_{i\alpha}\frac{\partial S}{\partial x_\alpha} + f_i \equiv K_{i\alpha}\frac{\partial S}{\partial x_\alpha} + f_i, \tag{6}$$

where a sum over the repeated Greek indices is assumed. S is the entropy of the entire system. $K_{ij} \equiv M_{ij} + L_{ij}$ is a kinetic coefficient which generally depends on $\mathbf{x}$. $M_{i\alpha}\partial S/\partial x_\alpha$ represents the reversible term where $M_{ij} = -M_{ji}$ is an antisymmetric matrix. $L_{i\alpha}\partial S/\partial x_\alpha$ represents the irreversible term, where L_{ij} is the ($\mathbf{x}$-dependent) Onsager coefficient. $f_i(\mathbf{x}, t)$ is a random noise which satisfies

$$\langle f_i(\mathbf{x}, t) f_j(\mathbf{x}', t')\rangle_{\mathbf{x}(t)=\mathbf{x}} = 2k_B L_{ij}(\mathbf{x})\delta(t - t'), \tag{7}$$

where $\langle\cdots\rangle_{\mathbf{x}(t)=\mathbf{x}}$ denotes the conditional average in which the ensemble average is taken with a fixed value of $\mathbf{x}(t) = \mathbf{x}$ at time t. The fact that the Onsager coefficient is a function of $\mathbf{x}$ means that the random noise is also the function of $\mathbf{x}$, i.e., it is multiplicative [22]. Equation (6) is the general expression for dynamical systems whose stationary distribution function in the absence of nonequilibrium constraints is given by the equilibrium ensemble. Examples include the (fluctuating) Navier–Stokes equation [23, 24] and the nonlinear diffusion equation (equation (1)). The response function $\chi_{ij}(t)$ is defined as the response to a time-dependent external force $\mathbf{F}(t)$ by

$$\langle \Delta x_i(t)\rangle_F = \int_{-\infty}^{t} \mathrm{d}t' \chi_{i\alpha}(t - t') F_\alpha(t'), \tag{8}$$

where $\langle \Delta x_i(t)\rangle_F$ is the deviation of $\langle x_i(t)\rangle$ from its equilibrium value due to the external force. The FDT asserts that $\chi_{ij}(t)$ is related to the correlation function in the absence of $\mathbf{F}(t)$, $C_{ij}(t) = \langle x_i(t)x_j(0)\rangle$, by

$$\chi_{ij}(t) = -\frac{1}{k_B T}\frac{\mathrm{d}C_{ij}(t)}{\mathrm{d}t} \qquad \text{for} \quad t \geqslant 0. \tag{9}$$

The FDT is proved easily using linear response theory [25]. The FDT is one of the strongest and the most robust statements of equilibrium statistical physics and it holds for any dynamical processes, classical or quantum, linear or nonlinear, as long as the system is stationary, satisfies the condition of detailed balance, and the perturbation is small enough.

The MSR formalism allows us for the use of a systematic loop expansion for the solution of the nonlinear Langevin equation in terms of the moments. MCT is regarded as the lowest order self-consistent approximation with no vertex correction in the MSR formalism. Deker *et al* [21] have proven that the FDT holds at each order of the loop expansion for three special classes of dynamical processes: 'class A' where, in equation (6), $M_{ij}(\mathbf{x}) = 0$, L_{ij} is a constant (independent of $\mathbf{x}$) and thus the noise is additive. The nonlinearity of the Langevin equation originates from the entropy S. 'Class B' where L_{ij} is constant and the entropy is a quadratic function of $\mathbf{x}$. The reversible matrix $M_{ij}(\mathbf{x})$ depends on $\mathbf{x}$ which is the origin of the nonlinearity. The equations discussed by Kawasaki to describe dynamical critical phenomena [26] belong to this class. 'Class C' involves Hamiltonian systems which do not have an irreversible part.

The problems is that, as discussed in the introduction, even equation (1), which describes the dynamics of supercooled fluids, does not belong to any of the classes listed above. The nonlinear term in equation (1) originates from the combination of the non-quadratic term of the entropy and the variable dependence of the Onsager coefficient. Extensions of the MSR formalism to more general cases have been discussed in [14, 27–29] but derivations of MCT equations have not been given so far. Since Deker's classifications do not cover these dynamical processes, it is convenient to re-categorize the nonlinear stochastic processes in a slightly more general way than classes A–C of Deker:

Class I. The nonlinearity comes solely from the entropy. K_{ij} is independent of $\mathbf{x}$. The noise is additive. The mean-field model of p-spin spin glasses also belongs to this type [12]. Again, it is trivial to show that the FDT holds at each order of the loop expansion.

Class II. The entropy is a quadratic function of $\mathbf{x}$ but $K_{ij}(\mathbf{x})$ is dependent on $\mathbf{x}$. $L_{ij}(\mathbf{x})$ can also be a function of $\mathbf{x}$ and, therefore, the noise can be multiplicative.

Class III. The entropy is an arbitrary function of $\mathbf{x}$ and $K_{ij}(\mathbf{x})$ is dependent on $\mathbf{x}$. Real fluids including the one described by equation (1) belong to this type.

In this letter, we shall focus here on class II. Here we restrict ourselves to the simplest situation where $K_{ij}(\mathbf{x})$ is a linear function of $\delta\mathbf{x}$:

$$K_{ij}(\mathbf{x}) = K_{ij}^{(0)} + K_{ij,\alpha}^{(1)}\delta x_\alpha, \tag{10}$$

because this is the most important case in the context of the glass transition. We shall show, for this class of Langevin equations, that MCT is consistent with the FDT even with the presence of multiplicative noise. We believe that this conclusion is valid for arbitrary function of $K_{ij}(\mathbf{x})$ of class II. We will end this work with some comments on the more interesting class III case which is problematic from the standpoint of the FDT within the MCT approximation.

3. MSR formalism for processes with multiplicative noise

In this section, we shall develop the MSR method for the class II case. In class II, the entropy in equation (6) is given by a quadratic form

$$S = S_0 + \tfrac{1}{2}\Omega_{\alpha\beta}\delta x_\alpha \delta x_\beta, \tag{11}$$

where $\delta\mathbf{x} = \mathbf{x} - \langle\mathbf{x}\rangle$ and $\Omega_{ij}^{-1} = -k_{\mathrm{B}}C_{ij}^{-1}(t=0)$ is the inverse of the equal time correlation function. Such an approximation is, perhaps, not as crude as it may superficially appear. Indeed, it has been shown via direct simulation that both simple [30] and complex liquids [31] have Gaussian density fluctuations over a wide range of length scales. Equation (11), within the canonical ensemble, is a precise statement of this approximation. The Fokker–Planck equation for the probability density function $P(\mathbf{x}, t)$ equivalent with equation (6) is written as

$$\frac{\partial P(\mathbf{x},t)}{\partial t} = -\frac{\partial}{\partial x_\alpha}\left\{K_{\alpha\beta}(\mathbf{x})\frac{\partial S}{\partial x_\beta} - k_{\mathrm{B}}L_{\alpha\beta}(\mathbf{x})\frac{\partial}{\partial x_\beta}\right\}P(\mathbf{x},t) \equiv \mathcal{T}P(\mathbf{x},t). \tag{12}$$

For the detailed balance condition to be satisfied, $M_{ij}(\mathbf{x})$ must satisfy the following condition (potential condition [25]): $\partial M_{i\alpha}(\mathbf{x})/\partial x_\alpha = 0$. We shall also assume the similar incompressible condition for $L_{ij}(\mathbf{x})$: $\partial L_{i\alpha}(\mathbf{x})/\partial x_\alpha = 0$. These two conditions are satisfied for most hydrodynamic equations including equation (1) (see equation (3)). The latter condition is especially useful because it enables us to avoid distinguishing between the Ito and Stratonovich interpretations which are associated with the multiplicative noise [22]. Using equations (10) and (11), equation (6) can be rewritten as

$$\dot{x}_i = \mu_{i\alpha}x_\alpha + \tfrac{1}{2}\mathcal{V}_{i\alpha\beta}x_\alpha x_\beta + f_i. \tag{13}$$

Hereafter we shall omit the 'δ' in front of $\mathbf{x}$. In equation (13), $\mu_{ij} = K_{i\alpha}^{(0)}\Omega_{\alpha j}$ is a bare transport coefficient and $\mathcal{V}_{ijk} = \mathcal{V}_{ikj}$ is the symmetrized vertex defined by

$$\mathcal{V}_{ijk} = M_{i\alpha,j}^{(1)}\Omega_{\alpha k} + M_{i\alpha,k}^{(1)}\Omega_{\alpha j} + L_{i\alpha,j}^{(1)}\Omega_{\alpha k} + L_{i\alpha,k}^{(1)}\Omega_{\alpha j} \equiv \mathcal{M}_{ijk} + \mathcal{L}_{ijk}, \tag{14}$$

where $\mathcal{M}_{ijk}$ and $\mathcal{L}_{ijk}$ are reversible and irreversible contributions of $\mathcal{V}_{ijk}$, respectively. $\mathcal{M}_{ijk}$ satisfies the cyclic condition given by

$$\Omega_{i\alpha}\mathcal{M}_{\alpha jk} + \Omega_{j\alpha}\mathcal{M}_{\alpha ki} + \Omega_{k\alpha}\mathcal{M}_{\alpha ij} = 0. \tag{15}$$

This is proved by using the condition that the reversible part does not contribute to the entropy production.

Following the standard MSR procedure [10], we shall introduce a spinor $\mathbf{z} \equiv (\mathbf{x}, \hat{\mathbf{x}})$, where $\hat{x}_i \equiv -\partial/\partial x_i$. It is convenient to define the generating function for $\mathbf{z}$ by

$$W[\boldsymbol{\xi}] \equiv \ln\left\langle \exp_+\left[\int \mathrm{d}t\, \boldsymbol{\xi}\cdot\mathbf{z}\right]\right\rangle, \tag{16}$$

where $\boldsymbol{\xi} = (\boldsymbol{\eta}, \hat{\boldsymbol{\eta}})$ is the auxiliary field conjugate to $\mathbf{z}$ which will be eventually set to zero. '$\exp_+$' implies the time ordering which aligns the quantities with larger t on the left. We define the cumulant function $\mathcal{G}_\xi(1,\ldots,n) = \langle\langle z(1)\cdots z(n)\rangle\rangle$ by

$$\mathcal{G}_\xi(1,\ldots,n) = \frac{\delta^n W[\boldsymbol{\xi}]}{\delta\xi(1)\cdots\delta\xi(n)}. \tag{17}$$

The index number $1 = (i, t, \pm)$, etc represents the index for field variables i, time t, and the index of the spinor defined by $\mathbf{z}(+) = \mathbf{x}$ and $\mathbf{z}(-) = \hat{\mathbf{x}}$, respectively. Let us construct the equation of motion for the first cumulant, $\langle\langle z(1)\rangle\rangle$. Substituting equation (13) into equation (17) for $n = 1$, we obtain the Schwinger equation:

$$\frac{\mathrm{d}\langle\langle z(1)\rangle\rangle}{\mathrm{d}t} = -\langle\langle[\mathcal{T}, z(1)]\rangle\rangle + \mathrm{i}\sigma(1,\underline{1})\xi(\underline{1}), \tag{18}$$

where a sum over the repeated underlined indices is assumed. $[\cdots]$ is the commutator, $\mathcal{T}$ is the Fokker–Planck operator defined by equation (12) represented in terms of $(\mathbf{x}, \hat{\mathbf{x}})$, and

$$\mathrm{i}\sigma(1,2) = \begin{pmatrix} \mathbf{0} & -\mathbf{1} \\ \mathbf{1} & \mathbf{0} \end{pmatrix}\delta(t_1 - t_2). \tag{19}$$

The explicit expression of equation (18) is given by

$$\begin{aligned}
\frac{\mathrm{d}\langle\langle x_i\rangle\rangle}{\mathrm{d}t} &= \hat{\eta}_i + \mu_{i\alpha}\langle\langle x_\alpha\rangle\rangle + \frac{1}{2}\mathcal{V}_{i\alpha\beta}\{\langle\langle x_\alpha x_\beta\rangle\rangle + \langle\langle x_\alpha\rangle\rangle\langle\langle x_\beta\rangle\rangle\} \\
&\quad + 2k_{\mathrm{B}}\Big[L^{(0)}_{i\alpha}\langle\langle\hat{x}_\alpha\rangle\rangle + L^{(1)}_{i\alpha,\beta}\{\langle\langle\hat{x}_\alpha x_\beta\rangle\rangle + \langle\langle\hat{x}_\alpha\rangle\rangle\langle\langle x_\beta\rangle\rangle\}\Big] \\
\frac{\mathrm{d}\langle\langle\hat{x}_i\rangle\rangle}{\mathrm{d}t} &= -\eta_i - {}^{\mathrm{t}}\mu_{i\alpha}\langle\langle\hat{x}_\alpha\rangle\rangle - \mathcal{V}_{\alpha\beta i}\{\langle\langle\hat{x}_\alpha x_\beta\rangle\rangle + \langle\langle\hat{x}_\alpha\rangle\rangle\langle\langle x_\beta\rangle\rangle\} + \mathcal{V}_{\alpha\alpha i} \\
&\quad - k_{\mathrm{B}}L^{(1)}_{\alpha\beta,i}\{\langle\langle\hat{x}_\alpha\hat{x}_\beta\rangle\rangle + \langle\langle\hat{x}_\alpha\rangle\rangle\langle\langle\hat{x}_\beta\rangle\rangle\},
\end{aligned} \tag{20}$$

where ${}^{\mathrm{t}}\mu_{ij}$ is the transverse of μ_{ij}. The last terms in these equations are due to the multiplicative noise. Equation (20) is written in short as

$$\mathcal{G}_0^{-1}(1,\underline{1})\langle\langle z(\underline{1})\rangle\rangle = \xi(1) + C(1) + \tfrac{1}{2}\gamma_3(1,\underline{1},\underline{2})\{\langle\langle z(\underline{1})z(\underline{2})\rangle\rangle + \langle\langle z(\underline{1})\rangle\rangle\langle\langle z(\underline{2})\rangle\rangle\}, \tag{21}$$

where $C(1) \equiv (\mathcal{V}_{\alpha\alpha i}, 0)$ is a constant which does not contribute to the following arguments and $\mathcal{G}_0(1,2)$ is the bare propagator whose inverse is written as

$$\mathcal{G}_0^{-1}(1,2) = \mathrm{i}\sigma(1,2)\frac{\mathrm{d}}{\mathrm{d}t_2} + \gamma_2(1,2) \tag{22}$$

with the symmetric matrix $\gamma_2(1,2)$ defined by

$$\gamma_2(1,2) = \begin{pmatrix} 0 & -{}^{\mathrm{t}}\boldsymbol{\mu} \\ -\boldsymbol{\mu} & -2k_{\mathrm{B}}\mathbf{L}^{(0)} \end{pmatrix}\delta(t_1 - t_2). \tag{23}$$

The non-zero components of $\gamma_3(1,2,3)$ are

$$\begin{cases} \gamma_3(i_1,t_1,-;i_2,t_2,+;i_3,t_3,+) = \mathcal{V}_{i_1i_2i_3}\delta(t_1-t_2)\delta(t_1-t_3) \\ \gamma_3(i_1,t_1,-;i_2,t_2,-;i_3,t_3,+) = 2k_{\mathrm{B}}L^{(1)}_{i_1i_2,i_3}\delta(t_1-t_2)\delta(t_1-t_3) \end{cases} \tag{24}$$

and its permutation of the indices $(1,2,3)$. Note that $\gamma_3(1,2,3)$ is a fully symmetric tensor. The second moment $\mathcal{G}(1,2) = \mathcal{G}_{\xi=0}(1,2) = \langle z(1)z(2)\rangle$ is given by taking the derivative of equation (21) with respect to $\langle\langle z(2)\rangle\rangle$ using equation (17) and then turning off $\boldsymbol{\xi} = 0$:

$$\mathcal{G}^{-1}(1,2) = \mathcal{G}_0^{-1}(1,2) - \Sigma(1,2), \tag{25}$$

where $\Sigma(1,2)$ is the self-energy. By neglecting the vertex correction, we obtain the MCT expression for the self-energy:

$$\Sigma(1,2) \simeq \tfrac{1}{2}\gamma_3(1,\underline{1},\underline{2})\mathcal{G}(\underline{1},\underline{3})\mathcal{G}(\underline{2},\underline{4})\gamma_3(2,\underline{3},\underline{4}). \tag{26}$$

We can write the components of these matrices as

$$\mathcal{G}(1,2) \equiv \begin{pmatrix} \mathbf{C} & \mathbf{G} \\ \mathbf{G}^\dagger & \mathbf{0} \end{pmatrix} \qquad \text{and} \qquad \Sigma(1,2) \equiv \begin{pmatrix} \mathbf{0} & \mathbf{E}^\dagger \\ \mathbf{E} & \mathbf{D} \end{pmatrix}, \tag{27}$$

where '†' represents the Hermitian conjugate defined by $A^\dagger_{ij}(t-t') = A^*_{ji}(t'-t)$. From the structure of equation (25), it is straightforward to show that $\mathcal{G}(-,-) = \Sigma(+,+) = 0$. $C_{ij}(t-t') = \langle x_i(t)x_j(t')\rangle$ is the correlation function and $G_{ij}(t-t') = \langle x_i(t)\hat{x}_j(t')\rangle$ is the propagator which describes the response of the system to the random noise. For $t > 0$, the equations for $\mathbf{C}(t)$ and $\mathbf{G}(t)$ can be written explicitly using equations (22)–(25) as

$$\frac{\mathrm{d}C_{ij}(t)}{\mathrm{d}t} = \mu_{i\alpha}C_{\alpha j}(t) + \int_{-\infty}^{t} \mathrm{d}t_1 E_{i\alpha}(t-t_1)C_{\alpha j}(t_1) + \int_{-\infty}^{0} \mathrm{d}t_1 D_{i\alpha}(t-t_1)G^\dagger_{\alpha j}(t_1) \tag{28}$$

$$\frac{\mathrm{d}G_{ij}(t)}{\mathrm{d}t} = \mu_{i\alpha}G_{\alpha j}(t) + \int_{0}^{t} \mathrm{d}t_1 E_{i\alpha}(t-t_1)G_{\alpha j}(t_1) \tag{29}$$

with the self-energies given by

$$\begin{cases} E_{ij}(t) = \mathcal{V}_{i\alpha\beta}G_{\alpha\lambda}(t)C_{\beta\mu}(t)\mathcal{V}_{\lambda\mu j} + k_\mathrm{B}\mathcal{V}_{i\alpha\beta}G_{\alpha\lambda}(t)G_{\beta\mu}(t)L^{(1)}_{\lambda\mu,j} \\ D_{ij}(t) = \frac{1}{2}\mathcal{V}_{i\alpha\beta}C_{\alpha\lambda}(t)C_{\beta\mu}(t)\mathcal{V}_{j\lambda\mu} + 2k_\mathrm{B}\mathcal{V}_{i\alpha\beta}G_{\alpha\lambda}(t)C_{\beta\mu}(t)L^{(1)}_{j\lambda,\mu}, \end{cases} \tag{30}$$

where use has been made of the causality condition: $\mathbf{G}(t) = 0$ for $t < 0$. The terms containing $L^{(1)}_{ij,k}$ in equation (30) originate from the multiplicative noise. Similar terms were derived by Kawasaki *et al* [14] but they were disregarded and their importance was not addressed. *Note that the propagator* $\mathbf{G}(t)$ *represents the response to the noise but it is not the response to the external force defined by equation (8).* The response function is obtained by evaluating the linear response of the average $\langle x_i(t)\rangle$ to the external force $\mathbf{F}(t)$. The term associated with the external force is introduced naturally by replacing the entropy with the one associated with the work done by the force as

$$S_F = S + \frac{\mathbf{x}\cdot\mathbf{F}}{T}. \tag{31}$$

Inserting this expression into the entropy term in equation (12) and taking the leading order of the formal solution, it is straightforward to derive the expression for the response function. The result is

$$\begin{aligned} \chi_{ij}(t-t') &= \frac{1}{T}\langle x_i(t)\hat{x}_\alpha(t')K_{\alpha j}(\mathbf{x}(t'))\rangle \\ &= \frac{1}{T}\langle x_i(t)\hat{x}_\alpha(t')\rangle K^{(0)}_{\alpha j} + \frac{1}{T}\langle x_i(t)\hat{x}_\alpha(t')x_\beta(t')\rangle K^{(1)}_{\alpha j,\beta}. \end{aligned} \tag{32}$$

The three-point correlation function $\langle x_i(t)\hat{x}_\alpha(t')x_\beta(t')\rangle$ in this expression is calculated in the same spirit as derivation of equation (26). Up to the one-loop level, neglecting the vertex correction, it is written as $\langle z(1)z(2)z(3)\rangle \simeq \mathcal{G}(1,\underline{1})\mathcal{G}(2,\underline{2})\mathcal{G}(3,\underline{3})\gamma_3(\underline{1},\underline{2},\underline{3})$. Substituting equation (24) into this, one obtains the MCT expression of the response function:

$$\chi_{ij}(t) = \frac{1}{T}G_{i\alpha}(t)K^{(0)}_{\alpha j} + \frac{1}{T}\int_0^t \mathrm{d}t_1 G_{i\alpha}(t-t_1)\mathcal{V}_{\alpha\beta\gamma}G_{\beta\lambda}(t_1)C_{\gamma\mu}(t_1)K^{(1)}_{\lambda j,\mu}. \tag{33}$$

In previous works, the propagator $T^{-1}\mathbf{G}(t)\cdot\mathbf{K}^{(0)}$ has been called the response function [21]. But as discussed above, it is not identical to the full response function in general. They become identical only if the kinetic coefficient K_{ij} is a constant, i.e., for class I.

4. FDT and MCT for class II

In this section, we prove that the correlation and response functions given by equations (28), (29) and (33) satisfy the FDT, equation (9). Deker *et al* have shown that, for the MCT equation of class B, there is a simple relation between the correlation function and the propagator if L_{ij} is a constant or $L^{(1)}_{ij,k} = 0$ [21]:

$$\mathbf{G}(t) = \theta(t)\mathbf{C}(t) \cdot \mathbf{C}^{-1}(0), \tag{34}$$

where $\theta(t)$ is the Heaviside function. We prove that this is also true when $L^{(1)}_{ij,k} \neq 0$ or the noise is multiplicative as follows: taking the time derivative of both sides of equation (34) and substituting the equation for the correlation function, equation (28), we have

$$\frac{\mathrm{d}\mathbf{G}}{\mathrm{d}t} = \mathbf{1} + \boldsymbol{\mu} \cdot \mathbf{G} + \theta(t)\{\mathbf{E} \otimes \mathbf{C} + \mathbf{D} \otimes \mathbf{G}^{\dagger}\} \cdot \mathbf{C}^{-1}(0), \tag{35}$$

where $\mathbf{A} \otimes \mathbf{B} \equiv \int_{-\infty}^{\infty} \mathrm{d}t_1 A_{i\alpha}(t - t_1) B_{\alpha j}(t_1)$. The terms containing $\mathcal{M}_{ijk}$ in $\mathbf{E} \otimes \mathbf{C} \cdot \mathbf{C}^{-1}(0)$ can be rearranged using equation (34) and the cyclic condition, equation (15). For example, if $t_1 \leqslant 0$,

$$\mathcal{V}_{i\alpha\beta} G_{\alpha\lambda}(\tau) C_{\beta\mu}(\tau) \mathcal{M}_{\lambda\mu\nu} C_{\nu j}(t_1) = -\tfrac{1}{2} \mathcal{V}_{i\alpha\beta} C_{\alpha\lambda}(\tau) C_{\beta\mu}(\tau) \mathcal{M}_{\nu\lambda\mu} G^{\dagger}_{\nu j}(t_1), \tag{36}$$

where $\tau = t - t_1$. Here we have used the fact that $C_{ij}(0) = -k_{\mathrm{B}} \Omega^{-1}_{ij}$. On the other hand, the terms containing $\mathcal{L}_{ijk}$ in $\mathbf{E} \otimes \mathbf{C} \cdot \mathbf{C}^{-1}(0)$ are rearranged as

$$\begin{aligned} &\left\{ G_{\alpha\lambda}(\tau) C_{\beta\mu}(\tau) \mathcal{L}_{\lambda\mu k} + k_{\mathrm{B}} G_{\alpha\lambda}(\tau) G_{\beta\mu}(\tau) L^{(1)}_{\lambda\mu,k} \right\} C_{kj}(t_1) \\ &\qquad = \left\{ -2k_{\mathrm{B}} G_{\alpha\lambda}(\tau) C_{\beta\mu}(\tau) L^{(1)}_{k\lambda,\mu} - \frac{1}{2} C_{\alpha\lambda}(\tau) C_{\beta\mu}(\tau) \mathcal{L}_{k\lambda\mu} \right\} G^{\dagger}_{kj}(t_1). \end{aligned} \tag{37}$$

Equation (36) combined with equation (37) cancels with $\mathbf{D} \otimes \mathbf{G}^{\dagger} \cdot \mathbf{C}^{-1}(0)$ of equation (35). Likewise, for $t_1 \geqslant 0$, $\mathbf{E} \otimes \mathbf{C} \cdot \mathbf{C}^{-1}(0)$ can be rewritten as $\mathbf{E} \otimes \mathbf{G}^{\dagger}$. Therefore, equation (35) becomes equivalent to the equation for $\mathbf{G}$, equation (29). This is the end of the proof.

Now let us prove the FDT. By taking the derivative of equation (34) with respect to time and using equation for $\mathbf{G}(t)$, equation (29), we have

$$\frac{\mathrm{d}\mathbf{C}(t)}{\mathrm{d}t} = -k_{\mathrm{B}} \mathbf{G}(t) \cdot \mathbf{K}^{(0)} + \int_0^t \mathrm{d}t_1 \mathbf{G}(t - t_1) \cdot \mathbf{E}(t_1) \cdot \mathbf{C}(0). \tag{38}$$

In this expression, $\mathbf{E}(t) \cdot \mathbf{C}(0)$ can be rewritten using equation (34) and antisymmetric property of $M^{(1)}_{ij,k}$ as

$$\{\mathbf{E}(t) \cdot \mathbf{C}(0)\}_{ij} = -k_{\mathrm{B}} \mathcal{V}_{i\alpha\beta} G_{\alpha\lambda}(t) C_{\beta\mu}(t) K^{(1)}_{\lambda j,\mu}. \tag{39}$$

Therefore, the right-hand side of equation (38) becomes identical to $-k_{\mathrm{B}} T \chi_{ij}(t)$ given by equation (33). Thus we arrive at equation (9) and the FDT is proved.

Finally let us derive the closed equation for $C_{ij}(t)$. $\mathbf{D} \otimes \mathbf{G}^{\dagger}$ again cancels with $\mathbf{E} \otimes \mathbf{C}$ for $t_1 \leqslant 0$. For $t_1 \geqslant 0$, $E_{ij}(t)$ is rewritten as

$$E_{ij}(t) = -\tfrac{1}{2} \mathcal{V}_{i\alpha\beta} C_{\alpha\lambda}(t) C_{\beta\mu}(t) (\mathcal{V} - 2\mathcal{L})_{\nu\lambda\mu} C^{-1}_{\nu j}(0) \tag{40}$$

and we arrive at the MCT equation for $C_{ij}(t)$;

$$\frac{\mathrm{d}C_{ij}(t)}{\mathrm{d}t} = \mu_{i\alpha} C_{\alpha j}(t) + \int_0^t \mathrm{d}t_1 E_{i\alpha}(t - t_1) C_{\alpha j}(t_1). \tag{41}$$

It is important to realize that $-2\mathcal{L}$ in the vertex in equation (40) is due to the multiplicative noise and the presence of it is essential. For example, for the pure dissipative case ($M_{ij}(\mathbf{x}) = 0$), neglect of the multiplicative noise leads to the wrong sign in front of the integral term (and thus leads to pathological behaviour). This term is neglected in [14].

5. FDT and MCT for class III

In this section, we shall consider the category of problems we call class III and elucidate generic reason why the MCT approximation for the class III dynamics is inconsistent with the FDT. For complete discussion of the technical aspects involved it would require a longer discussion than we provide here. Our main point is simply to sketch the difficulties that arise in attempting to formulate a simple MCT (namely a self-consistent one-loop theory for both the propagation and the response function) that satisfies the FDT. The important conclusion is that the standard idealized MCT of Götze and coworkers [1, 2] cannot be consistently derived via field-theoretic techniques, at least via the usual one-loop approximations applied to equation (1). This is discussed both in this section and in section 6.

For problems of class III, the entropy is not a quadratic function but has higher order terms;

$$S = S_0 + \frac{1}{2}\Omega_{\alpha\beta}\delta x_\alpha \delta x_\beta + \frac{1}{3!}\Lambda_{\alpha\beta\gamma}\delta x_\alpha \delta x_\beta \delta x_\gamma + \cdots . \tag{42}$$

We again assume that the kinetic coefficient $K_{ij}(\mathbf{x})$ is a linear function of $\mathbf{x}$. Up to the quadratic order in $\mathbf{x}$, the nonlinear Langevin equation for $\mathbf{x}$ is given by equation (13) but the vertex, equation (14), is now modified as

$$\mathcal{V}_{ijk} = K^{(0)}_{i\alpha}\Lambda_{\alpha jk} + M^{(1)}_{i\alpha,j}\Omega_{\alpha k} + M^{(1)}_{i\alpha,k}\Omega_{\alpha j} + L^{(1)}_{i\alpha,j}\Omega_{\alpha k} + L^{(1)}_{i\alpha,k}\Omega_{\alpha j} \equiv \mathcal{V}^{(I)}_{ijk} + \mathcal{V}^{(II)}_{ijk}, \tag{43}$$

where $\mathcal{V}^{(I)}_{ijk} \equiv K^{(0)}_{i\alpha}\Lambda_{\alpha jk}$ is the vertex that originates from the nonlinearity of the entropy whereas $\mathcal{V}^{(II)}_{ijk}$, which is the same as equation (14), originates from the $\mathbf{x}$-dependence of the kinetic coefficient $K_{ij}(\mathbf{x})$. The one-loop equations for $C_{ij}(t)$ and $G_{ij}(t)$, equations (28) and (29), remain the same except that the vertex is given by equation (43) instead of equation (14). First, we shall show that there is no simple relation such as equation (34) which relates the correlation function to the propagator. The starting point is the formal solution of equation (28) for the correlation function;

$$\mathbf{C} = \mathbf{G} \otimes (k_{\mathrm{B}}\mathbf{K}^{(0)} + k_{\mathrm{B}}\mathbf{K}^{(0)\dagger} + \mathbf{D}) \otimes \mathbf{G}^{\dagger}. \tag{44}$$

This is derived by eliminating $\mathbf{E}(t)$ from the formal solution of $\mathbf{C}(t)$ by substituting the formal solution for $\mathbf{G}(t)$. Substituting the equation for $\mathbf{G}$, equation (29), equation (44) can be rewritten as

$$\mathbf{C} = -k_{\mathrm{B}}\mathbf{G}\cdot\boldsymbol{\Omega}^{-1} + \mathbf{G}\otimes\mathbf{f}\otimes\mathbf{G}^{\dagger} \tag{45}$$

for $t \geqslant 0$. In this expression, $\mathbf{f}(t) \equiv -k_{\mathrm{B}}\mathbf{E}(t)\cdot\boldsymbol{\Omega}^{-1} - k_{\mathrm{B}}\boldsymbol{\Omega}^{-1}\cdot\mathbf{E}(t) + \mathbf{D}(t)$. Following the similar steps as equations (37)–(39), $\mathbf{f}(t)$ can be reduced as

$$f_{ij}(t) = -\mathcal{V}_{i\alpha\beta}G_{\alpha\lambda}(t)C_{\beta\mu}(t)\mathcal{V}^{(I)}_{\lambda\mu\nu}\Omega^{-1}_{\nu j} + \tfrac{1}{2}\mathcal{V}_{i\alpha\beta}C_{\alpha\lambda}(t)C_{\beta\mu}(t)\mathcal{V}^{(I)}_{j\lambda\mu} + (\text{higher order loops}), \tag{46}$$

where only the first two terms (one loop) are explicitly shown. The other terms consist of the higher order loops. These loops appear to contain at least one multiple of $\mathcal{V}^{(I)}\mathcal{V}^{(II)}$. This means that these higher order loops do not appear in either class I or class II problems. If one takes the time derivative of equation (45), one obtains, after straightforward but tedious calculations, the following expression:

$$\frac{\mathrm{d}\mathbf{C}(t)}{\mathrm{d}t} = -k_{\mathrm{B}}T\boldsymbol{\chi}(t) + (\text{higher order loops}). \tag{47}$$

Again, the higher order loops always contain at least one multiple of $\mathcal{V}^{(I)}\mathcal{V}^{(II)}$. One of the lowest order terms is an integral such as

$$\int \mathrm{d}t'\ \mathcal{V}_{i\alpha\beta}G_{\alpha\lambda}(t-t')C_{\beta\mu}(t-t')\mathcal{V}^{(I)}_{l\lambda\mu}\mathcal{V}^{(II)}_{l\alpha'\beta'}G^{\dagger}_{\alpha'\lambda'}(t')C_{\beta'\mu'}(t')\mathcal{V}_{j\lambda'\mu'}. \tag{48}$$

Note that this term is an irreducible loop in the field-theoretic language, which means that this cannot be represented by any simpler renormalized diagram. Since the original Langevin equation (13) itself *does* satisfy the FDT, the failure of the FDT in equation (47) is attributed to the inconsistencies of the loop expansion with the FDT. In other words, a naive loop expansion using the bare fields $\mathbf{z} = (\mathbf{x}, \hat{\mathbf{x}})$ for class III problems does not preserve the FDT at the each level of expansion. The higher order diagrams shown in equation (47) are cancelled only by taking the next higher order loops in the MSR loop expansion in section 3.

The failure to derive the FDT at the one-loop level for class III leads to the failure of deriving a MCT-type equation such as equation (41). By substituting equations (45) and (47) into the equation for $C_{ij}(t)$, equation (28), we obtain

$$\frac{\mathrm{d}C_{ij}(t)}{\mathrm{d}t} = \mu_{i\alpha}C_{\alpha j}(t) + \int_0^t \mathrm{d}t_1 E_{i\alpha}(t-t_1)C_{\alpha j}(t_1) + (\text{higher order loops}), \tag{49}$$

where $E_{ij}(t)$ is given by the same expression as equation (40) except that $\mathcal{V}$ is given by equation (43) and $\mathcal{L}$ in the second vertex, $(\mathcal{V}-2\mathcal{L})$ in equation (40) is replaced by

$$L^{(0)}_{i\alpha}\Lambda_{\alpha jk} + L^{(1)}_{i\alpha,j}\Omega_{\alpha k} + L^{(1)}_{i\alpha,k}\Omega_{\alpha j}. \tag{50}$$

The higher order loops in equation (49) are again irreducible diagrams which do not appear if either $\mathcal{V}^{(I)}$ or $\mathcal{V}^{(II)}$ is absent. Equation (49) shows that the standard MCT equation for the class III is regarded as the *uncontrollable* approximation in the field-theoretic sense in that it neglects terms which are present in the original set of equations, equations (28) and (29). The consequences of this are discussed below.

6. Discussion

In order to see how the results shown in the previous sections are related to real fluids, let us consider the Langevin equation for a colloidal suspension given by equation (1). The class II equation is derived by approximating the entropy given by equation (4) with its Gaussian form. Neglecting the terms of higher order than quadratic, one has

$$S \simeq S_0 - \frac{k_{\mathrm{B}}}{2}\int \mathrm{d}\mathbf{k}\frac{|\delta\rho_{\mathbf{k}}|^2}{NS(k)}, \tag{51}$$

is the Fourier transform of $\delta\rho(\mathbf{r})$, N is the total number of the particles and $S(k) = N^{-1}\langle|\delta\rho_{\mathbf{k}}|^2\rangle$ is the static structure factor. As mentioned in section 3, this approximation has been shown to hold over a wide range of length scales in real liquids [30, 31]. It is also compatible with the functionals used to derive integral equations of fluid structure [32]. It is not expected to hold on very short length scales where density fluctuations are Poissonian and the ideal gas entropy is essential. On the other hand, density fluctuations on such length scales are not expected physically to be effective in providing glassy behaviour. It is plausible that the approximation equation (51) may be used (along with an appropriate large wavevector cutoff) in the treatment of realistic fluids. Indeed, approximation (51) is used in [13]. Using equations (3) and (51), the MCT equation, equation (41), for the density correlation function $F(k,t) = N^{-1}\langle\delta\rho_{\mathbf{k}}(t)\delta\rho_{-\mathbf{k}}(0)\rangle$ is written as

$$\frac{\partial F(k,t)}{\partial t} = -\frac{Dk^2}{S(k)}F(k,t) + \int_0^t \mathrm{d}t_1 M'(k,t-t_1)F(k,t_1) \tag{52}$$

with the memory kernel given by

$$M'(k,t) = \frac{D^2k^2}{2\rho_0 S(k)} \int \frac{\mathrm{d}\mathbf{q}}{(2\pi)^3} \left\{ \frac{\hat{\mathbf{k}}\cdot\mathbf{q}}{S(q)} + \frac{\hat{\mathbf{k}}\cdot\mathbf{p}}{S(p)} \right\}^2 F(q,t)F(p,t), \tag{53}$$

where $\mathbf{p} = \mathbf{k} - \mathbf{q}$. Note that the vertex $\hat{\mathbf{k}}\cdot\mathbf{q}/S(q) + \hat{\mathbf{k}}\cdot\mathbf{p}/S(p)$ is precisely the one that appears in [13], where the Gaussian approximation to the entropy is also made. This equation should be compared with the standard MCT equation which has been derived using the projection operator method with the decoupling approximation [18] where[3]

$$M(k,t) = \frac{\rho_0 D^2 k^2}{2} \int \frac{\mathrm{d}\mathbf{q}}{(2\pi)^3} \{\hat{\mathbf{k}}\cdot\mathbf{q}c(q) + \hat{\mathbf{k}}\cdot\mathbf{p}c(p)\}^2 F(q,t)F(p,t). \tag{54}$$

We observe that there is a difference between equations (53) and (54): $1/S(q)$ appears in the vertex function of equation (53), whereas the direct correlation function $\rho_0 c(q) = 1 - 1/S(q)$ shows up in equation (54). This difference traces back to the Gaussian approximation, equation (51) [34]. The entropy of fluids is not Gaussian due primarily to ideal gas part, $\rho \ln \rho$, in equation (4). As discussed in the introduction, the nonlinearities of realistic fluids come both from the entropy (in this case, $\rho \ln \rho$) and the kinetic coefficient and therefore realistic fluids are destined to belong to class III over the entire range of wavevectors. Indeed, if the full expression for S, equation (4), instead of approximated form of equation (51) is used, one sees that the non-quadratic term of S gives a vertex of the form of $-D(\hat{\mathbf{k}}\cdot\mathbf{q} + \hat{\mathbf{k}}\cdot\mathbf{p})$ which, combined with $D\{\hat{\mathbf{k}}\cdot\mathbf{q}/S(q) + \hat{\mathbf{k}}\cdot\mathbf{p}/S(p)\}$, leads to $\rho_0 D\{\hat{\mathbf{k}}\cdot\mathbf{q}c(q) + \hat{\mathbf{k}}\cdot\mathbf{p}c(p)\}$. Using the full expression for S means that the dynamics now belongs to class III. As illustrated in section 5, one obtains the memory kernel in the form of equation (54) but there are always extra terms which are a direct consequence of inconsistencies of the FDT with the one-loop approximation in the MSR formalism, at least if the loop expansion is made directly with physical density modes as field variables[4]. This argument is true for arbitrary orders of the loop expansion. This conclusion implies that there is no simple systematic way to derive equation (54) from equation (1) using the standard field-theoretic method. This inconsistency is not relevant as far as one is concerned only with the equilibrium state, because one may always adopt an approximation where one neglects higher order terms in equation (49) and 'define' the response function via the FDT instead of solving the equation for $\mathbf{G}(t)$, equation (29), separately. But we cannot do so if the system is in nonequilibrium state. It is desirable to develop such an expansion method that preserves the FDT relation at each level of the perturbative expansion.

Even with the difficulties discussed in this letter, the MSR or field-theoretic approach is still an attractive route to attack out-of-equilibrium supercooled fluids, in that it is systematic and one does not need to evaluate the nonequilibrium measure which is required in alternative approaches such as the projection operator technique. Models which belong to class I have been already discussed extensively in the context of spin glasses and even for supercooled fluids [35]. However, it is difficult to construct realistic models of class I which can incorporate

[3] Note that equation (52) is different from the MCT equation used in the supercooled fluids; $-Dk^2F(k,t_1)$ appears in the place of $\partial F(k,t_1)/\partial t_1$ [18, 20, 33]. The difference originates from the overdamped nature of the starting diffusion equation, equation (1). One obtains the $\partial F(k,t_1)/\partial t_1$ term if one incorporates the momentum density as well as the number density as stochastic variables. Technically, this is equivalent to using the irreducible projection operator introduced by Cichocki *et al* [18].

[4] Recent unpublished work by G Biroli, A Lefevre and J-P Bouchaud shows that if a transformed set of modes is used, FDT can be recovered for the full class III problem, although, at the time of submission of this work, other mathematical difficulties arise that render the resulting equations unusable in the present form. In particular, the vertex that results is distinct from that of equation (54), and thus the one-loop approximation does not yield the usual form of the standard idealized MCT of Götze and coworkers.

the effect of changes of structure embodied in $S(k)$ observed in simulations of aging [7] and sheared systems [36, 37], which is argued to be essential for the violation of the FDT of real fluids [38]. The class II system derived here is a better candidate as a realistic 'model fluid'. As discussed above, the Gaussian approximation for the entropy is known to be a good description for wide ranges of densities and length scales [30, 31]. The equation for equal-time correlation functions such as $S(k)$ should be constructed in the same manner as the MCT equation derived here. The solution could be plugged into the vertex functions of the set of the MCT equations, equations (28) and (29). Eventually, these three equations can be solved self-consistently. Performing such a calculation will require some consideration of a wavevector cutoff that would eliminate spurious divergence that arises from the *approximate* vertex functions (equation (53)). Calculations in this direction are underway.

In summary, in the present work, we have broadened the range of applicability of the MSR approach by extending the method to dynamical processes with the multiplicative noise. This is a necessary step in the treatment of the Brownian dynamics of colloidal suspensions. This formalism still does not cover real fluids over all length scales and it is found that there is no direct compatibility between the MCT equations derived from MSR approach and from the projection operator method. We proposed an approximate but feasible method to explore nonequilibrium supercooled fluids using the formalism discussed in this paper. The MSR method is not restricted to evaluation of the two point correlation functions nor to the lowest order loop expansion. Extension of the method to the multipoint correlation functions and to higher order loops will be essential for understanding growing length scales which are hidden in the supercooled fluids [3]. The formulation presented in this paper will serve as a first step towards such extensions.

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