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FAST TRACK COMMUNICATION

The mode coupling theory in the FDR-preserving field theory of interacting Brownian particles

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Abstract
We develop a renormalized perturbation theory for the dynamics of interacting Brownian particles, which preserves the fluctuation–dissipation relation order by order. We then show that the resulting one-loop theory gives a closed equation for the density correlation function, which is identical with that in the standard mode coupling theory.

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As a first-principle approach, the mode coupling theory (MCT) [1–3] has not only enjoyed considerable success in explaining the slowing down of the weakly supercooled liquids, but also had enormous impact on the area by stimulating further experiments, simulations and other theoretical developments. However, the foundation of the theory leaves much to be desired since the theory is fraught with uncontrolled approximations. It is helpful if one can develop systematic field-theoretical treatment for a model system containing smallness parameter. The earlier field-theoretic formulations [4, 5] of MCT are found to be incompatible with the fluctuation–dissipation relation (FDR) [6, 7]. It was thus an urgent task to develop a consistent field-theoretic formulation capable of describing the dynamics of glass-forming liquids, for which a systematic perturbation expansion preserving the FDR is possible. Quite recently, Andreanov, Biroli and Lefevre (ABL) [7] provided a remarkable insight into this problem by focusing on the symmetry properties of the action integral under time reversal (TR). In particular, ABL has identified the transformation of fields under TR, which leaves the action invariant. The FDR naturally follows from the TR transformation combined with causality. Moreover, the nonlinear nature of the TR transformation is shown to be the underlying reason why the renormalized perturbation theory (i.e., the loop expansion) does not preserve the FDR order by order. By introducing a new set of auxiliary fields to linearize the time-reversal transformation, ABL have attempted to develop a FDR-preserving field theory. Although ABL's work is a remarkable step forward, the one-loop results of ABL's theory are found to have some pathological features. The equation for the nonergodicity parameter gives nontrivial results even for noninteracting Brownian systems, which is suspicious and should be examined carefully. Furthermore, their vertex is ill behaved, leading to the divergence of the memory integral. We tend to believe that these ill-behaved results are intimately connected to their linearization scheme.

In this communication, by proposing a simpler but crucial linearization scheme, we show that the one-loop result in the FDR-preserving renormalized perturbation theory yields a closed dynamic equation for the density correlation function and show that this closed equation turns out to be the same as that in the standard MCT. We thus have established the precise relationship of the FDR-preserving field theory with the standard MCT.

We start with the following Langevin equation for the density field $\rho(\mathbf{r}, t)$ of interacting Brownian particles:

$$\partial_t \rho(\mathbf{r}, t) = \nabla \cdot \left(\rho(\mathbf{r}, t) \nabla \frac{\delta F[\rho]}{\delta \rho(\mathbf{r}, t)} \right) + \eta(\mathbf{r}, t) \tag{1}$$

where the Gaussian thermal noise $\eta(\mathbf{r}, t)$ has zero mean and variance of the form

$$\langle \eta(\mathbf{r}, t)\eta(\mathbf{r}', t')\rangle = 2T\nabla \cdot \nabla' \big(\rho(\mathbf{r}, t)\delta(\mathbf{r} - \mathbf{r}')\delta(t - t')\big). \tag{2}$$

Note that the noise correlation depends on the density variable, i.e., the noise is multiplicative. In (1), $F[\rho]$ is the free-energy density functional which takes the following form:

$$F[\rho] = T \int \mathrm{d}\mathbf{r}\, \rho(\mathbf{r}) \left(\ln \frac{\rho(\mathbf{r})}{\rho_0} - 1 \right) + \frac{1}{2} \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}'\, \delta\rho(\mathbf{r}) U(\mathbf{r} - \mathbf{r}') \delta\rho(\mathbf{r}') \tag{3}$$

where $\delta\rho(\mathbf{r}, t) \equiv \rho(\mathbf{r}, t) - \rho_0$ is the density fluctuation around the equilibrium density ρ_0. In (3), the first term is the ideal-gas part of the free energy, $F_{\mathrm{id}}[\rho]$, and the second term the interaction part of the free energy, $F_{\mathrm{int}}[\rho]$. Using Ito calculus, Dean [8] has derived the above nonlinear Langevin equation for the density field of system of interacting Brownian particles with pair potential $U(\mathbf{r})$. Earlier, Kawasaki [9] has obtained the same Langevin equation with $U(\mathbf{r})$ replaced by $-Tc(\mathbf{r})$, with $c(\mathbf{r})$ being the direct correlation function, by adiabatically eliminating the momentum field in the fluctuating nonlinear hydrodynamic equations [4] of the glass-forming liquids. For this case, the free-energy density functional (3) takes the Ramakrishnan–Yussouff form.

We consider the corresponding action integral $\mathcal{S}[\rho, \hat{\rho}]$ which governs the stochastic dynamics of the coarse-grained density variable:

$$\mathcal{S}[\rho, \hat{\rho}] = \int \mathrm{d}\mathbf{r} \int \mathrm{d}t \left\{ \hat{\rho} \left[-\partial_t \rho + \nabla \cdot \left(\rho \nabla \frac{\delta F}{\delta \rho} \right) \right] + T\rho(\nabla\hat{\rho})^2 \right\} \tag{4}$$

where the field $\hat{\rho}$ is pure imaginary and the last term comes from the multiplicative thermal noise. (A similar action was given in [5] but with real $\hat{\rho}$.) It is a crucial observation of ABL to recognize that the above action is *invariant* under the TR field transformation:

$$\rho(\mathbf{r}, -t) = \rho(\mathbf{r}, t) \qquad \hat{\rho}(\mathbf{r}, -t) = -\hat{\rho}(\mathbf{r}, t) + \frac{1}{T} \frac{\delta F}{\delta \rho(\mathbf{r}, t)}. \tag{5}$$

The FDR follows from the above TR transformation. The response function $R(\mathbf{r}, t; \mathbf{r}'t')$ is defined as a link between induced density change $\Delta\langle\rho(\mathbf{r}, t)\rangle$ and an external infinitesimal field $h_e(\mathbf{r}', t')$:

$$\Delta\langle\rho(\mathbf{r}, t)\rangle \equiv \int \mathrm{d}\mathbf{r}' \int \mathrm{d}t'\, R(\mathbf{r}, t; \mathbf{r}'t') h_e(\mathbf{r}', t') \tag{6}$$

where $\langle\cdots\rangle \equiv \int \mathcal{D}\rho \int \mathcal{D}\hat{\rho} (\cdots) \exp(\mathcal{S}[\rho, \hat{\rho}])$. To find the induced change of density, one should add the contribution of the external field to the free energy $F, \Delta F \equiv -\int \mathrm{d}\mathbf{r} \int \mathrm{d}t\, \delta\rho(\mathbf{r}, t) h_e(\mathbf{r}, t)$. It is straightforward to show, by obtaining the induced density change, that the response function $R(\mathbf{r}, t; \mathbf{r}', t')$ is given by

$$R(\mathbf{r}, t; \mathbf{r}', t') = -\langle \rho(\mathbf{r}, t)\nabla' \cdot (\rho(\mathbf{r}', t')\nabla'\hat{\rho}(\mathbf{r}', t'))\rangle. \quad (7)$$

Note that the response function is *not* given by the conventional response function $-\langle \rho(\mathbf{r}, t)\nabla'^2\hat{\rho}(\mathbf{r}', t')\rangle$ which is the true response function for the Langevin equation with additive noise. Instead, the response function is given by (7) due to the multiplicative nature of the Langevin equation noise for the density fluctuation [6, 7]. Using the identity $\langle \rho(\mathbf{r}, t)\delta S/\delta\hat{\rho}(\mathbf{r}', t')\rangle = 0$ and the TR transformation (5), one obtains the FDR

$$-\frac{1}{T}\partial_t G_{\rho\rho}(\mathbf{r} - \mathbf{r}', t - t') = -R(\mathbf{r} - \mathbf{r}', t' - t) + R(\mathbf{r} - \mathbf{r}', t - t') \quad (8)$$

where $G_{\rho\rho}(\mathbf{r} - \mathbf{r}', t - t') \equiv \langle \delta\rho(\mathbf{r}, t)\delta\rho(\mathbf{r}', t')\rangle$. Since $R(\mathbf{r} - \mathbf{r}', t' - t) = 0$ for $t > t'$ due to causality, equation (8) gives the standard form of the FDR:

$$R(\mathbf{r} - \mathbf{r}', t - t') = -\Theta(t - t')\frac{1}{T}\partial_t G_{\rho\rho}(\mathbf{r} - \mathbf{r}', t - t') \quad (9)$$

where $\Theta(t)$ is the Heaviside step function.

Using the form of the free energy given in (3), one can explicitly write $\delta F/\delta\rho$ as

$$\begin{aligned} \frac{1}{T}\frac{\delta F_{\mathrm{id}}[\rho]}{\delta\rho(\mathbf{r}, t)} &= \ln\frac{\rho(\mathbf{r}, t)}{\rho_0} \equiv \frac{\delta\rho(\mathbf{r}, t)}{\rho_0} + f(\delta\rho(\mathbf{r}, t)), \\ \frac{1}{T}\frac{\delta F_{\mathrm{int}}[\rho]}{\delta\rho(\mathbf{r}, t)} &= \frac{1}{T}\int \mathrm{d}\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}')\delta\rho(\mathbf{r}', t) \end{aligned} \quad (10)$$

where $f[\delta\rho(\mathbf{r}, t)]$ is the contribution of the non-Gaussian part of $F_{\mathrm{id}}[\rho]$ and is given by an infinite series $f[\delta\rho(\mathbf{r}, t)] \equiv -\sum_{n=2}^{\infty}\frac{1}{n}(-\delta\rho(\mathbf{r}, t)/\rho_0)^n$. Using (10), one can explicitly write the TR transformation (5) as

$$\begin{aligned} \rho(\mathbf{r}, -t) &= \rho(\mathbf{r}, t) \\ \hat{\rho}(\mathbf{r}, -t) &= -\hat{\rho}(\mathbf{r}, t) + f[\delta\rho(\mathbf{r}, t)] + \hat{K} * \delta\rho(\mathbf{r}, t) \end{aligned} \quad (11)$$

where $\hat{K}*$ is convolution with the kernel $K(\mathbf{r}) \equiv (\delta(\mathbf{r})/\rho_0 + U(\mathbf{r})/T)$. Note that equation (11) is *nonlinear* due to the non-Gaussian contribution $f[\delta\rho(\mathbf{r}, t)]$ to the ideal-gas part of the free energy[3]. ABL has shown that this nonlinear nature of the transformation is the origin of the incompatibility of the renormalized perturbation theory with FDR. Naturally, this incompatibility would be resolved if the transformation (11) is made linear by ignoring $f[\delta\rho(\mathbf{r}, t)]$. However, as shown below, in this Gaussianized case, the nonlinear term generated by dynamics (the second term in (13)) would give rise to the spurious contribution to the one-loop result, incorrectly yielding a nontrivial result [6, 7, 10] even in the noninteracting system.

As the most natural way to make the transformation (11) linear, we introduce a new field $\theta(\mathbf{r}, t)$ so as to satisfy the nonlinear constraint

$$\theta(\mathbf{r}, t) = f[\delta\rho(\mathbf{r}, t)] \equiv -\sum_{n=2}^{\infty}\frac{1}{n}\left(-\frac{\delta\rho}{\rho_0}\right)^n. \quad (12)$$

Note that our approach here differs from that of ABL in that whereas ABL defines the new variable as the functional derivative of the full free energy with respect to density: $\theta_{\mathrm{ABL}}(\mathbf{r}, t) \equiv \delta F/\delta\rho(\mathbf{r}, t)$, we limit the new variable $\theta(\mathbf{r}, t)$ only to the nonlinear part of it.

[3] Throughout the paper the term 'ideal gas' is meant only for static behaviour. Dynamics for our system is of course different from the ideal-gas behaviour even without interparticle interactions.

Using the constraint (12), we obtain the ideal-gas contribution to the body force as

$$\nabla\cdot\left(\rho\nabla\frac{\delta F_{\mathrm{id}}}{\delta\rho}\right)=T\nabla^2\rho+\frac{T}{\rho_0}\nabla\cdot(\delta\rho\nabla\rho)+\rho_0T\nabla^2\theta+T\nabla\cdot(\delta\rho\nabla\theta) \tag{13}$$

where the first (the last) two terms are the contributions from the Gaussian (non-Gaussian) parts of the ideal-gas free energy. Since due to cancellation of the two nonlinear effects, the entire ideal-gas contribution to the dynamics is pure diffusion, i.e., $\nabla\cdot(\rho\nabla\delta F_{\mathrm{id}}/\delta\rho)=T\nabla^2\rho$, the sum of the last three terms in (13) should *vanish if* the constraint (12) is taken into account:

$$\frac{T}{\rho_0}\nabla\cdot(\delta\rho\nabla\rho)+\rho_0T\nabla^2\theta+T\nabla\cdot(\delta\rho\nabla\theta)=0. \tag{14}$$

As shown below, this nonperturbative cancellation plays a crucial role in obtaining a closed equation for the density correlation function alone.

Incorporating the new variable $\theta(\mathbf{r},t)$ and its conjugate $\hat{\theta}(\mathbf{r},t)$, the action (4) can now be explicitly rewritten as

$$\begin{aligned}
\mathcal{S}[\psi]&\equiv\mathcal{S}_g[\psi]+\mathcal{S}_{\mathrm{ng}}[\psi]\\
\mathcal{S}_g[\psi]&\equiv\int\mathrm{d}\mathbf{r}\int\mathrm{d}t\left\{\hat{\rho}\left[-\partial_t\rho+T\nabla^2\rho+\underline{\rho_0T\nabla^2\theta}+\rho_0\nabla^2\hat{U}*\delta\rho\right]+T\rho_0(\nabla\hat{\rho})^2+\hat{\theta}\theta\right\}\\
\mathcal{S}_{\mathrm{ng}}[\psi]&\equiv\int\mathrm{d}\mathbf{r}\int\mathrm{d}t\left\{\hat{\rho}\left[\nabla\cdot(\delta\rho\nabla\hat{U}*\delta\rho)+\underline{\frac{T}{\rho_0}\nabla\cdot(\delta\rho\nabla\rho)}+\underline{T\nabla\cdot(\delta\rho\nabla\theta)}\right]\right.\\
&\qquad\left.+T\delta\rho(\nabla\hat{\rho})^2-\hat{\theta}f(\delta\rho)\right\}
\end{aligned} \tag{15}$$

where ψ denotes the entire set of the fields collectively, and the full action $\mathcal{S}[\psi]$ is separated into its Gaussian part $\mathcal{S}_g[\psi]$ and non-Gaussian part $\mathcal{S}_{\mathrm{ng}}[\psi]$.[4] Now the actions $\mathcal{S}_g[\psi]$ and $\mathcal{S}_{\mathrm{ng}}[\psi]$ are *separately* invariant under the following *linear* TR transformation:

$$\begin{aligned}
\rho(\mathbf{r},-t)&=\rho(\mathbf{r},t)\\
\hat{\rho}(\mathbf{r},-t)&=-\hat{\rho}(\mathbf{r},t)+\theta(\mathbf{r},t)+\hat{K}*\delta\rho(\mathbf{r},t)\\
\theta(\mathbf{r},-t)&=\theta(\mathbf{r},t)\\
\hat{\theta}(\mathbf{r},-t)&=\hat{\theta}(\mathbf{r},t)-\partial_t\rho(\mathbf{r},t).
\end{aligned} \tag{16}$$

It is easy to show that the modulus of the associated transformation matrix is unity. Though the three underlined terms in (15) vanish when summed together, their presence is crucial for the actions $\mathcal{S}_g[\psi]$ and $\mathcal{S}_{\mathrm{ng}}[\psi]$ to be separately time-reversal invariant. Also note that the separate invariance of the actions under the linear transformation (16) is not tied to the form of the constraint (12). This separate invariance of $\mathcal{S}_g[\psi]$ and $\mathcal{S}_{\mathrm{ng}}[\psi]$ enables us to construct the FDR-preserving renormalized perturbation theory from these actions.

With the new action (15), it is easy to show that the response function is given by

$$R(\mathbf{r},t;\mathbf{r}',t')=\frac{1}{T}\langle\delta\rho(\mathbf{r},t)\hat{\theta}(\mathbf{r}',t')\rangle. \tag{17}$$

One can then obtain the FDR (8) by taking correlation of the last member of (16) with $\delta\rho(\mathbf{r},t)/T$.

Incorporating the above new set of variables into the theory, we are now ready to develop a renormalized perturbation theory which preserves the FDR order by order. We

[4] We recall that separation into Gaussian and non-Gaussian parts do not coincide for the action $\mathcal{S}$ and for the free energy functional F.

first consider the noninteracting case ($U = 0$) and show that the noninteracting part of the action, $\mathcal{S}_{\mathrm{id}}[\psi] \equiv \mathcal{S}[\psi; U = 0]$, yields the dynamic behaviour consistent for the noninteracting system. We begin with the following identities:

$$\left\langle \delta\rho(\mathbf{2}) \frac{\delta \mathcal{S}_{\mathrm{id}}[\psi]}{\delta\hat{\rho}(\mathbf{1})} \right\rangle = 0, \qquad \left\langle \delta\rho(\mathbf{2}) \frac{\delta \mathcal{S}_{\mathrm{id}}[\psi]}{\delta\theta(\mathbf{1})} \right\rangle = 0 \tag{18}$$

where $\mathbf{1} \equiv (\mathbf{r}, t)$ and $\mathbf{2} \equiv (\mathbf{0}, 0)$. The first identity can be explicitly written as

$$0 = \left\langle \delta\rho(\mathbf{2}) \frac{\delta \mathcal{S}_{\mathrm{id}}[\psi]}{\delta\hat{\rho}(\mathbf{1})} \right\rangle = \left(-\frac{\partial}{\partial t} + T\nabla^2 \right) G_{\rho\rho}(\mathbf{1}-\mathbf{2}) - 2T\rho_0 \nabla^2 \langle \hat{\rho}(\mathbf{1})\delta\rho(\mathbf{2})\rangle \\ - 2T\langle \delta\rho(\mathbf{2})\nabla\cdot(\delta\rho(\mathbf{1})\nabla\hat{\rho}(\mathbf{1}))\rangle \tag{19}$$

where we used the fact that the sum of the three underlined terms in (15) vanishes. Similarly, using the second identity in (18), we obtain

$$0 = \left\langle \delta\rho(\mathbf{2}) \frac{\delta \mathcal{S}_{\mathrm{id}}[\psi]}{\delta\theta(\mathbf{1})} \right\rangle = \rho_0 T\nabla^2 \langle \hat{\rho}(\mathbf{1})\delta\rho(\mathbf{2})\rangle + \langle \hat{\theta}(\mathbf{1})\delta\rho(\mathbf{2})\rangle + T\langle \delta\rho(\mathbf{2})\nabla\cdot(\delta\rho(\mathbf{1})\nabla\hat{\rho}(\mathbf{1}))\rangle. \tag{20}$$

In (20), $\langle \hat{\rho}(\mathbf{1})\delta\rho(\mathbf{2})\rangle = 0$ and $\langle \hat{\theta}(\mathbf{1})\delta\rho(\mathbf{2})\rangle = 0$ for $t > 0$ due to the causality, and hence we obtain

$$\langle \delta\rho(\mathbf{2})\nabla\cdot(\delta\rho(\mathbf{1})\nabla\hat{\rho}(\mathbf{1}))\rangle = 0 \qquad \text{for} \quad t > 0. \tag{21}$$

Using (21) and causality, we obtain from (19)

$$\frac{\partial}{\partial t} G_{\rho\rho}(\mathbf{r}, t) = T\nabla^2 G_{\rho\rho}(\mathbf{r}, t) \qquad \text{for} \quad t > 0. \tag{22}$$

This result is expected for the noninteracting system.

There are five nonlinear terms in the full action (15) where the two underlined nonlinear terms $(T/\rho_0)\hat{\rho}\nabla\cdot(\delta\rho\nabla\rho)$ and $T\hat{\rho}\nabla\cdot(\delta\rho\nabla\theta)$ are shown to cancel the linear diffusion term $\hat{\rho}\rho_0 T\nabla^2\theta$. Then, the remaining nonlinearities are $\hat{\rho}\nabla\cdot(\delta\rho\nabla\hat{U} * \delta\rho)$, $T\delta\rho(\nabla\hat{\rho})^2$ and $\hat{\theta} f[\delta\rho]$. The first two come from the particle interaction and the multiplicative thermal noise, respectively. The last cubic nonlinear term $\frac{1}{2}\hat{\theta}(\delta\rho/\rho_0)^2$, the only one contributing in the one-loop order, comes from the non-Gaussian part of the ideal-gas free energy. In order to analyse the effect of these three nonlinear terms, one should examine the structures of the relevant self-energies.

Let us write

$$\mathcal{S}_g[\psi] = \frac{1}{2}\psi^T(\mathbf{1})\cdot G_0^{-1}(\mathbf{12})\cdot\psi(\mathbf{2}) \\ \mathcal{S}_{\mathrm{ng}}[\psi] = \frac{1}{3!}V(123)\psi(1)\psi(2)\psi(3) + \hat{\theta}(\mathbf{1})\sum_{n=3}^{\infty}\frac{1}{n}\left(-\frac{\delta\rho(\mathbf{1})}{\rho_0}\right)^n \tag{23}$$

where $\psi(\mathbf{1})$ and $\psi^T(\mathbf{1})$ are, respectively, column and row vectors of the four fields ρ, $\hat{\rho}$, θ and $\hat{\theta}$, and the term with $n = 2$ in the summation is incorporated into $V(123)$. The unperturbed inverse matrix propagator $G_0^{-1}(\mathbf{12})$ can be read off from the action (15) as

$$G_0^{-1}(\mathbf{12}) = \begin{pmatrix} 0 & \tilde{D}_1\delta(\mathbf{12}) + \rho_0\nabla_1^2 U(\mathbf{12}) & 0 & 0 \\ D_1\delta(\mathbf{12}) + \rho_0\nabla_1^2 U(\mathbf{12}) & -2T\rho_0\nabla_1^2\delta(\mathbf{12}) & T\rho_0\nabla_1^2\delta(\mathbf{12}) & 0 \\ 0 & T\rho_0\nabla_1^2\delta(\mathbf{12}) & 0 & \delta(\mathbf{12}) \\ 0 & 0 & \delta(\mathbf{12}) & 0 \end{pmatrix} \tag{24}$$

where $D_1 \equiv \left(-\partial/\partial t_1 + T\nabla_1^2\right)$ and $\tilde{D}_1 \equiv \left(\partial/\partial t_1 + T\nabla_1^2\right)$. $V(123)$ in fact is a collection of vertices $V_{\alpha_1\alpha_2\alpha_3}(\mathbf{123})$ with α's standing for four field types, $\mathbf{1}, \mathbf{2}, \mathbf{3}, \ldots$, for spacetime coordinates, and $\psi(1)$, etc stand for $\psi_{\alpha_1}(\mathbf{1})$, etc. Repeated thin (thick) numbers indicate spacetime integrations and summations over the field types (spacetime integrations only). We give expressions for the vertices $V_{\alpha_1\alpha_2\alpha_3}(\mathbf{123})$:

$$\begin{aligned}
V^{\mathrm{id}}_{\hat\rho\rho\rho}(\mathbf{123}) &= -\frac{T}{\rho_0}[\nabla_1\delta(\mathbf{12})\cdot\nabla_3\delta(\mathbf{23}) + \nabla_1\delta(\mathbf{13})\cdot\nabla_2\delta(\mathbf{23})],\\
V^{\mathrm{int}}_{\hat\rho\rho\rho}(\mathbf{123}) &= -[\nabla_1\delta(\mathbf{12})\cdot\nabla_3\hat{U}(\mathbf{23}) + \nabla_1\delta(\mathbf{13})\cdot\nabla_2\hat{U}(\mathbf{23})]\delta(t_2 - t_3),\\
V_{\hat\rho\rho\theta}(\mathbf{123}) &= -T\nabla_1\cdot\nabla_3\delta(\mathbf{123}), \qquad V_{\rho\hat\rho\hat\rho}(\mathbf{123}) = T\nabla_2\delta(\mathbf{12})\cdot\nabla_3\delta(\mathbf{13}),\\
V_{\hat\theta\rho\rho}(\mathbf{123}) &= \frac{1}{2\rho_0^2}\delta(\mathbf{12})\delta(\mathbf{23}),
\end{aligned} \tag{25}$$

where we have separated the vertex $V_{\hat\rho\rho\rho}(\mathbf{123})$ into the ideal-gas contribution $V^{\mathrm{id}}_{\hat\rho\rho\rho}(\mathbf{123})$ and the interaction contribution $V^{\mathrm{int}}_{\hat\rho\rho\rho}(\mathbf{123})$. In (24) and (25), the spacetime delta function is defined as $\delta(\mathbf{12}) \equiv \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(t_1 - t_2)$.

The dynamic equations for the correlation and response functions are formally given by the matrix Schwinger–Dyson (SD) equation:

$$G_0^{-1}(\mathbf{13})\cdot G(\mathbf{32}) - \Sigma(\mathbf{13})\cdot G(\mathbf{32}) = \delta(\mathbf{12}). \tag{26}$$

In order to obtain the equation of motion for the density correlation function, we compute $\hat\rho\rho$-element of the SD equation by taking $\mathbf{1} \equiv (\mathbf{r}, t)$, $\mathbf{2} \equiv (\mathbf{0}, 0)$ and $\mathbf{3} \equiv (\mathbf{r}_s, s)$. It is now convenient to introduce the Fourier transform in space

$$\Sigma(\mathbf{r}, t) = \int_{\mathbf{k}} \Sigma(\mathbf{k}, t)\,\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}} \tag{27}$$

etc, where $\int_{\mathbf{k}} \equiv \int \mathrm{d}\mathbf{k}/(2\pi)^3$. Using (24) and causality of the response functions, we obtain for $t > 0$

$$\left[G_0^{-1}\cdot G\right]_{\hat\rho\rho}(\mathbf{k}, t) = -(\partial_t + \rho_0 T\mathbf{k}^2 K(\mathbf{k}))G_{\rho\rho}(\mathbf{k}, t) - \rho_0 T\mathbf{k}^2 G_{\theta\rho}(\mathbf{k}, t). \tag{28}$$

Likewise using the causalities of the self-energy functions and the response functions, we obtain for $t > 0$

$$\begin{aligned}
[\Sigma\cdot G]_{\hat\rho\rho}(\mathbf{k}, t) = &\int_{-\infty}^{t} \mathrm{d}s[\Sigma_{\hat\rho\rho}(\mathbf{k}, t-s)G_{\rho\rho}(\mathbf{k}, s) + \Sigma_{\hat\rho\theta}(\mathbf{k}, t-s)G_{\theta\rho}(\mathbf{k}, s)]\\
&+ \int_{-\infty}^{0} \mathrm{d}s[\Sigma_{\hat\rho\hat\rho}(\mathbf{k}, t-s)G_{\hat\rho\rho}(\mathbf{k}, s) + \Sigma_{\hat\rho\hat\theta}(\mathbf{k}, t-s)G_{\hat\theta\rho}(\mathbf{k}, s)]
\end{aligned} \tag{29}$$

where the upper limits of the time integration are due to the causality of the self-energy functions in the first two integrations and due to the causality of the response functions in the last two integrations. It can be shown that (29) can be simplified using the TR properties of the self-energies and the response functions as

$$\begin{aligned}
[\Sigma\cdot G]_{\hat\rho\rho}(\mathbf{k}, t) = &-\int_0^t \mathrm{d}s[\Sigma_{\hat\rho\hat\theta}(\mathbf{k}, t-s)\partial_s G_{\rho\rho}(\mathbf{k}, s) + K(\mathbf{k})\Sigma_{\hat\rho\hat\rho}(\mathbf{k}, t-s)G_{\rho\rho}(\mathbf{k}, s)]\\
&- \int_0^t \mathrm{d}s\;\Sigma_{\hat\rho\hat\rho}(\mathbf{k}, t-s)G_{\theta\rho}(\mathbf{k}, s).
\end{aligned} \tag{30}$$

Using (28) and (30), the $\hat{\rho}\rho$-element of the SD equation for $t > 0$ can now be explicitly written as

$$\partial_t G_{\rho\rho}(\mathbf{k},t) = -\rho_0 T\mathbf{k}^2 K(\mathbf{k}) G_{\rho\rho}(\mathbf{k},t) - \rho_0 T\mathbf{k}^2 G_{\theta\rho}(\mathbf{k},t) + \int_0^t \mathrm{d}s [\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k},t-s)\partial_s G_{\rho\rho}(\mathbf{k},s) + K(\mathbf{k})\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t-s)G_{\rho\rho}(\mathbf{k},s)] + \int_0^t \mathrm{d}s\, \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t-s)G_{\theta\rho}(\mathbf{k},s). \tag{31}$$

One can likewise obtain $\hat{\theta}\rho$-element of the SD equation as

$$G_{\theta\rho}(\mathbf{k},t) = \Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{k},0)G_{\rho\rho}(\mathbf{k},t) - \int_0^t \mathrm{d}s\, \Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{k},t-s)\partial_s G_{\rho\rho}(\mathbf{k},s) - \int_0^t \mathrm{d}s [K(\mathbf{k})\Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{k},t-s)G_{\rho\rho}(\mathbf{k},s) + \Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{k},t-s)G_{\theta\rho}(\mathbf{k},s)]. \tag{32}$$

We now take the two crucial steps which can give a closed equation for $G_{\rho\rho}(\mathbf{r},t)$ in *one-loop* order:

- Equations (31) and (32) imply that the integral involving $G_{\theta\rho}(\mathbf{r},t)$ in (31) are of higher order and hence can be discarded in the one-loop order (note that $[G_0]_{\theta\rho} = 0$ from (24)).
- As we alluded earlier, using the cancellation of the three underlined terms in (15), one can eliminate $-\rho_0 T\mathbf{k}^2 G_{\theta\rho}(\mathbf{k},t)$ in (31), together with the two vertices $V^{\mathrm{id}}_{\hat{\rho}\rho\rho}$ and $V_{\hat{\rho}\rho\theta}$ appearing in (30). It should be emphasized that the cancellation is a nonperturbative effect which is required to hold only when the constraint (12) is employed. Therefore, we discard $-\rho_0 T\mathbf{k}^2 G_{\theta\rho}(\mathbf{k},t)$ and retain only those terms arising from the remaining three vertices $V^{\mathrm{int}}_{\hat{\rho}\rho\rho}$, $V_{\rho\hat{\rho}\hat{\rho}}$ and $V_{\hat{\theta}\rho\rho}$ in evaluating the one-loop self-energies.

Taking these steps we obtain the equation of motion for the density correlation function up to the *one-loop order* as

$$\partial_t G_{\rho\rho}(\mathbf{k},t) = -\rho_0 T\mathbf{k}^2 K(\mathbf{k}) G_{\rho\rho}(\mathbf{k},t) + \int_0^t \mathrm{d}s [\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k},t-s)\partial_s G_{\rho\rho}(\mathbf{k},s) + K(\mathbf{k})\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t-s)G_{\rho\rho}(\mathbf{k},s)]. \tag{33}$$

In the absence of particle interaction ($U = 0$), equation (33) reduces to the diffusion equation (22) since, as shown below, the self-energies in (33) involve the vertex $V^{\mathrm{int}}_{\hat{\rho}\rho\rho}$ which contains the interaction potential $\hat{U}$.

Now we compute the one-loop self-energies $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k},t)$ and $\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)$. With the three surviving vertices $V^{\mathrm{int}}_{\hat{\rho}\rho\rho}$, $V_{\rho\hat{\rho}\hat{\rho}}$ and $V_{\hat{\theta}\rho\rho}$, we find that there is the only one nonvanishing diagram for $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{12})$, which is given by

$$\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{12}) = V^{\mathrm{int}}_{\hat{\rho}\rho\rho}(\mathbf{134})V_{\hat{\theta}\rho\rho}(\mathbf{256})G_{\rho\rho}(\mathbf{35})G_{\rho\rho}(\mathbf{46}). \tag{34}$$

Likewise, there are three nonvanishing contributions to the self-energy $\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{12})$:

$$\begin{aligned}\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{12}) &= \Sigma^{(1)}_{\hat{\rho}\hat{\rho}}(\mathbf{12}) + \Sigma^{(2)}_{\hat{\rho}\hat{\rho}}(\mathbf{12})\\ \Sigma^{(1)}_{\hat{\rho}\hat{\rho}}(\mathbf{12}) &= V^{\mathrm{int}}_{\hat{\rho}\rho\rho}(\mathbf{134})V^{\mathrm{int}}_{\hat{\rho}\rho\rho}(\mathbf{256})G_{\rho\rho}(\mathbf{35})G_{\rho\rho}(\mathbf{46})\\ \Sigma^{(2)}_{\hat{\rho}\hat{\rho}}(\mathbf{12}) &= V^{\mathrm{int}}_{\hat{\rho}\rho\rho}(\mathbf{134})V_{\rho\hat{\rho}\hat{\rho}}(\mathbf{562})G_{\rho\rho}(\mathbf{35})G_{\rho\hat{\rho}}(\mathbf{46}),\end{aligned} \tag{35}$$

$\Sigma^{(1)}_{\hat{\rho}\hat{\rho}}$ in (35) comes solely from the interaction contribution to the body force and $\Sigma^{(2)}_{\hat{\rho}\hat{\rho}}$ from the cross-contribution of the interaction and multiplicative thermal noise. By multiplying the second equation in the transformation (16) by $\delta\rho(\mathbf{0},0)$, taking average and using causality, one obtains

$$G_{\rho\hat{\rho}}(\mathbf{r},t)=\Theta(t)(\hat{K}*G_{\rho\rho}(\mathbf{r},t)+G_{\rho\theta}(\mathbf{r},t)). \tag{36}$$

When (36) is substituted into (35), the correlation function $G_{\rho\theta}(\mathbf{r},t)$ will make null contribution in the one-loop order. Using this fact one can rewrite (35) as

$$\begin{aligned}\Sigma^{(1)}_{\hat{\rho}\hat{\rho}}(\mathbf{12})&=V^{\rm int}_{\hat{\rho}\rho\rho}(\mathbf{134})V^{\rm int}_{\hat{\rho}\rho\rho}(\mathbf{256})G_{\rho\rho}(\mathbf{35})G_{\rho\rho}(\mathbf{46})\\ \Sigma^{(2)}_{\hat{\rho}\hat{\rho}}(\mathbf{12})&=V^{\rm int}_{\hat{\rho}\rho\rho}(\mathbf{134})V_{\rho\hat{\rho}\hat{\rho}}(\mathbf{562})G_{\rho\rho}(\mathbf{35})\hat{K}*G_{\rho\rho}(\mathbf{46})\Theta(t_4-t_6).\end{aligned} \tag{37}$$

Therefore, equations (33), (34) and (37) consist of a closed equation for the density correlation function $G_{\rho\rho}(\mathbf{r},t)$ alone.

The Fourier components of the self-energies are now computed as

$$\begin{aligned}\Sigma_{\hat{\rho}\theta}(\mathbf{k},t)&=-\frac{1}{2\rho_0^2}\int_{\mathbf{q}}V(\mathbf{k},\mathbf{q})G_{\rho\rho}(\mathbf{q},t)G_{\rho\rho}(\mathbf{k}-\mathbf{q},t)\\ \Sigma^{(1)}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)&=\int_{\mathbf{q}}V^2(\mathbf{k},\mathbf{q})G_{\rho\rho}(\mathbf{q},t)G_{\rho\rho}(\mathbf{k}-\mathbf{q},t)\\ \Sigma^{(2)}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)&=-\frac{1}{2}\int_{\mathbf{q}}V^2(\mathbf{k},\mathbf{q})G_{\rho\rho}(\mathbf{q},t)G_{\rho\rho}(\mathbf{k}-\mathbf{q},t)\\ &\quad-\frac{T}{2\rho_0}\mathbf{k}^2\int_{\mathbf{q}}V(\mathbf{k},\mathbf{q})G_{\rho\rho}(\mathbf{q},t)G_{\rho\rho}(\mathbf{k}-\mathbf{q},t)\end{aligned} \tag{38}$$

where $V(\mathbf{k},\mathbf{q})\equiv[(\mathbf{k}\cdot\mathbf{q})U(\mathbf{q})+\mathbf{k}\cdot(\mathbf{k}-\mathbf{q})U(\mathbf{k}-\mathbf{q})]$. Arranging the sum of the self-energies $\Sigma^{(i)}_{\hat{\rho}\hat{\rho}}$ with $i=1,2$, one can rewrite $\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)$ as

$$\begin{aligned}\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)&\equiv\mathbf{k}^2\,\tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)\\ \tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)&=\frac{1}{2}\int_{\mathbf{q}}\left[\hat{V}^2(\mathbf{k},\mathbf{q})-\frac{T}{\rho_0}V(\mathbf{k},\mathbf{q})\right]G_{\rho\rho}(\mathbf{q},t)G_{\rho\rho}(\mathbf{k}-\mathbf{q},t)\end{aligned} \tag{39}$$

with $\hat{V}(\mathbf{k},\mathbf{q})\equiv V(\mathbf{k},\mathbf{q})/|\mathbf{k}|$.

In order to appreciate what the closed equation (33) implies, let us for the moment ignore the contribution of the self-energy $\Sigma_{\hat{\rho}\theta}(\mathbf{k},t)$ in (28). Then, equation (33) would become

$$\partial_t G_{\rho\rho}(\mathbf{k},t)=-\rho_0 T\mathbf{k}^2K(\mathbf{k})G_{\rho\rho}(\mathbf{k},t)+\mathbf{k}^2K(\mathbf{k})\int_0^t \mathrm{d}s\,\tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t-s)G_{\rho\rho}(\mathbf{k},s). \tag{40}$$

One can see from (40) that the effect of the nonlinear contribution $\tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k},t)$ is to renormalize the 'bare' lifetime $\tau_0(\mathbf{k})\equiv(\rho_0 T\mathbf{k}^2K(\mathbf{k}))^{-1}$[5] as follows. Defining the Laplace transform $G^L_{\rho\rho}(\mathbf{k},z)\equiv\int_0^\infty \mathrm{d}t\,\mathrm{e}^{-zt}G_{\rho\rho}(\mathbf{k},t)$, etc, we obtain the Laplace transform of (40) as

$$G^L_{\rho\rho}(\mathbf{k},z)=G_{\rho\rho}(\mathbf{k},0)\cdot\left[z+\tau_R^{-1}(\mathbf{k},z)\right]^{-1} \tag{41}$$

where we defined the renormalized lifetime as

$$\tau_R(\mathbf{k},z)\equiv\tau_0(\mathbf{k})\left(1-\frac{1}{\rho_0 T}\tilde{\Sigma}^L_{\hat{\rho}\hat{\rho}}(\mathbf{k},z)\right)^{-1}=\tau_0(\mathbf{k})\left(1+\frac{1}{\rho_0 T}\tilde{\Sigma}^L_{\hat{\rho}\hat{\rho}}(\mathbf{k},z)\right), \tag{42}$$

the last equality holding in the one-loop theory. Substituting (42) into (41), one obtains

$$G^L_{\rho\rho}(\mathbf{k},z)=G_{\rho\rho}(\mathbf{k},0)\cdot\left[z+\frac{\tau_0^{-1}(\mathbf{k})}{1+\tilde{\Sigma}^L_{\hat{\rho}\hat{\rho}}(\mathbf{k},z)/\rho_0 T}\right]^{-1}. \tag{43}$$

The corresponding equation in time domain is given by

$$\partial_t G_{\rho\rho}(\mathbf{k}, t) = -\rho_0 T \mathbf{k}^2 K(\mathbf{k}) G_{\rho\rho}(\mathbf{k}, t) + \int_0^t \mathrm{d}s\, \tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k}, t-s)(-\partial_s G_{\rho\rho}(\mathbf{k}, s)/\rho_0 T). \tag{44}$$

Equation (44) is in fact obtained by replacing $G_{\rho\rho}(\mathbf{k}, s)$ in the convolution integral in (40) by $(-\partial_s G_{\rho\rho}(\mathbf{k}, s)/\rho_0 T \mathbf{k}^2 K(\mathbf{k}))$, which is valid up to the one-loop order.

It is clear that the so far ignored self-energy $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k}, t)$ makes a new contribution to the renormalization of the bare lifetime $\tau_0(\mathbf{k})$, which comes from the consistency requirement of the perturbation theory with the FDR. Now restoring the contribution of $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k}, t)$ and adding it to (44), we obtain

$$\begin{aligned} \partial_t G_{\rho\rho}(\mathbf{k}, t) &= -\rho_0 T \mathbf{k}^2 K(\mathbf{k}) G_{\rho\rho}(\mathbf{k}, t) - \int_0^t \mathrm{d}s\, \Sigma_{MC}(\mathbf{k}, t-s)\partial_s G_{\rho\rho}(\mathbf{k}, s), \\ \Sigma_{MC}(\mathbf{k}, t) &\equiv \left(-\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k}, t) + \frac{1}{\rho_0 T}\tilde{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{k}, t)\right) \\ &= \frac{1}{2\rho_0 T}\int_{\mathbf{q}} \hat{V}^2(\mathbf{k}, \mathbf{q}) G_{\rho\rho}(\mathbf{q}, t) G_{\rho\rho}(\mathbf{k}-\mathbf{q}, t). \end{aligned} \tag{45}$$

This equation reduces to the closed dynamic equation for the density correlation function in the standard MCT if $U(\mathbf{k})$ is replaced by $-Tc(\mathbf{k})$. The self-energy $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{k}, t)$ and the part of the self-energy $\Sigma^{(2)}_{\hat{\rho}\hat{\rho}}(\mathbf{k}, t)$ which is multiplied by $\rho_0 T$ cancel against each other to yield the last line in (45).

The equation for the nonergodicity parameter (NEP) $f(\mathbf{k})$ defined as $f(\mathbf{k}) \equiv G_{\rho\rho}(\mathbf{k}, t \to \infty)/G_{\rho\rho}(\mathbf{k}, 0)$ can be obtained from (45) as

$$\frac{f(\mathbf{k})}{1-f(\mathbf{k})} = \frac{\Sigma_{MC}(\mathbf{k}, \infty)}{\rho_0 T \mathbf{k}^2 K(\mathbf{k})} = \frac{1}{2\rho_0 T^2 \mathbf{k}^2}\int_{\mathbf{q}} \hat{V}^2(\mathbf{k}, \mathbf{q}) S(\mathbf{k}) S(\mathbf{q}) S(\mathbf{k}-\mathbf{q}) f(\mathbf{q}) f(\mathbf{k}-\mathbf{q}). \tag{46}$$

In obtaining (46), we used the inverse relationship between $K(\mathbf{k})$ and the static structure factor $S(\mathbf{k})$: $\rho_0 K(\mathbf{k}) = (1+\rho_0 U(\mathbf{k})/T) = (1-\rho_0 c(\mathbf{k})) = S^{-1}(\mathbf{k})$. The NEP equation (46) is similar to the corresponding one in ABL, except that there is no memory kernel involved in ABL. It naturally occurs from (46) that $f(\mathbf{k}) = 0$ is the only solution for the noninteracting case $(U = 0)$. But this feature is absent in ABL's theory.

In summary, in order to obtain the FDR-preserving perturbation theory, we here proposed a simpler but crucial linearization scheme in which the new variable is employed to take care of only the nonlinear part of the TR transformation. We then recognized that there is a characteristic nonperturbative cancellation effect in the theory. This feature enables us to obtain in the one-loop theory a closed dynamic equation for the two-point density correlation function. This equation is shown to be the same as that of the standard MCT.

Having established the relation of our field-theoretical treatment with the standard MCT, we are now at the starting point to venture into ambitious tasks such as higher order loop calculations and multibody correlations.

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