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# Dynamical van der Waals Model of Glassy Behavior[1]

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A model fluid system whose intermolecular forces consist of the short range part characterizing usual stable fluid and the long range part of Kac-type is considered. When the ratio of the force ranges is large enough, the system can be described by regarding the system having only short range interactions as a reference fluid treated as a continuum, on which long range forces acting among tiny fluid elements are superimposed. We discuss the glassy behavior of this model relating it to the mode coupling theory and using real replica theory. These theories lead to the two equations for non-ergodicity parameter which are totally different from each other. We argue that our model can be a basis for examining nature of the drastic approximations entering derivations of the mode coupling equations. We further explore the possibility of developing the dynamical real replica approach for our model system with the hope of providing a framework to cope with different time scales characterizing complex glassy behavior.

**KEY WORDS:** Glassy behavior; van der Waals; replica approach; mode coupling theory.

## 1. INTRODUCTION

The problem of supercooled liquids and glass transiton is said to be characterized by short length scales of several to a few tens of Ångstroms

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[1] It is a great plesure to dedicate this acrticle to Michael Fisher on his 70th birthday. His monumental works on phase transitions served as invaluable guiding posts for the author over the past four decades.

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and long time scales ranging from a few nanoseconds to more than a year.(1) Because of this variation in scales, treatment of a real glassy liquid, especially its dynamical properties has gotten to be an extremely difficult problem. Existing analytical approaches to dynamics(1) are much hampered by this difficulty despite surprising successes in describing in some detail the behavior before the onset of glass transitions.(1) There appears to be no first principles theory of comparable success describing what happens after onset of the predicted ergodic-nonergodic transition including dynamics. In order to get further insights into the problems of glass transitions including dynamics, it should be useful to construct a model where this microscopic length scale can be extended to a macroscopic level (or, more accurately, to a mesocopic level). In such a model both space and time scales are clearly separated into microscopic and macroscopic (or mesoscopic) scales which should make clear theoretical analyses possible.

There are in nature glassy systems which seem to achieve such separation. Examples are colloidal systems (for instance ref. 2) and physical gel systems (for instance ref. 3). These, however, are beset with other complications. For colloids, treatment of hydrodynamic interactions presents a formidable problem. Although such interactions are believed to influence only short time behavior well separated from long time glassy behavior, there appears to be no convincing demonstration of this fact. Physical gels have a relevant mesoscopic length scale, but they are complicated generally ill-defined polymer systems. For instance, polymerization indices of polymers constituting such sytems are not easy to control.

Here we propose a model which is a dynamical extension of van der Waals model of fluid which played such a prominent role in the developments of phase transition theories. Here physically important behaviors such as phase transitions and glassy dynamics are attributed to long range interactions whereas short range forces mainly serve to endow the reference fluid the properties of a stable liquid with no unusual behavior. The short distance aspects can then be safely treated by macroscopic means. In other words a fluid in the absence of long range interactions can be treated merely as some stable reference fluid whose properties can be put in by hand according to our convenience. This is especialy relevant for dynamics since microscopic dynamical treatment of dense fluid is an old prohibitively difficult problem. In other words, we can now treat glassy *dynamics* of our van der Waals liquid by mapping it onto a nonlinear and nonlocal field theory. This is one crucial novel feature of our approach

Here we comment on the meaning of model construction which generally contains unrealistic elements. (1) If the model is nontrivial and is exactly solvable it has interests in its own right for mathematical physicists. (2) If the model can be treated with controlled approximation even though not exactly solvable, the model permits sharpening of concepts introduced in computational studies or by crude approximations. (3) There are possibilities of extracting features that can go beyond the specific model. Their correct interpretations and proper generalizations might lead to new insights into nature of real systems.[4]

There already exist models for glassy behavior with long-range intermolecular potentials.(7–10) The dynamics in these models is introduced through Monte-Carlo algorithm or by writing down ususal time-dependent Ginzburg–Landau equations where glassy behavior entirely comes from Hamiltonians or free-energy functionals. In contrast, one main objectve of this work is to propose a model of fluid where dynamics naturally occurs through *hydrodyanmic equations for the "reference fluid"* which will be precisely defined below. We shall see that the glassy behavior of fluid can not be ascribed entirely to the free energy functional.

Such a clear-cut dynamical van der Waals model was used before in connection with critical dynamics(11) but has not yet been proposed for studying glassy behavior to the author's knowledge.

This paper is organized as follows. In Section 2 we present our model whose equilibrium properties are given in Section 3. Section 4 discusses the mode coupling theory (MCT) on the basis of our model. In Section 5 we consider an application of the real replica approach to the model followed in Section 6 by discussion of possible nonergodic states where close correspondence of our model with stripe glass(7) is indicated. In Section 7 we attempt dynamical extension of real replica theory. At this moment there is no generally accepted approach to tackle complicated glassy dynamics having many different time scales.(12) So this section proposes our modest first step. In Section 8 we derive the model from a microscopic starting point.

## 2. THE MODEL CONSTRUCTION

We consider a certain reference fluid which is superimposed with an additional long range pair interaction. Basic philosophy of the model was discussed in connection with critical dynamics of fluids in ref. 11. Namely, we initially consider a classical many particle system interacting with short

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[4] As for the toy model recently introduced by us(4, 5) we found that (a) a new kind of gross variable other than the usual density and velocity variables emerge in the region where non-ergodicty is expected, (2) emergence of non-ergodic states is intimately connected to the singular nature of the diffusion matrix of the reduced Fokker–Planck equation for the density variables.

range forces of range $r_0$ and long range forces of range $l$. We assume the ratio to be extremely small: $r_0/l \ll 1$. We focus on the processes occurring over the length scales of $l$ or longer. Then it is legitimate to treat the system as consisting of a reference fluid with only short range interactions where we can superpose long range interactions regarded as interactions among tiny but *macroscopic* elements of the reference fluid. Here we must assume the reference fluid to be a stable fluid devoid of any unusual behavior like phase transitions.

In this section we first write down a complete model equation for a single-component fluid. We then give its simplified version which can be used for supercooled liquids. We also mention equations more appropriate for solids.

## 2.1. Full Model Equation of Single-Component Fluid

At the beginning we introduce the notation for various quantities pertaining to the reference fluid as follows.(6)

$\rho(\mathbf{r}, t)$: number density, $m$: particle mass, $\mathbf{v}(\mathbf{r}, t)$: velocity field

$p(\mathbf{r}, t)$: local pressure, $\eta$: shear viscosity, $\zeta$: bulk viscosity

$\lambda$: thermal conductivity, $T(\mathbf{r}, t)$: local tempertature

$\sigma_{jl}(\mathbf{r}, t)$: $jl$-component of the local stress tensor containing dissipative and fluctuating parts

$\mathbf{q}(\mathbf{r}, t)$: the local heat flux density containing dissipative and fluctuating parts

$s(\mathbf{r}, t)$: the local entropy per molecule

The long-range interaction potential $U(r)$ can generally be taken to be of the following Kac form:(13)

$$U(r) = l^{-d} U^*(r/l) \tag{2.1}$$

where $d$ is the dimensionality of space and $U^*(x)$ is a function such that

$$l^{-d} \int d\mathbf{r}\, U^*(r/l) < \infty \tag{2.2}$$

With this notation, the set of model equations is written as follows,

$$\frac{\partial}{\partial t}\rho(r,t)+\nabla\cdot\rho(\mathbf{r},t)\,\mathbf{v}(\mathbf{r},t)=0 \tag{2.3}$$

$$\begin{aligned}\rho(\mathbf{r},t)\left[\frac{\partial}{\partial t}v_j(\mathbf{r},t)+\mathbf{v}(\mathbf{r},t)\cdot\nabla v_j(\mathbf{r},t)\right]=&-\frac{1}{m}\frac{\partial}{\partial x_j}p(\mathbf{r},t)+\frac{1}{m}\frac{\partial}{\partial x_l}\sigma_{jl}(\mathbf{r},t)\\&-\rho(\mathbf{r},t)\frac{\partial}{\partial x_j}\int d\mathbf{r}'\,\frac{U(\mathbf{r}-\mathbf{r}')}{m}\,\rho(\mathbf{r}',t)\end{aligned} \tag{2.4}$$

$$\begin{aligned}\rho(\mathbf{r},t)\,T(\mathbf{r},t)\left[\frac{\partial}{\partial t}+\mathbf{v}(\mathbf{r},t)\cdot\nabla\right]s(\mathbf{r},t)=&\frac{1}{2}\sigma_{jl}(\mathbf{r},t)\left(\frac{\partial v_j(\mathbf{r},t)}{\partial x_l}+\frac{\partial v_l(\mathbf{r},t)}{\partial x_j}\right)\\&-\nabla\cdot\mathbf{q}(\mathbf{r},t)\end{aligned} \tag{2.5}$$

where the summation convention is used for repeated indices. The above equations are to be supplemented by the definitions of the $\sigma'$s and $\mathbf{q}$ as follows,

$$\begin{aligned}\sigma_{jl}(r,t)\equiv{}&\eta\left(\frac{\partial v_j(\mathbf{r},t)}{\partial x_l}+\frac{\partial v_l(\mathbf{r},t)}{\partial x_j}-\frac{2}{3}\delta_{jl}\nabla\cdot\mathbf{v}(\mathbf{r},t)\right)\\&+\zeta\delta_{jl}\nabla\cdot\mathbf{v}(\mathbf{r},t)+R_{jl}(\mathbf{r},t)\end{aligned} \tag{2.6}$$

$$\mathbf{q}(\mathbf{r},t)\equiv-\lambda\,\nabla T(\mathbf{r},t)+\mathbf{R}(\mathbf{r},t) \tag{2.7}$$

where the random forces the $R's$ satisfy the following fluctuation-dissipation relations:

$$\begin{aligned}\langle R_{jl}(\mathbf{r},t)\,R_{mn}(\mathbf{r}',t')\rangle={}&2k_BT(\eta(\delta_{jm}\delta_{ln}+\delta_{jn}\delta_{lm})+(\zeta-\tfrac{2}{3}\eta)\,\delta_{jl}\delta_{mn})\\&\times\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\end{aligned} \tag{2.8}$$

$$\langle R_j(\mathbf{r},t)\,R_l(\mathbf{r}',t')\rangle=2\lambda k_BT^2\delta_{jl}\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t') \tag{2.9}$$

$$\langle R_{jl}(\mathbf{r},t)\,R_m(\mathbf{r}',t')\rangle=0 \tag{2.10}$$

Here we want to make some remarks.

- *Nature of the Quantities Appearing in this Model.* Here all the hydrodynamic variables and transport coefficients are for the reference fluid valid over the length scales such that $r_0 \ll$ (length scale) $< l$. One can also discuss truly macroscopic processes occurring over the scales much longer than $l$ on the basis of the proposed model equation. The variables as well as transport coefficients entering these macroscopic equations are generally quite different from those appearing in this section. See, e.g., ref. 11.

- *Derivation of the Model Equations (2.3)–(2.5).* Equation (2.3) is the continuity equation always valid when the reference fluid is conserved. Equation (2.5) in the absence of $\sigma_{jl}$ and $\mathbf{q}$ expresses isoentropic change. Equation (2.4) in the absence of $\sigma_{jl}$ expresses Euler's equation modified in the presence of a slowly varying external force field per molecule denoted as $\mathbf{g}(\mathbf{r}, t)$, that is,

$$\left[\frac{\partial}{\partial t}+\mathbf{v}(\mathbf{r}, t)\cdot\nabla\right]\mathbf{v}(\mathbf{r}, t)=-\frac{1}{m\rho(\mathbf{r}, t)}\nabla p(\mathbf{r}, t)+\mathbf{g}(\mathbf{r}, t) \tag{2.11}$$

We can then choose

$$\mathbf{g}(\mathbf{r}, t)=-\nabla\int d\mathbf{r}'\,\frac{U(\mathbf{r}-\mathbf{r}')}{m}\,\rho(\mathbf{r}', t) \tag{2.12}$$

Dissipative and noise terms can then be added, which are not affected by slowly varying external force field which can functionally depend on the density profile $\rho(\mathbf{r}', t)$ elsewhere. More about this derivation will be given in Section 8.

- *Consistency.* It was shown in ref. 14 that the Landau–Lifshitz theory of hydrodynamic fluctuation is consistent with the known equilibrium properties of fluctuations only if the quantities $T^2\lambda, T\eta$ and $T\zeta$ are constant independent of their space-time coordinates where $T, \lambda, \eta$ and $\zeta$ can separately depend on these coordinates. The source of the difficulty is attributed to the presence of multiplicative noise. That is, the r.h.s. of (2.5) containing a term,

$$\frac{1}{2}\,R_{jl}(\mathbf{r}, t)\left(\frac{\partial v_j(\mathbf{r}, t)}{\partial x_l}+\frac{\partial v_l(\mathbf{r}, t)}{\partial x_j}\right)$$

This deficiency of the Landau–Lifshitz theory was rectified in subsequent investigations.(15) Here we refrain from considering this problem since we will be concerned mostly with simplified models in which only the local density and local velocity variables enter.

- *Fluctuations.* Since the reference fluid is macroscopic on the scale between $r_0$ and $l$, its fluctuations can be safely assumed to be Gaussian. However, this is not always exactly true. For instance in the vicinity of a critical point in which the Ginzburg criterion[5] is violated, non-Gaussian density fluctuations are responsible for non-mean field critical exponents. Nevertheless, for supercooled liquids and glasses, density fluctuations with

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[5] Here this is $l^d\epsilon^{2-d/2}\gg 1$, $\epsilon$ being the dimensionless distance from criticality.

wavelength comparable to $l$ would be most relevant. Still, large amplitude frozen density fluctuations associated with spatial heterogeneity below the so-called mode coupling $T_c$ will not be Gaussian.[6] Even here, fluctuations around heterogeneous density profile in each ergodic component state are Gaussian in this model. The model could provide a good framework for classifying different types of fluctuations according to their amplitudes and space and time scales, which play different roles in different situations like in critical phenomena or non-ergodic states.

- *Strategy*. As it stands the model looks quite complicated since we have used for the reference fluid full nonlinear hydrodynamic equations for a one-component fluid. However, drastic simplifications are possible in individual applications. For instance in solids the hydrodynamic equation can be replaced by simple diffusion or relaxation equation. Such simplifications will be discussed below.

## 2.2. Simplified Fluid Equation

We consider simplifications of the model which will be appropriate to treat supercooled liquids and glasses. Here we will be concerned with length scales of the order of $l$. We expect that heat transfer is rapid enough that the variables $s(\mathbf{r}, t)$ will not play a crucial role, and will be dropped. Further more, the temperature $T$ is assumed to be a constant. In the following we drop terms nonlinear in $\mathbf{v}$ which do not play important roles here. We also retain only the longitudinal component of $\mathbf{v}$, that is, $\nabla \times \mathbf{v} = 0$.

The simplified equation of motion is

$$\frac{\partial}{\partial t} \rho(r, t) + \nabla \cdot \rho(\mathbf{r}, t)\, \mathbf{v}(\mathbf{r}, t) = 0 \tag{2.13}$$

$$\frac{\partial}{\partial t} \mathbf{v}(\mathbf{r}, t) = -\frac{1}{m\rho_0} \nabla p(\mathbf{r}, t) + \nu \nabla^2 \mathbf{v}(\mathbf{r}, t) - \nabla \int d\mathbf{r}' \, \frac{U(\mathbf{r}-\mathbf{r}')}{m} \rho(\mathbf{r}', t) + \mathbf{R}(\mathbf{r}, t) \tag{2.14}$$

where $\nu \equiv (\frac{4}{3}\eta + \zeta)/(m\rho_0)$ is the kinetic longitudinal viscosity, $\rho_0$ being the constant average density. The thermal noise $\mathbf{R}(\mathbf{r}, t)$ satisfies

$$\langle \mathbf{R}(\mathbf{r}, t)\, \mathbf{R}(\mathbf{r}', t') \rangle = \frac{2k_B T \nu}{m^2 \rho_0} \nabla \nabla' \delta(\mathbf{r} - \mathbf{r}')\, \delta(t - t') \tag{2.15}$$

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[6] Here the word "fluctuation" may not be appropriate.

Here we further linearize the local pressure $p(\mathbf{r}, t)$, which is regarded as a function of $\rho(\mathbf{r}, t)$, with respect to $\delta\rho(\mathbf{r}, t) \equiv \rho(\mathbf{r}, t) - \rho_0$ to find

$$p(\mathbf{r}, t) = p(\rho(\mathbf{r}, t)) = p(\rho_0) + \left( \left.\frac{dp(\rho)}{d\rho}\right|_{p=p_0} \right) \delta\rho(\mathbf{r}, t) = p(\rho_0) + mc_0^2\, \delta\rho(\mathbf{r}, t) \tag{2.16}$$

where $c_0$ is the sound speed of the reference fluid. Hence (2.14) is rewritten as

$$\frac{\partial}{\partial t} \mathbf{v}(\mathbf{r}, t) = -\frac{c_0^2}{\rho_0} \nabla\rho(\mathbf{r}, t) + \nu\nabla^2\mathbf{v}(\mathbf{r}, t) - \nabla \int d\mathbf{r}' \, \frac{U(\mathbf{r}-\mathbf{r}')}{m} \rho(\mathbf{r}', t) + \mathbf{R}(\mathbf{r}, t) \tag{2.17}$$

Note that the above equation is completely linear in $\mathbf{v}$ and $\delta\rho$.

If desired the pressure term need not be linearized. For instance, short-range repulsive interaction can be taken into account by assuming $p(\rho) = k_B T/(\rho^{-1} - b)$, $b$ being some positive number.

Further simplifications can be achieved if $\mathbf{v}(\mathbf{r}, \mathbf{t})$ is eliminated between (2.13) and (2.17) and we keep only the nonlinearity associated with the long-range force potential $U(\mathbf{r})$. The result is

$$\begin{aligned} m \frac{\partial^2}{\partial t^2} \delta\rho(\mathbf{r}, t) &= mc_0^2 \nabla^2\, \delta\rho(\mathbf{r}, t) + \rho_0 \nabla^2 \int d\mathbf{r}' \, U(|\mathbf{r}-\mathbf{r}'|)\, \delta\rho(\mathbf{r}', t) \\ &\quad + \nu m \nabla^2 \frac{\partial}{\partial t} \delta\rho(\mathbf{r}, t) + \nabla \cdot \delta\rho(\mathbf{r}, t)\, \nabla \int d\mathbf{r}' \, U(|\mathbf{r}-\mathbf{r}'|)\, \delta\rho(\mathbf{r}', t) \\ &\quad + R(\mathbf{r}, t) \end{aligned} \tag{2.18}$$

where

$$R(\mathbf{r}, t) \equiv -\rho_0 m \nabla \cdot \mathbf{R}(\mathbf{r}, \mathbf{t}) \tag{2.19}$$

$$\langle R(\mathbf{r}, t)\, R(\mathbf{r}', t') \rangle = 2k_B T \rho_0 \nu \nabla^4\, \delta(\mathbf{r}-\mathbf{r}')\, \delta(t-t') \tag{2.20}$$

For the case of solids absence of the velocity field variable further simplifies the problem. One can write down the ususal time-dependent Ginzburg–Landau type equations for the order parameter with or without the conservation law where the Ginzburg–Landau free energy fucntional contains long-range potentials. We shall not dwell on that since this is straightforward although we shall come back to his type of model in Section 7.

In closing this section we emphasize that a reference system is chosen to be always stable with weak Gaussian noise. The usual gradient terms appearing in Ginzburg–Landau type free energy functional are not needed. Non-Gaussian fluctuations near criticality and emergence of spatial heterogeneity in non-ergodic glassy states are due to combined effects of $U$ and reference fluid properties as we shall see below.

## 3. EQUILIBRIUM PROPERTIES OF THE MODEL

We discuss equilibrium properties of the model when long wavelength fluctuations crucial in critical phenomena are neglected. The equilibrium density distribution functional $D_e(\{\rho\}, T)$ is given, apart from normalization factor, by

$$D_e(\{\rho\}, T) \equiv \exp\left[ -\beta F^{\mathrm{ref}}(\{\rho\}, T) - \beta \tfrac{1}{2} \int d\mathbf{r} \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r})\, \rho(\mathbf{r}') \right] \tag{3.1}$$

where $\beta \equiv 1/(k_B T)$ and $F^{\mathrm{ref}}(\{\rho\}, T)$ is the Helmholtz free energy functional of the reference fluid[7] which depends on the temperature and functionally on the density profile.

### 3.1. Mean Field Equilibrium Properties

The mean field theory assumes that $D_e(\{\rho\}, T)$ has a sharp maximum at some $\{\rho\}$ with small fluctuations which we neglect here. For uniform state this maximum occurs at $\rho(\mathbf{r}) = \rho_0 \equiv N/V$, $N$ and $V$ being the total number of particles and the system volume, respectively. Hereafter we consider the system with fixed $N$ which will not be written explicitly unless needed. The Helmholtz free energy is,

$$F(V, T) = F^{\mathrm{ref}}(V, T) + \frac{N^2}{2V} U_{\mathbf{0}} \tag{3.2}$$

where $F^{\mathrm{ref}}(V, T) \equiv F^{\mathrm{ref}}(\{\rho = \rho_0\}, T)$ is the Helmholtz free energy of the reference system and

$$U_{\mathbf{k}} \equiv \int d\mathbf{r}\, U(\mathbf{r})\, e^{i\mathbf{k}\cdot\mathbf{r}} \tag{3.3}$$

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[7] Quantities referring to the reference fluid is shown by a superfix ref.

That is, $U_0 = \int d\mathbf{r}\, U(\mathbf{r})$. Hence the entropy is

$$S = -\left.\frac{\partial F(V,T)}{\partial T}\right|_V = S^{\text{ref}} \tag{3.4}$$

Likewise

$$P = P^{\text{ref}} + \frac{N^2}{2V^2} U_0 \quad \text{(pressure)} \tag{3.5}$$

$$E(S,V) = E^{\text{ref}}(S,V) + \frac{N^2}{2V} U_0 \quad \text{(internal energy)} \tag{3.6}$$

$$\chi_T^{-1} = (\chi_T^{\text{ref}})^{-1} + \left(\frac{N}{V}\right)^2 U_0 \quad \text{(inverse isothermal compressibility)} \tag{3.7}$$

The pressure equation above can be found in refs. 16 and 17. The last equation above can be readily extended to the wave vector dependent inverse susceptibility or the inverse scatttering structure function $S_{\mathbf{k}}^{-1}$ if the wave number is of the order of the inverse range $l^{-1}$ of $U$ as ref. 18

$$\chi_T^{-1}(\mathbf{k}) = (k_B T \rho_0)\, S_{\mathbf{k}}^{-1} = (\chi_T^{\text{ref}})^{-1} + \left(\frac{N}{V}\right)^2 U_{\mathbf{k}} \tag{3.8}$$

We can extend the foregoing to spatially non-uniform situations with length scale equal to or greater than $l$. The total free energy functional is

$$F(\{\rho\}, T) = \int d\mathbf{r}\, f^{\text{ref}}(\rho(\mathbf{r}), T) + \tfrac{1}{2} \int d\mathbf{r} \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \delta\rho(\mathbf{r})\, \delta\rho(\mathbf{r}') \tag{3.9}$$

where $f^{\text{ref}}(N/V, T) \equiv F^{\text{ref}}(V,T)/V$ is the free energy density of the reference systems in a spatially uniform state which naturally includes the entropy contribution associated with the reference system. An additional contribution to the total *thermodynamic* entropy arises from the density profile probability distribution (3.1).

Now, there should be a term linear in $\delta\rho$ in (3.9) as follows,

$$\rho_0 \int d\mathbf{r} \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \delta\rho(\mathbf{r}').$$

If the system volume $V$ is much greater than the volume $l^3$ in which the long range interaction is working (we exclude power-law type interaction), $\int d\mathbf{r}\, U(\mathbf{r}-\mathbf{r}')$ is independent of $\mathbf{r}'$. See (2.2). Hence the above linear expression becomes proportional to $\int d\mathbf{r}\, \delta\rho(\mathbf{r})$ and vanishes.

In this subsection we have dropped gradient terms for reference systems, which are common to Ginzburg–Landau type models because we have chosen our reference systems to be stable, and hence the local order parameter will not vary on a microscopic scale of $r_0$. On macroscopic scales greater than $l$ such gradient terms are generated by performing gradient expansions of the terms containing long-range forces.

## 3.2. Free Energy Functional of the Reference Fluid

So far we have not specified the reference fluid free energy functional which is $F^{\text{ref}}(\{\rho\}, T) = \int d\mathbf{r}\, f^{\text{ref}}(\rho(\mathbf{r}, T))$. We therefore focus our attention on the thermodynamics of the reference fluid. Let us denote the reference system free energy simply as $A(N, V, T)$ (which was previously denoted as $F^{\text{ref}}(V, T)$). This takes the form

$$A(N, V, T) = V a(N/V) \tag{3.10}$$

where $a(\rho)$ is the Helmholtz free energy density of the unifrom reference fluid with the number density $\rho = N/V$ and $T$ dependence of $a$ was suppressed since it is always taken to be constant in this subsection. Then

$$\mu = \left(\frac{\partial A}{\partial N}\right)_{V,T} \quad \text{(chemical potential)}, \qquad P = -\left(\frac{\partial A}{\partial V}\right)_{N,T} \quad \text{(pressure)} \tag{3.11}$$

etc.

What we need is the expansion of $a(\rho)$ around another density $\rho_0$ which will be identified as the average density of our original nonuniform system. Putting $\rho = \rho_o + \phi$ and denoting derivatives with primes as $\frac{dX(\rho)}{d\rho} = X'(\rho)$, we need

$$a(\rho) = a(\rho_0) + a'(\rho_0)\,\phi + \frac{1}{2}\,a''(\rho_0)\,\phi^2 + \frac{1}{3!}\,a'''(\rho_0)\,\phi^3 + \frac{1}{4!}\,a''''(\rho_0)\,\phi^4 + \cdots \tag{3.12}$$

Combining (3.11) with (3.12) we get

$$\mu(\rho) = a'(\rho), \quad P = -a + \rho a'(\rho) = -a + \rho\mu \quad \text{etc.} \tag{3.13}$$

(Note $a$ is a quantity per unit volume, but $\mu$ is a quantity per one molecule.)

Likewise higher order derivatives are

$$a'' = \mu', \qquad a''' = \mu'', \qquad a'''' = \mu''',\ldots \tag{3.14}$$

We would like to express these derivatives in terms of the isothermal compressiblity $\chi_T(\rho)$ and its density derivatives. First note $\mu' = (\partial\mu(P,T)/\partial\rho)_T = (\partial P/\partial\rho)_T\,(\partial\mu(P,T)/\partial P)_T = \rho(\partial P/\partial\rho)_T = 1/\chi_T$. Next we get $\mu'' = (\frac{1}{\chi_T})'$, $\mu''' = (\frac{1}{\chi_T})''$, and so forth.

This results in

$$a(\rho) = a(\rho_0) + \mu(\rho_0)\,\phi + \frac{1}{2}\left(\frac{1}{\chi_T(\rho_0)}\right)\phi^2 + \frac{1}{3!}\left(\frac{1}{\chi_T(\rho_0)}\right)'\phi^3 + \frac{1}{4!}\left(\frac{1}{\chi_T(\rho_0)}\right)''\phi^4 + \cdots \tag{3.15}$$

Thus in the following we start from the expansion as

$$a(\rho) = a(\rho_0) + u_1\phi + \tfrac{1}{2}u_2\phi^2 + \tfrac{1}{3}u_3\phi^3 + \tfrac{1}{4}u_4\phi^4 + \cdots \tag{3.16}$$

where the coefficients $u_i$ are defined by comparison with (3.15).

### 3.3. Model Free Energy Functional

The free energy functional (3.9) now becomes

$$F(\{\phi\},T) = \int d\mathbf{r}(\tfrac{1}{2}u_2\phi(\mathbf{r})^2 + \tfrac{1}{3}u_3\phi(\mathbf{r})^3 + \tfrac{1}{4}u_4\phi(\mathbf{r})^4) + \tfrac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\,U(\mathbf{r}-\mathbf{r}')\,\phi(\mathbf{r})\,\phi(\mathbf{r}') + \tfrac{1}{2}VU_0\rho_0^2 + \cdots \tag{3.17}$$

where $\cdots$ denotes the higher order terms of the expansion of (3.16) and the preceding term to it is the van der Waals term. Here we do not need a term with $(\nabla\phi(\mathbf{r}))^2$ since the length scales of interest are much greater than the correlation length of the reference fluid which is of the order of $r_0$.

Here we make one simplifying assumption that the coefficient $u_3$ vanishes. Then our model reduces to that of familiar $\phi^4$ field theory where instead of the gradient term we have a long-range force term with $U$. We write down the final form of $F(\{\phi\},T)$ which will be analyzed in Sections 5 and 7:

$$F(\{\phi\},T) = \int d\mathbf{r}\,(\tfrac{1}{2}u_2\phi(\mathbf{r})^2 + \tfrac{1}{4}u_4\phi(\mathbf{r})^4) + \tfrac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\,U(\mathbf{r}-\mathbf{r}')\,\phi(\mathbf{r})\,\phi(\mathbf{r}') + \tfrac{1}{2}VU_0\rho_0^2 \tag{3.18}$$

where $u_2, u_4 > 0$ due to stability of the reference fluid.

## 4. DERIVATIONS OF THE IDEALIZED MODE COUPLING THEORY EQUATION

Since the mode coupling theory is undoubtedly the most sucessful first principle dynamical theory of supercooled liquids and glass transitions above certain crossover temperature,(1, 19, 20) any model or theory that purports to describe experimentally observed glassy dynamics of fluid must be able to relate itself to the MCT in the region where the latter is considered to be valid. This problem is taken up for our model in this section. Our aim is to derive the known MCT equatation of fluid for our model where nature of the uncontrolled approximations entering the earlier derivations can be examined.

### 4.1. Langevin Equation Approach

This derivation of the MCT equation follows the same method given in Section 3 of ref. 20 where analogy with Brownian motion theory is used. We start from the simplified model equations obtained before, (2.18), (2.19) and (2.20),

We express (2.18) in terms of Fourier components $\rho_{\mathbf{k}}(t)$ of $\delta\rho(\mathbf{r}, t)$ to find

$$m\frac{\partial^2}{\partial t^2}\rho_{\mathbf{k}}(t) = -m\omega_k^2\rho_{\mathbf{k}}(t) - \nu m k^2\frac{\partial}{\partial t}\rho_{\mathbf{k}}(t) + \mathscr{R}_{\mathbf{k}}(t) + R_{\mathbf{k}}(t) \tag{4.1}$$

where

$$\omega_k^2 \equiv k^2\left(c_0^2 + \frac{\rho_0}{m}U_{\mathbf{k}}\right) \tag{4.2}$$

$$\mathscr{R}_{\mathbf{k}}(t) \equiv -\int_{\mathbf{q}} \mathbf{k}\cdot\mathbf{q}U_{\mathbf{q}}\rho_{\mathbf{k}-\mathbf{q}}(t)\,\rho_{\mathbf{q}}(t) \tag{4.3}$$

and $\int_{\mathbf{q}} \equiv \frac{1}{(2\pi)^d}\int d\mathbf{q}$, $d$ being the dimensionality of space. The constant $\nu$ is the bare kinetic viscosity determined by the microscopic thermal noise of molecular scale $R_{\mathbf{k}}(t)$ by the ususal fluctuation-dissipation relation, with $R_{\mathbf{k}}(t)$ the Fourier transform of (2.19).

Now we make several approximations or assumptions.(20) In a supercooled liquid near the mode coupling critical temperature $T_c$, $\nu$ as well as $R_{\mathbf{k}}(t)$ are not important and will be dropped. On the other hand, $\mathscr{R}_{\mathbf{k}}(t)$ contains many terms of products of the two $\rho$'s with different wave vectors. Hence it will change in irregular manner in time like thermal noise. Thus we assume that $\mathscr{R}_{\mathbf{k}}(t)$ can be treated as a new thermal noise which replaces $R_{\mathbf{k}}(t)$. Accordingly the $\nu$ term is replaced by a term containing the memory kernel which we denote as $\mathscr{M}_{\mathbf{k}}(t)$. The result is the following linear Langevin equation with memory:

$$\frac{\partial^2}{\partial t^2}\,\rho_{\mathbf{k}}(t) = -\omega_k^2 \rho_{\mathbf{k}}(t) - \int_0^t ds\; \mathscr{M}_{\mathbf{k}}(t-s)\,\frac{\partial}{\partial s}\,\rho_{\mathbf{k}}(s) + \frac{1}{m}\,\mathscr{R}_{\mathbf{k}}(t) \tag{4.4}$$

where, denoting time derivative by an overdot,

$$\begin{aligned}\mathscr{M}_{\mathbf{k}}(t) &\equiv \frac{1}{m^2\langle|\dot{\rho}_{\mathbf{k}}|^2\rangle}\,\langle \mathscr{R}_{\mathbf{k}}(t)\,\mathscr{R}_{-\mathbf{k}}(0)\rangle \\ &= \frac{1}{mNk_BTk^2}\int_{\mathbf{q}}\int_{\mathbf{q}'} (\mathbf{k}\cdot\mathbf{q})(\mathbf{k}\cdot\mathbf{q}')\,U_{\mathbf{q}}U_{-\mathbf{q}'}\,\langle \rho_{\mathbf{k}\,q}(t)\,\rho_{\mathbf{q}}(t)\,\rho_{\mathbf{q}\,k}(0)\,\rho_{-\mathbf{q}'}(0)\rangle\end{aligned} \tag{4.5}$$

which replaces the earlier fluctuation-dissipation relation (2.20). The equations (4.4) and (4.5) have similar status as the Mori equation for Brownian dynamics(23) except that our starting point is not completely microscopic. Here one finds $\langle|\dot{\rho}_{\mathbf{k}}|^2\rangle = Nk_BTk^2/m$. We now assume that the density fluctuations obey Gaussian statistics. This will permit us to factorize a correlation function of four density fluctuations into a sum of products of two pair correlation functions. Then, defining the normalized time correlation function of density and the static scattering structure function, respectively, by

$$\phi_{\mathbf{k}}(t) \equiv \frac{\langle \rho_{\mathbf{k}}(t)\,\rho_{-\mathbf{k}}(0)\rangle}{\langle|\rho_{\mathbf{k}}|^2\rangle} \tag{4.6}$$

$$S_k \equiv \frac{1}{N}\,\langle|\rho_{\mathbf{k}}|^2\rangle \tag{4.7}$$

we have

$$\mathscr{M}_{\mathbf{k}}(t) = \frac{\rho_0}{2mk^2k_BT}\int_{\mathbf{q}} |(\mathbf{k}\cdot\mathbf{q})U_{\mathbf{q}} + (\mathbf{k}\cdot(\mathbf{k}-\mathbf{q}))\,U_{\mathbf{k}-\mathbf{q}}|^2\,S_qS_{|\mathbf{k}-\mathbf{q}|}\,\phi_q(t)\,\phi_{|\mathbf{k}-\mathbf{q}|}(t) \tag{4.8}$$

Here we note that (4.4) without $\frac{1}{m}\,\mathscr{R}_{\mathbf{k}}(t)$ is satisfied by $\phi_k(t)$:

$$\left(\frac{\partial^2}{\partial t^2} + \omega_k^2\right)\phi_{\mathbf{k}}(t) = -\int_0^t ds\; \mathscr{M}_{\mathbf{k}}(t-s)\,\frac{\partial}{\partial s}\,\phi_{\mathbf{k}}(s) \tag{4.9}$$

The equations (4.9) and (4.8) with the initial conditions given, by

$$\phi_{\mathbf{k}}(0) = 1, \qquad \dot{\phi}_{\mathbf{k}}(0) = 0 \tag{4.10}$$

constitute a self-consistent set for the $\phi$'s. If $U_{\mathbf{k}}$ is replaced by $-k_B T C_{\mathbf{k}}$,[8] $C_{\mathbf{k}}$ being Fourier component of direct correlation function of liquid theory,(21) this reduces to the self-consistent equations of Götze and others(1, 19) called the idealized MCT equation. In this latter equation one generally does not separate molecular forces into short range and long range parts, hence no justification can be given for the factorization approximation made there as well because the direct correlation function $C(r)$ of a liquid is generally short-ranged. In any event their results suggest that for appropriate choice of the potential $U(r)$ in our equation we can expect the same kind of ergodic-nonergodic transition at the mode coupling $T_c$ of our model with the same kind of properties(1)

The above simple derivation kept silent about the sublte but important fact that the dynamics of $\mathscr{R}_{\mathbf{k}}(t)$ is not the ususal one but the "projected" one in Mori–Zwanzig's sense.(23, 24) An alternative method is to follow the procedure of Section 6 of ref. 20.

Another possibility is to apply the procedure of Dawson and the coworkers(22) which relies on the Gaussian assumption for density fluctuations. This assumption is more naturally acceptable with our model. This is described in the following subsection.

### 4.2. The Dawson Approach

An alternative derivation was proposed by Dawson and the coworkers(22) which avoids use of the projected dynamics mentioned in the preceding subsection. Here we start from (4.4), (4.2), and (4.3) with (4.2) rewritten as:

$$\omega_k^2 \equiv k^2 \left( c_0^2 + \frac{\rho_0}{m} U_{\mathbf{k}} \right) = \frac{k^2 k_B T}{m S_k} \tag{4.11}$$

where (3.8) and $c_0^2 = 1/(m \rho_0 \chi_T^{\mathrm{ref}})$ have been used.[9] We rewrite these as

$$\left( \frac{\partial^2}{\partial t^2} + \omega_k^2 \right) \rho_{\mathbf{k}}(t) = \mathscr{F}_{\mathbf{k}}(t) \tag{4.12}$$

$$\mathscr{F}_{\mathbf{k}}(t) \equiv \frac{1}{m} \mathscr{R}_{\mathbf{k}}(t) \tag{4.13}$$

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[8] Here $C_{\mathbf{k}}$ is not just for the reference fluid but for the original fluid model including long range intermolecular forces if the latter can be separated as in our model.

[9] Here the entropy variable is dropped, and heat transfer is not considered. Thus we simply assume that all the processes are isothermal. This simplification is justified since our primary interest is in slow dynamics in which kinetic energy (or temperature of the reference system) is equilibrated.

Now, as before, if we regard

$$\mathscr{F}_{\mathbf{k}}(t) = \frac{1}{m}\mathscr{R}_{\mathbf{k}}(t) = -\frac{1}{m}\int_{\mathbf{q}} \mathbf{k}\cdot\mathbf{q}U_{\mathbf{q}}\rho_{\mathbf{k}-\mathbf{q}}(t)\,\rho_{\mathbf{q}}(t) \tag{4.14}$$

as a random force, but consider the fact that the real random force, here denoted by $f_{\mathbf{k}}(t)$ in the Brownian motion theory of Mori,(23) takes the same form as (4.14) except that the time dependence is governed by Mori's projected dynamics. These two are related by ref. 23

$$\mathscr{F}_{\mathbf{k}}(t) = -\int_0^t ds\,\mathscr{M}_{\mathbf{k}}(t-s)\,\frac{\partial}{\partial s}\rho_{\mathbf{k}}(s) + f_{\mathbf{k}}(t) \tag{4.15}$$

Here the memory function is rigorously expressed as

$$\mathscr{M}_{\mathbf{k}}(t) = \frac{\langle f_{\mathbf{k}}(t)\,f_{-\mathbf{k}}(0)\rangle}{\langle|\dot{\rho}_{\mathbf{k}}|^2\rangle} \tag{4.16}$$

We then replace $f_{\mathbf{k}}(t)$, and $f_{-\mathbf{k}}(0)$ by the expressions obtained from (4.15), and we have another rigorous expression,

$$\mathscr{M}_{\mathbf{k}}(t) = \frac{1}{\langle|\dot{\rho}_{\mathbf{k}}|^2\rangle}\left\langle\left(\mathscr{F}_{\mathbf{k}}(t) + \int_0^t ds\,\mathscr{M}_{\mathbf{k}}(t-s)\,\frac{\partial}{\partial s}\rho_{\mathbf{k}}(s)\right)\mathscr{F}_{-\mathbf{k}}(0)\right\rangle \tag{4.17}$$

Here we introduce the main ansatz of this approach, namely the density fluctuations with various time arguments obey Gaussian statistics. Then, since $\mathscr{F}_{\mathbf{k}}$ is bilinear in the density fluctuations by (4.14) the second term of (4.17) drops out. The remainder of (4.17) is identical to (4.5). The Gaussian ansatz further reduces this to (4.8). This completes the derivation of self-consistent MCT equation by the Dawson approach.

## 4.3. Non Ergodicity Parameter

We can discuss within the framework of the MCT the non-ergodicity parameter defined in ref. 1

$$f_{\mathbf{k}} \equiv \phi_{\mathbf{k}}(t\to\infty) \tag{4.18}$$

The equation for this is readily obtained from (4.9) and (4.10) as

$$\frac{f_{\mathbf{k}}}{1-f_{\mathbf{k}}} = \frac{1}{\omega_k^2}\,\mathscr{M}_{\mathbf{k}}(\phi\to f) \tag{4.19}$$

where the form of $\mathscr{M}_{\mathbf{k}}(\phi)$ is given by (4.8).

## 4.4. On Justification of the MCT Equation

Central to the above two derivations was the assumption that the $\rho_{\mathrm{k}}(t)$'s obey Gaussian statistics, which permits the factorization approximation of correlation functions of products of four density fluctuations and dropping of the second term of (4.17). This assumption can never be justified for usual liquid models where wave numbers involved are of the orders of inverse intermolucular distances required in MCT. For our van der Waals model the relevant wave numbers involved are of the orders of inverse force range $l$ of $U(\mathbf{r})$. This gives rise to a possibility to examine this crucial Gaussian approximation making use of the fact that this approximation is trivially valid for the reference system for the wave numbers concerned. Let us consider as an example the equal time four body correlation function, which can be obtained by knowing the equilibrium four body distribution function, $F_4(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4)$, where the distances between points, $\mathbf{r}_i$ and $\mathbf{r}_j$ with $i \neq j$, are of the orders of $l$. Since $l$ is much longer than microscopic correlatation length of the reference system, $F_4$ for the reference system is trivially factorizable, and deviations from factorized $F_4$ must involve $U(\mathbf{r})$ which gives rise to an additional factor of the smallness parameter $(r_0/l)^{-d}$. A similar analysis can be made for $F_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$. Expansion of distribution functions in powers of this smallness parameter was discussed in ref. 25. There are two problems in applying this argument to examine validity of the factoriztion approximation in the context of MCT.

- A naïve expansion in powers of the smallness parameter is inadequate since the two body distribution functions $F_2$ that appear in the factorized form of $F_4$ should in fact include effects of $U$. Thus the factozied form of $F_4$ already include partial resummation in powers of the smallness parameter.

- In fact we have to deal with the time-displaced four-body correlation function rather than the equal time one. This difference will not affect a naïve expansion in powers of the smallness parameter since the simplified model equation (2.18) is a linear hydrodynamicc equation in the absence of $U$. Thus a naïve expansion in powers of the smallness parameter is still formally similar with extra nonlinear contributions arising from time evolution. By the same reason as before, we will need resummation where effects of the nonlinear $U$ term must be properly handled.

Clearly, examination of the drastic approximations entering the MCT equation requires careful considerations of the points made here and will be deferred to future research.

## 4.5. Beyond MCT

Here we discuss what to be expected beyond MCT. We go back to the time-dependent Ginzburg–Landau type model described at the end of Section 2.2. This model is the continuum version of the Monte Carlo simulation of single-spin-flip (non-conserved) or spin-exchange (conserved) kinetic Ising models. For $F(\{\phi\})$ of the model we may choose the following:

$$F(\{\phi\}) = F^{\mathrm{ref}}(\{\phi\}) + \tfrac{1}{2}\int d\mathbf{r} \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \phi(\mathbf{r})\, \phi(\mathbf{r}') \tag{4.20}$$

$$F^{\mathrm{ref}}(\{\phi\}) = \int d\mathbf{r}[a(\phi(\mathbf{r}, t)) + \tfrac{1}{2}\, b(\nabla\phi(\mathbf{r}, t))^2] \tag{4.21}$$

with $b$ some positive constant and $a(x)$ a function having a minimum at $x = 0$. Here we have temporarily restored a gradient term in (4.21).

Let us take $a(x) \equiv \frac{1}{2} a_2 x^2 + \frac{1}{4} a_4 x^4$ with $a_2, a_4$ some positive numbers, and $x$ an arbitrary argument in the function $a(x)$. Since $\sqrt{b/a_2}$ is the characteristic length of the reference system, we must choose $l \gg \sqrt{b/a_2}$, $l$ the force range of $U$.

Now, Klein and coworkers(8) found that the static properties of the model similar to ours have clump states at some low temperature region where frozen inhomogeneous density profile appears. If this were true for our fluid model as suggested by static linear instability of the model with a suitable $U(r)$ against density fluctuations, we should expect the density fluctuation $\delta\rho = \rho - \rho_0$ is no longer small and will be frozen. Here $\delta\rho$ can no longer be considered as Gaussian and the self-consistent MCT equation loses its validity. This is in accord with the general belief (not proof) that the mode coupling critical temperature $T_c$ is the point where the Götze type MCT breaks down.

In the following two sections we shall describe our attempts to go beyond the MCT.

## 5. REPLICA ANALYSIS

A promising analytical way to include effects on non-ergodicity is to employ the new replica trick of Monasson.(26) This will be explained in this section in the present context. A technique of similar nature was successfully applied to the first principle calculation of the low temperature properties of amorphous solids from the side of liquids at higher temperatures.(27)

## 5.1. Real Replica Theory

The real replica approach for our model is described following the methods of refs. 7 and 26 where the self-consistent screening approximation(28) was used to implement the theory.

We start from the model (3.18). The equilibrium thermodynamic free energy including density fluctuation effects $A(V,T)$ is given, denoting $\beta \equiv 1/(k_B T)$, by

$$A(V,T) \equiv -\beta^{-1} \ln \int d\{\phi\}\, e^{-\beta F(\{\phi\},T)} \tag{5.1}$$

and also

$$A^{\mathrm{ref}}(V,T) \equiv -\beta^{-1} \ln \int d\{\phi\}\, e^{-\beta F^{\mathrm{ref}}(\{\phi\},T)} \tag{5.2}$$

Now, following(7, 26) we introduce a kind of reference density profile $\{\psi\}$ to which the system is attracted, and also the new free energy functional $\mathscr{F}$ as

$$\mathscr{F}(\{\psi\}) \equiv -\beta^{-1} \ln \int d\{\phi\} \exp\left( -\beta F(\{\phi\},T) - \frac{g}{2} \int d\mathbf{r}[\phi(\mathbf{r}) - \psi(\mathbf{r})]^2 \right) \tag{5.3}$$

where $g > 0$. (We suppress dependence of $\mathscr{F}$ on thermodynamic variables like $T$ other than the density profile.)

We then prepare $M$ replicas and introduce the free energy functional of replicated system divided by $M$ as

$$\begin{aligned} A(M) &\equiv -(\beta M)^{-1} \ln \int d\{\psi\}\, e^{-\beta M \mathscr{F}(\{\psi\})} \\ &= -(\beta M)^{-1} \ln \int \prod_{a=1}^{M} d\{\phi^a\} \exp\left( -\beta \sum_b F(\{\phi^b\},T) \right. \\ &\quad \left. - \frac{g}{2M} \sum_{b>c} \int d\mathbf{r}[\phi^b(\mathbf{r}) - \phi^c(\mathbf{r})]^2 \right) \\ &\equiv -(\beta M)^{-1} \ln \int \prod_{a=1}^{M} d\{\phi^a\}\, e^{-\beta F(\{\boldsymbol{\phi}\},M)} \end{aligned} \tag{5.4}$$

where $a, b$, and $c$ label replicas. Equation (5.4) also defines $F(\{\boldsymbol{\phi}\}, M)$ where $\{\boldsymbol{\phi}\} \equiv \{\phi^1\}, \{\phi^2\}, \{\phi^3\},\ldots, \{\phi^M\}$, superscripts labelling replicas. The thermodynamic equilibrium free energy is obtained by taking $g \to 0+$ and $M = 1$,

$$A = A(M = 1) \tag{5.5}$$

Here the term with $g$ has no effects on ergodic states after taking $g \to 0+$. On the other hand, if the system is in a non-ergodic state which is trapped in a metastable state, $g$ has the effect of keeping $M$ replicated systems in the same metastable state. Then, any observable physical quantity is an average of such a quantity in each metable state, which is further averaged over all metastable states with proper weights. We denote such an averaged quantity by attaching asuffix $c$. Then, in this formalism we find[(26)]

$$S_c = \beta(A_c - A) = \beta\, \partial A(M)/\partial M|_{M=1} \tag{5.6}$$

$$A_c = \partial(MA(M))/\partial M|_{M=1} \tag{5.7}$$

where $S_c$ is the configurational entropy and $A_c$ is called the "internal energy."[(26)]

We now turn to (5.4),

$$A(M) = -(\beta M)^{-1} \ln \int \prod_{a=1}^{M} d\{\phi^a\}\, e^{-\beta F(\{\phi\}, M)} \tag{5.8}$$

The correlation matrix $\hat{\mathscr{G}}$ in replica space is expressed through its $ab$-element as

$$(\hat{\mathscr{G}}_{\mathbf{k}})_{ab} = \beta\langle \phi^a_{\mathbf{k}} \phi^b_{-\mathbf{k}} \rangle \tag{5.9}$$

Then the two types of correlation functions are defined as

$$\begin{aligned} \mathscr{G}(\mathbf{r}-\mathbf{r}') &\equiv \beta\langle \phi(\mathbf{r})^a\, \phi(\mathbf{r}')^a \rangle \\ \mathscr{F}(\mathbf{r}-\mathbf{r}') &\equiv \lim_{t\to\infty} \beta\langle \phi(\mathbf{r}, t)^a\, \phi(\mathbf{r}', 0)^a \rangle \end{aligned} \tag{5.10}$$

$a$ being any replica index. The second member of the above equation is the Edwards-Anderson (EA) type correlation function whereas the first one is the usual equal time correlation function. Here one has assumed symmetry in replica indices in view of the symmetry of the $g$-term. This permits us to make the following Ansatz[(7)] (indices $\mathbf{k}$ are occasionally suppressed):

$$\hat{\mathscr{G}} = (\mathscr{G} - F)\, \mathbf{1} + \mathscr{F}\, \mathbf{E} \tag{5.11}$$

where **1** is the unit matrix in the replica space and **E** is the matrix, all of whose elements are unity. We can then write

$$\hat{\mathscr{G}}^{-1} = \mathscr{G}_0^{-1}\mathbf{1} + \hat{\Sigma} - \frac{g}{M}\mathbf{E} \tag{5.12}$$

where $\mathscr{G}_0$ is the correlation function in the absence of both the nonlinear and $g$-terms. This also defines the self-energy part $\hat{\Sigma}$. Next we make an Ansatz similar to (5.11) as

$$\hat{\Sigma} = (\Sigma_{\mathscr{G}} - \Sigma_{\mathscr{F}})\,\mathbf{1} + \Sigma_{\mathscr{F}}\mathbf{E} \tag{5.13}$$

After some algebra we find in the limits $M \to 1, \quad g \to 0+$ the following(7)

$$\mathscr{G}^{-1} = \mathscr{G}_0^{-1} + \Sigma_{\mathscr{G}} \tag{5.14}$$

We then find(7)

$$\mathscr{F} = -(\mathscr{G} - F)^{-1}\,\mathscr{G}\Sigma_{\mathscr{F}}$$

or,

$$\mathscr{F} = \mathscr{G} - K, \qquad \mathscr{K}^{-1} \equiv \mathscr{G}^{-1} - \Sigma_{\mathscr{F}} \tag{5.15}$$

In order to proceed further we need to introduce an approximation to deal with the free energy functional (3.18). Thus we first tried the random phase approximation which is considered to be good for fluids with long-range interaction potential. See ref. 21, Chapt. 6. We found that there is no possibility of producing non-ergodic states in this aproximation since the density fluctuations of the reference fluid are dealt with in the linear approximation here.

## 5.2. Self-Consistent Screening Approximation (SCSA)

The replica analysis of a model of stripe glass was recently carried out(7) using the self-consistent-screening approximation(SCSA) of Bray.(28) The importance of SCSA(28) as compared to other approximation scheme such as RPA arises from the fact that SCSA seems to be capable of dealing with symmetry breaking in a very natural way. This together with replica trick described in the preceding section would permit us to treat glass-like behavior with a complicated (free) energy landscape.(7, 26) In the following we try to map our problem to this analysis. The main difference is the absence of the gradient term and a general form of long-range interaction in our case, which will only affect linear propagator $\mathscr{G}_{0\mathbf{k}}$, which in our case arises from the quadratic terms in $\phi$ in (3.18), is

$$\mathscr{G}_{0\mathbf{k}}^{-1} = u_2 + U_{\mathbf{k}} \tag{5.16}$$

We argue later that in fact there is a close connection between the two models of self-generated glassy systems, i.e., stripe and van der Waals glasses.

We now rewrite $F(\{\boldsymbol{\phi}\}, M)$ entering (5.4) as

$$\begin{aligned} F(\{\boldsymbol{\phi}\}, M) = & \sum_a \int d\mathbf{r} \left( \frac{u_2}{2} [\phi^a(\mathbf{r})]^2 + \frac{u_4}{4} [\phi^a(\mathbf{r})]^4 \right) \\ & + \sum_a \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \, U(\mathbf{r}-\mathbf{r}') \, \phi^a(\mathbf{r}) \, \phi^a(\mathbf{r}') \\ & - \frac{g}{2M\beta} \sum_{a,b} \int d\mathbf{r} \, \phi^a(\mathbf{r}) \, \phi^b(\mathbf{r}) + \frac{1}{2} MVU_0 \rho_0^2 \end{aligned} \tag{5.17}$$

We consider that the difference

$$\frac{g}{2M\beta} \sum_{a>b} \int d\mathbf{r} ([\phi^a(\mathbf{r})]^2 + [\phi^b(\mathbf{r})]^2) - \frac{g}{2M\beta} \int d\mathbf{r} \sum_a [\phi^a(\mathbf{r})]^2$$

is absorbed into the first term of (5.17). First, we must extend the scalar order parameter $\phi$ into the $n$ component vector order parameter $\phi_i$ with $i = 1, 2, \ldots, n$. Then we initially suppose that $n$ is large and assume $u_4 \propto 1/n$. We attempt an expansion in $1/n$ and assume an analytic continuation to smaller $n$, even to $n \leqslant 1$.(26)

The subsequent algebra is similar to that in ref. 7 and we can omit intermediate steps. The final outcome is summarized by the following self-consistent set of equations for Fourier components of (5.10), $\mathscr{G}_{\mathbf{k}}$, $\mathscr{F}_{\mathbf{k}}$.

$$\mathscr{G}_{\mathbf{k}}^{-1} = \mathscr{G}_{0\mathbf{k}}^{-1} + \Sigma_{\mathscr{G}\mathbf{k}} \tag{5.18}$$

$$\mathscr{F}_{\mathbf{k}} = \mathscr{G}_{\mathbf{k}} - \mathscr{K}_{\mathbf{k}}, \qquad \mathscr{K}_{\mathbf{k}}^{-1} \equiv \mathscr{G}_{\mathbf{k}}^{-1} - \Sigma_{\mathscr{F}\mathbf{k}} \tag{5.19}$$

$$\Sigma_{\mathscr{G}\mathbf{k}} = \Sigma^H + \frac{2}{n} \int_{\mathbf{q}} \mathscr{D}_{\mathscr{G}\mathbf{q}} \mathscr{G}_{\mathbf{k}-\mathbf{q}} \tag{5.20}$$

$$\Sigma_{\mathscr{F}\mathbf{k}} = \frac{2}{n} \int_{\mathbf{q}} \mathscr{D}_{\mathscr{F}\mathbf{q}} \mathscr{F}_{\mathbf{k}-\mathbf{q}} \tag{5.21}$$

$$\Sigma^{H} = nu_4 \int_{\mathbf{q}} \mathscr{G}_{\mathbf{q}} \tag{5.22}$$

$$\mathscr{D}_{\mathscr{G}\mathbf{k}} = \left( \frac{\beta}{nu_4} + \Pi_{\mathscr{G}\mathbf{k}} \right)^{-1} \tag{5.23}$$

$$\mathscr{D}_{F\mathbf{k}} = -[1 - \Pi_{\mathscr{F}\mathbf{k}} \mathscr{D}_{G\mathbf{k}}]^{-1} \, \mathscr{D}_{G\,\mathbf{k}}^{\,2} \Pi_{\mathscr{F}\mathbf{k}} \tag{5.24}$$

$$\Pi_{\mathscr{G}\mathbf{k}} = \int_{\mathbf{q}} \mathscr{G}_{\mathbf{q}} \mathscr{G}_{\mathbf{k}-\mathbf{q}} \tag{5.25}$$

$$\Pi_{\mathscr{F}\mathbf{k}} = \int_{\mathbf{q}} \mathscr{F}_{\mathbf{q}} \mathscr{F}_{\mathbf{k}-\mathbf{q}} \tag{5.26}$$

The configurational entropy $S_c$ or its density $s_c \equiv S_c/V$ is given by refs. 7 and 26

$$\begin{aligned} s_c = & -\frac{1}{2} \int_{\mathbf{k}} \left\{ \ln\left(1 - \frac{\mathscr{F}_{\mathbf{k}}}{\mathscr{G}_{\mathbf{k}}}\right) + \frac{\mathscr{F}_{\mathbf{k}}}{\mathscr{G}_{\mathbf{k}}} \right\} \\ & + \frac{1}{2} \int_{\mathbf{k}} \left\{ \ln\left(1 - \frac{v\Pi_{\mathscr{F}\mathbf{k}}}{1 + v\Pi_{\mathscr{G}\mathbf{k}}}\right) + \frac{v\Pi_{\mathscr{F}\mathbf{k}}}{1 + v\Pi_{\mathscr{G}\mathbf{k}}} \right\} \end{aligned} \tag{5.27}$$

with $v \equiv \frac{nu_4}{\beta}$. The expressions (5.18)–(5.20) follow by minimizing the following expression for the free energy appropriate for the SCSA approximation with respect to $\mathscr{G}_{\mathbf{k}}$.(7, 29)

$$\frac{\beta A(M)}{2M} = \mathrm{Tr} \ln \hat{\mathscr{G}}^{-1} + \mathrm{Tr} \ln \hat{\mathscr{D}}^{-1} - \mathrm{Tr}\, \hat{\Sigma} \hat{\mathscr{G}} \tag{5.28}$$

where matrix product includes integration over wave vectors. We will take $n = 1$ in the following.

Before concluding this subsection we briefly touch upon related aproximate methods to handle nonlinear field equations. Some time ago Kraichnan introduced one of the first successful methods now known as direct interaction approximation (DIA),(30) which is, in fact, closer in spirit to the MCT of Section 4. Generalizations of DIA and alternative more elaborate approaches were introduced and examined in ref. 31, which include SCSA under the name of the renormalized vertex approximation. This incorporates screening of interactions between different fluctuating fields. We shall come back to this approximation again in Section 7.

### 5.3. Equations for Non-Ergodicity Parameters

The set of Eqs. (5.18)–(5.26) can only be analyzed via numerical mehtods reliably.[10] But we can hope to gain some insights by an approximate analysis. Here we make use of the spirit of MCT that the static quantities such as $\mathscr{G}$ are smoothly varying as we cross from ergodic to nonergodic states.[11] Thus $\mathscr{G}$, $\Pi_{\mathscr{G}}$, and $\mathscr{D}_{\mathscr{G}}$ are assumed to be given. The only unknown is $\mathscr{F}$ which is obtained by solving the following equation:

$$\mathscr{F}_{\mathbf{k}} = \mathscr{G}_{\mathbf{k}} - \frac{1}{\mathscr{G}_{\mathbf{k}}^{-1} - 2\int_{\mathbf{q}} \mathscr{D}_{\mathscr{F}\mathbf{q}} \mathscr{F}_{\mathbf{k}-\mathbf{q}}} \tag{5.29}$$

After rearranging this and defining the wave-number dependent non-ergodicity parameter by $f_{\mathbf{k}} \equiv \mathscr{F}_{\mathbf{k}} / \mathscr{G}_{\mathbf{k}}$ the above equation becomes, with $\mathscr{D}_{\mathscr{F}}$, (5.24), now expressed in terms of $f_{\mathbf{k}}$ defined above,

$$\frac{f_{\mathbf{k}}}{1-f_{\mathbf{k}}} = 2\mathscr{G}_{\mathbf{k}} \int_{\mathbf{q}} \frac{\mathscr{D}^2_{\mathscr{G}\mathbf{q}} \int_{\mathbf{p}} \mathscr{G}_{\mathbf{p}} \mathscr{G}_{\mathbf{q}-\mathbf{p}} f_{\mathbf{p}} f_{\mathbf{q}-\mathbf{p}}}{1 - \mathscr{D}_{\mathscr{G}\mathbf{q}} \int_{\mathbf{p}} \mathscr{G}_{\mathbf{p}} \mathscr{G}_{\mathbf{q}-\mathbf{p}} f_{\mathbf{p}} f_{\mathbf{q}-\mathbf{p}}} \mathscr{G}_{\mathbf{k}-\mathbf{q}} f_{\mathbf{k}-\mathbf{q}} \tag{5.30}$$

In case when $f_{\mathbf{k}}$ is small, this corresponds to the MCT-like equation (4.19) where $\mathscr{M}$ is now cubic in the $f$'s. Such equation would follow if one applies the method of Kirkpatrick and Thirumalai.(32) In fact they obtained equations with $\mathscr{M}$ quadratic in $f$ for a cubic Hamiltonian. They obtained the same equation dynamically by using MCT-like approach for a simple relaxational type model we mentioned before. Then they comment that glassy behavior should be independent of the dynamics used. However, this is not generally true as we have shown for the case of MCT driven by reversible MC mechanism. In fact the two corresponding equations (5.30) and the earlier one, (4.19), are totally different though the Hamiltonian (free-energy functional) are the same.

Perhaps it is important to point out that the difference cannot solely be attributed to dissipative and non-dissipative nature of nonlinearities

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[10] One promising numerical approach can be based on the fact that in (5.18)–(5.26) the determinations of $\mathscr{G}_{\mathbf{k}}$ and $\mathscr{F}_{\mathbf{k}}$ are separated. Namely, for given $u_2$ and $U_{\mathbf{k}}$ we can solve (5.18), (5.20), (5.22), (5.23), and (5.25) to find $\mathscr{G}_{\mathbf{k}}$. Then, with this knowledge we can solve (5.19), (5.21), (5.24), and (5.26) to obtain $\mathscr{F}_{\mathbf{k}}$. However, we can reverse the first part of this computation. We first make a reasonable guess for $\mathscr{G}_{\mathbf{k}}$ which leads to desired results for $\mathscr{F}_{\mathbf{k}}$. We can then use (5.18) and (5.16) with $g = 0$ to deduce $u_2 + U_{\mathbf{k}}$. It is only necessary that the resulting $u_2 + U_{\mathbf{k}}$ is such that it has a negative minimum at some finite value of $k$ and $U(\mathbf{r})$ is such as to prevent global instability of the system. An example of such an analysis is given in ref. 47.

[11] This will not be rigorously true for model systems with long-range forces such as that described in ref. 8.

in the two cases. A crucial point seems to be whether the ergodic-to-nonergodic transition is driven by a non-trivial Hamiltonian or by purely kinetic effects or by both. See the next section for further discussions. The replica approach as presented here takes into account only the properties of Hamiltonian. On the other hand, elsewhere we have derived purely dissipative dynamical density functional equation from a Langevin or Fokker–Planck equation type model with reversible mode coupling.(38) This equation contains non-trivial kinetic effects in addition to the non-trivial Hamiltonian (free energy density functional). We note in passing that the same dynamical density functional equation can be derived from the Smoluchowski equation for colloidal systems without hydrodynamic interactions.(38) In order to reproduce from this equation the long-time form of the self-consistent MCT equation for the density correlator, (4.8) and (4.9), we needed both nontrivial kinetic effects and nontrivial free energy function(al)s as we have emphasized in ref. 39 and the point will be also discussed in the next section.

Finally we note that both (4.19) and (5.30) possess multiple solutions including $f_{\mathbf{k}} = 0$ which corresponds to a supercooled liquid state. For the case of MCT it is shown that the state with the largest $f_{\mathbf{k}}$ is always reached.(1) For (5.30) no such property is found so far.

## 6. MECHANISMS OF NON-ERGODICITY

In contrast to more conventional phase trasition problems, widely different ideas about the so-called glass transitions appear to coexist. Consequently, more than one mehacnisms of glass transitions or ergodic-nonergodic transitions have been proposed. Here we look at some of these proposals.

### 6.1. Three Types of Mechanisms

In general three distinct types of mechanisms driving ergodic-nonergodic transitons have been proposed as enumerated below.

1. Those driven by non-trivial Hamiltonians (or free-energy function(al)s). Most spin glass type models(33) belong to this type.
2. Those driven by purely kinetic effects but with trivial Hamiltonians[12] (e.g., facilitated kinetic Ising models reviewed in ref. 42)
3. Those driven by combination of nontrivial Hamiltonians and kinetic effects (e.g., the MCT for liquids(1))

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[12] Here trivial Hamiltonians are meant typically to be those without interaction such as spin models with independent spins in an external magnetic field or an ideal gas system.

The cases (1) and (2) are well-documented and need no further comments. We here argue that the mode coupling theories of supercooled liquids$^{(1)}$ belong to the case (3). Now, in this type of theories, the body force acting on a fluid element at $\mathbf{r}$, which is nonlinear in density fluctuations and can produce cage effects, plays the central role. In its simplest form the body force appears in the following overdamped equation of motion for density fluctuations obtained after adiabaticlly eliminating the velocity field:$^{(38)}$

$$\frac{\partial}{\partial t}\rho(\mathbf{r},t) = -L\nabla\cdot\mathbf{f}(\mathbf{r},t) = L\nabla\cdot\rho(\mathbf{r})\,\nabla\frac{\delta H(\{\rho\})}{\delta\rho(\mathbf{r})} \tag{6.1}$$

where $L$ is an Onsager kinetic coefficient and $H(\{\rho\})$ is the free energy density fucntional. The expression for the body force density denoted as $\mathbf{f}(\mathbf{r})$ is given by

$$\mathbf{f}(\mathbf{r}) = -\rho(\mathbf{r})\,\nabla\frac{\delta H(\{\rho\})}{\delta\rho(\mathbf{r})} \tag{6.2}$$

Typically the free energy density fucntional takes the following form:$^{(34)}$

$$H(\{\rho\}) = H_0(\{\rho\}) + H_I(\{\rho\}) \tag{6.3}$$

$$H_0(\{\rho\}) \equiv T\int d\mathbf{r}\,\rho(\mathbf{r})\ln\left(\frac{\rho(\mathbf{r})\,\rho(\mathbf{r})}{\rho_0}\right) \tag{6.4}$$

ideal gas part, highly nonlinear in $\delta\rho$

$$H_I(\{\rho\}) \equiv \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\,\rho(\mathbf{r})\,\rho(\mathbf{r}') \tag{6.5}$$

interaction part, quadratic in $\delta\rho$

The usual choice for the interaction $V(\mathbf{r})$ is $-TC(\mathbf{r})$, $C(\mathbf{r})$ being the direct correlation function.$^{(21,34)}$ This will lead to the MCT results of Section 4.$^{(1)}$ For derivation see, e.g., ref. 20.

The linear and nonlinear contributions to the body force are:

$$\mathbf{f}_l(\mathbf{r}) = -T\nabla\,\delta\rho(\mathbf{r}) - \rho_0\nabla\int d\underline{\mathbf{r}}V(\mathbf{r}-\underline{\mathbf{r}})\,\delta\rho(\underline{\mathbf{r}}) \tag{6.6}$$

linear *due to the presence of an extra* $\rho$ *factor in front of* $\mathbf{f}(\mathbf{r})$ in (6.2)

$$\mathbf{f}_{nl}(\mathbf{r}) = -\delta\rho(\mathbf{r})\,\nabla\int d\underline{\mathbf{r}}V(\mathbf{r}-\underline{\mathbf{r}})\,\delta\rho(\underline{\mathbf{r}}) \tag{6.7}$$

nonlinear *due to the presence of an extra* $\rho$ *factor in front of* $\mathbf{f}(\mathbf{r})$ in (6.2)

The well-known cage effect(1) comes from the nonlinear body force (6.7) which combines the interaction part of the free eenrgy functional and the kinetic effect in the fom of an extra factor of $\delta\rho(\mathbf{r}, t)$.

It should be emphasized that the situation is totally different for the conventional TDGL type models like:

$$\frac{\partial}{\partial t}\rho(\mathbf{r}, t) = L'\nabla^2 \frac{\delta H(\{\rho(t)\})}{\delta\rho(\mathbf{r}, t)}$$

$$\frac{\partial}{\partial t}\rho(\mathbf{r}, t) = -L'' \frac{\delta H(\{\rho(t)\})}{\delta\rho(\mathbf{r}, t)}$$

where driving mechanisms are entirely contained in the non-trivial Hamiltonians, or free nergy function(al)s $H(\{\rho\})$.

*Understanding the relationship between these different types of mechanisms is a problem of utmost importance.*

One nagging question is that how much (free-)energetic considerations alone can go towards full understanding of glassy behavior, in particular, meaning of the often quoted mode coupling critical temperature.(43)

A possible first step toward addressing this question is to look at dynamical extensions of SCSA.(45, 46) These models are of purely relaxational type driven by non-trivial Hamiltonians. If we follow Kirkpatrick and Thirumalai(32) and if extention of these equations or preferably their replicated version[13] to non-ergodic regions is possible, we expect to recover (5.30) for the nonergodicity parameter. An attempt toward this direction will be given in Section 7. Then we can start to consider including non-trivial kinetics. In this endeaver it might be useful to know that SCSA can be cast into a form of MCT approximation if besides the usual order parameter field, say, $\phi(\mathbf{r})$, we take $\phi(\mathbf{r})^2$ as an independent composite field(44) and formally treat both on equal footings.(45, 46)

## 6.2. Relationship Between van der Waals and Stripe Glasses

We expect that our model with appropriate choice of $U(\mathbf{r})$ possesses non-ergodic states and hence rich "energy landscape" as it is with the stripe

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[13] In this connection it is useful to remember the situation in spin glasses.(33) The dynamical treatment of spin glass was introduced first to avoid somewhat artificial nature of the old replica treatment which requires one to take the number of replicated sytems to zero in the end. This became the standard tool to deal with spin glass and other problems with quenched randomness. De Domonicis(35) then noted that if the problem is formulated dynamically using the MSR method,(36) averages over quenched randomness can be performed without relying on the artificial old replica trick. Then, in discussing the influence of an initial condition on out-of-equilibiurm dynamics, it became necessary to incorporate the replica trick into dynamical theory again.(37)

glass.$^{(7)}$ We now examine the relationship between the two glass models explicitly.

Let Fourier transform of $U(\mathbf{r})$ be $U_k = U_{\mathbf{k}}$ (real quantity depending only on $k$ for isotropic interaction) and assume that it has a negative minimum $-U_m$ at some value of wavenumber $k_m$.$^{(8)}$ If $\min_k(u_2 + U_k) < 0$ for some values of $k$ around $k_m$, that is,

$$u_2 - U_m < 0 \tag{6.8}$$

the system described by (3.18) is linearly unstable against small periodic density fluctuations having Fourier components $\phi_{\mathbf{k}}$ with $k$ inside the region of negative $u_2 + U_k$. In this sense,

$$u_2 = u_2^s \equiv U_m \tag{6.9}$$

represents a mean field spinodal. Let us expand $U_k$ near the minimum up to second order as

$$U_k = -U_m + b(k - k_m)^2 \tag{6.10}$$

where $b$ is a positive coefficient. The "bare" static density-density correlation function $\mathscr{G}_{0\mathbf{k}}$ in this neighborhood is then

$$\mathscr{G}_{0\mathbf{k}} = \frac{1}{u_2 + U_k} = \frac{1}{b} \frac{1}{\xi^{-2} + (k - k_m)^2} \tag{6.11}$$

where

$$\xi^{-2} \equiv \frac{u_2 - u_2^s}{b} \qquad \text{for} \quad u_2 \geqslant u_2^s \tag{6.12}$$

On the other hand, for $u_2 \leqslant u_2^s$, $\mathscr{G}_{0\mathbf{k}}$ can become negative, that is, we enter the region inside the mean field spinodal, and then we have to use the Hartree approximation result

$$\mathscr{G}_{H\mathbf{k}} = \frac{1}{b} \frac{1}{\xi_H^{-2} + (k - k_m)^2} \tag{6.13}$$

with

$$\xi_H^{-2} \equiv \frac{u_2 - u_2^s + u_4 \int_{\mathbf{q}} \mathscr{G}_{H\mathbf{q}}}{b} \tag{6.14}$$

Here $\mathscr{G}_{H\mathbf{q}}$ is the self-consistently determined equal time density correlation function and the r.h.s. of (6.14) is now expected to be positive in some region inside the mean field spinodal of crystalline-liquid transition.We may assume an upper cut-off for $q$ in order to avoid any undesirable divergence.

In this form our model is now very close to that of stripe glass.(7) In that case existence of two characteristic length scales were essential to obtain a glassy state. Here we also have such two length scales near the mean field spinodal. One is the length $l_m \equiv \frac{2\pi}{k_m}$ associated with the minimum of $U_k$. Another is $\xi = [b/(u_2 - u_2^s)]^{1/2}$ or $\xi_H = [b/(u_2 - u_2^s + u_4 \int_{\mathbf{q}} \mathscr{G}_{H\mathbf{q}})]^{1/2}]$, the latter including the Hartree correction term. Thus here proximity to the mean field spinodal is an important factor for emergence of of glassy phase in our system. We then should be able to map the analyses of ref. 7 to our model to discuss glassy behavior, which in fact was indicated in Section 5.3.

Now, Klein and coworkers' findings(8) mentioned in Section 4.3 suggests emergence of frozen inhomogeneous density profiles in our model system as well if quenched into states inside the mean field spinodal of crystalline-liquid transition. Indeed, very recently Loh and his coworkers(47) succeeded in numerically solving the SCSA equation (5.30) corresponding to Klein's model with a slight modification and found a finite non-ergodicity paramenter and hence a finite configurational entropy for a certain temperature region in agreement with the argument of this subsection and Klein's simulation results.(8)

## 7. REAL REPLICA APPROACH TO DYNAMICS

One motivation to apply the real replica concept to dynamics is a hope to find a new way to separate two qualitatively different types of dynamics that characterize regions below the so-called mode coupling critical temperature. One is the processes occurring in states within individual domains of attraction of free energy landscape. Another is slower processes that involve barrier crossings between different minima. If two replicated sytems are interacting with each other by weak attractions, these sytems will likely to remain in the same domain of attraction where physical quantities averged over replicated systems will be slowly varying compared to quantities which represent fluctuations from averages. In fact some recent replica calculations envisage formation of "molecules" consisting of "atoms" belonging to different replica members.(27, 40, 41) [14] The idea of considering

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[14] The last reference makes use of this concept for a field theoretical model of stripe glass.

replicated system (also called clones) in non-equilibrium problems has appeared in literature (e.g., refs. 48–51).

As a first step in this direction we describe below a formalism for dynamical replica theory where nonergodicity is driven by complex Hamiltonian or more specifically the nontrivial free energy functional of our model system. In Section 5.3 we saw the equation for the non-ergodicity parameter obtained by MCT and by static real replica approach are radically different. This is the case even if equal time correlations in the two equations are taken to be the same and the nonergodicity parameter is assumed to be small. If we recall the widely recognized fact that the underlying MCT is considered to be valid only in ergodic states, this seems to require a dynamical theory that can also deal with complex free energy landscape that characterizes non-ergodic states. Combining the real replica idea with the SCSA successfully implemented in refs. 7, 26, 27 thus motivated us to develop its dynamical version in order to deal with dynamics involving nonergodic states. Generalizing this approach to include kinetic effects is left for future.

We will mainly use a generating functional formalism for non-equilibrium problems. This has formal similarity to equilibrium problems,^(45) which facilitates use of replica theory.

## 7.1. Separation into Slow and Fast Processes

Here we start from the following Langevin equation for $M$ replicated systems, each replica being labelled by symbols $a$, $b$,...:

$$\frac{\partial}{\partial t}\phi^a(\mathbf{r},t) = -L\left[u_2\phi^a(\mathbf{r},t)+\int d\mathbf{r}'\,U(r-r')\,\phi^a(\mathbf{r}',t)-\frac{g}{\beta}\Phi(\mathbf{r},t)+u_4\phi^a(\mathbf{r},t)^3\right]+R^a_\phi(\mathbf{r},t) \tag{7.1}$$

$$\langle R^a_\phi(\mathbf{r},t)\,R^b_\phi(\mathbf{r}',t')\rangle = 2LT\delta_{ab}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t') \tag{7.2}$$

where

$$\Phi \equiv \frac{1}{M}\sum_{a=1}^{M}\phi^a \tag{7.3}$$

The term in $[\cdots]$ in (7.1) comes from $\delta F(\{\boldsymbol{\phi}\};M)/\delta\phi^a(\mathbf{r})$ with $F(\{\boldsymbol{\phi}\};M)$ given by (5.17) where $\boldsymbol{\phi}$ stands for the entire set $\phi^a$, $a=1,2,\ldots,M$. Here

$\Phi(\mathbf{r}, t)$, an average over weakly interacting replicated systems, will be slowly varying compared with fluctuations from the average in nonergodic states. In ref. 41 $\Phi(\mathbf{r}, t)$ is taken to represent a frozen field in a nonergodic state.

We now write

$$\phi^a(\mathbf{r}, t) = \Phi(\mathbf{r}, t) + \varphi^a(\mathbf{r}, t) \tag{7.4}$$

with

$$\sum_{a=1}^{M} \varphi^a(\mathbf{r}, t) = 0 \tag{7.5}$$

Sustituting (7.4) and (7.5) into (7.1) we find

$$\begin{aligned} \frac{\partial}{\partial t} \Phi(\mathbf{r}, t) = & -L\left[\left(u_2 - \frac{g}{\beta}\right) \Phi(\mathbf{r}, t) + \int d\mathbf{r}'\, U(r-r')\, \Phi(\mathbf{r}', t)\right] \\ & - L u_4 \left(\Phi(\mathbf{r}, t)^3 + 3\Phi(\mathbf{r}, t) \frac{1}{M} \sum_{a=1}^{M} \varphi^a(\mathbf{r}, t)^2 + \frac{1}{M} \sum_{a=1}^{M} \varphi^a(\mathbf{r}, t)^3\right) \\ & + R_\Phi(\mathbf{r}, t) \end{aligned} \tag{7.6}$$

$$\begin{aligned} \frac{\partial}{\partial t} \varphi^a(\mathbf{r}, t) = & -L\left[u_2 \varphi^a(\mathbf{r}, t) + \int d\mathbf{r}'\, U(r-r')\, \varphi^a(\mathbf{r}', t)\right] \\ & - L u_4 \left[3\Phi(\mathbf{r}, t) \left(\varphi^a(\mathbf{r}, t)^2 - \frac{1}{M} \sum_{b=1}^{M} \varphi^b(\mathbf{r}, t)^2\right)\right. \\ & \left. + 3\Phi(\mathbf{r}, t)^2 \varphi^a(\mathbf{r}, t) + \varphi^a(\mathbf{r}, t)^3 - \frac{1}{M} \sum_{b=1}^{M} \varphi^b(\mathbf{r}, t)^3\right] + R_\varphi^a(\mathbf{r}, t) \end{aligned} \tag{7.7}$$

where thermal noises are defined by

$$R_\phi^a(\mathbf{r}, t) = R_\Phi(\mathbf{r}, t) + R_\varphi^a(\mathbf{r}, t), \qquad R_\Phi(\mathbf{r}, t) \equiv \frac{1}{M} \sum_{a=1}^{M} R_\phi^a(\mathbf{r}, t) \tag{7.8}$$

Therefore the fluctuation-dissipation relation (7.2) expressed in the new variables takes the following forms:

$$\langle R_\Phi(\mathbf{r}, t)\, R_\Phi(\mathbf{r}', t')\rangle = \frac{2LT}{M}\, \delta(\mathbf{r}-\mathbf{r}')\, \delta(t-t') \tag{7.9}$$

$$\langle R_\varphi^a(\mathbf{r}, t)\, R_\varphi^b(\mathbf{r}', t')\rangle = 2LT \left(\delta_{ab} - \frac{1}{M}\right) \delta(\mathbf{r}-\mathbf{r}')\, \delta(t-t') \tag{7.10}$$

$$\langle R_\varphi^a(\mathbf{r}, t)\, R_\Phi(\mathbf{r}', t')\rangle = 0 \tag{7.11}$$

Let us now write the MSR action integral[(36)] as[15]

$$
\begin{aligned}
&\mathscr{S}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M)\\
&\quad \equiv \sum_a \int d\mathbf{r}\, dt\, i\hat{\phi}^a(\mathbf{r}, t) \left[ \dot{\phi}^a(\mathbf{r}, t) + L \frac{\delta F(\{\boldsymbol{\phi}(t)\}; M)}{\delta \phi^a(\mathbf{r}, t)} + LTi\hat{\phi}^a(\mathbf{r}, t) \right]\\
&\quad = \sum_a \int d\mathbf{r}\, dt\, i\hat{\phi}^a(\mathbf{r}, t) \left[ \dot{\phi}^a(\mathbf{r}, t) + L \left( u_2 \phi^a(\mathbf{r}, t) + \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \phi^a(\mathbf{r}', t) \right.\right.\\
&\qquad \left.\left. + u_4 \phi^a(\mathbf{r}, t)^3 - \frac{g}{\beta} \Phi(\mathbf{r}, t) \right) + LTi\hat{\phi}^a(\mathbf{r}, t) \right]
\end{aligned}
\tag{7.12}
$$

where $\hat{\phi}^a$ is a field conjugate to $\phi^a$. In the operator representation, $\hat{\phi}^a(\mathbf{r})$ behaves like $-\delta/\delta\phi^a(\mathbf{r})$.[(36)] Substituting (7.4) and (7.5) the action integral becomes, denoting partial time derivative by an overdot,

$$
\begin{aligned}
&\mathscr{S}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M)\\
&\quad = \sum_a \int d\mathbf{r}\, dt\, i\hat{\phi}^a(\mathbf{r}, t) \left\{ \dot{\Phi}(\mathbf{r}, t) + L \left[ \left( u_2 - \frac{g}{\beta} \right) \Phi(\mathbf{r}, t) + \int d\mathbf{r}'\, U(r-r')\, \Phi(\mathbf{r}', t) \right.\right.\\
&\qquad \left. + u_4 \left( \Phi(\mathbf{r}, t)^3 + 3\Phi(\mathbf{r}, t) \frac{1}{M} \sum_{a=1}^{M} \varphi^a(\mathbf{r}, t)^2 + \frac{1}{M} \sum_{a=1}^{M} \varphi^a(\mathbf{r}, t)^3 \right) \right]\\
&\qquad + \dot{\varphi}^a(\mathbf{r}, t) + L \left[ u_2 \varphi^a(\mathbf{r}, t) + \int d\mathbf{r}'\, U(r-r')\, \varphi^a(\mathbf{r}', t) \right.\\
&\qquad + u_4 \left( 3\Phi(\mathbf{r}, t) \left( \varphi^a(\mathbf{r}, t)^2 - \frac{1}{M} \sum_{b=1}^{M} \varphi^b(\mathbf{r}, t)^2 \right) \right.\\
&\qquad \left.\left.\left. + 3\Phi(\mathbf{r}, t)^2\, \varphi^a(\mathbf{r}, t) + \varphi^a(\mathbf{r}, t)^3 - \frac{1}{M} \sum_{b=1}^{M} \varphi^b(\mathbf{r}, t)^3 \right) \right] + LTi\hat{\phi}^a(\mathbf{r}, t) \right\}
\end{aligned}
\tag{7.13}
$$

Since a hat variable plays the role of a functional differentiation, we find

$$
\begin{aligned}
\hat{\Phi} &= \sum_a \hat{\phi}^a\\
\hat{\phi}^a &= \hat{\varphi}^a + \frac{1}{M} \hat{\Phi} - \frac{1}{M} \sum_b \hat{\varphi}^b
\end{aligned}
\tag{7.14}
$$

---

[15] Here and after $[\phi]$ etc. denote functional dependence on $\phi(\mathbf{r}, t)$, etc. and $\{\phi\}$ etc. denote functional dependence on $\phi(\mathbf{r})$ etc. without $t$.

These are not enough to determine the $\hat{\varphi}$'s uniquely. Hence we can require

$$\sum_a \hat{\varphi}^a = 0 \tag{7.15}$$

and put

$$\hat{\phi}^a = \hat{\varphi}^a + \frac{1}{M} \hat{\Phi} \tag{7.16}$$

We then find, for instance,

$$\sum_a i\hat{\phi}^a i\hat{\phi}^a = \frac{1}{M} i\hat{\Phi} i\hat{\Phi} + \sum_a i\hat{\varphi}^a i\hat{\varphi}^a \tag{7.17}$$

With these the action becomes

$$\mathscr{S}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) = \mathscr{S}_{\hat{\Phi}\Phi}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) + \mathscr{S}_{\hat{\varphi}\varphi}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) \tag{7.18}$$

where

$$\begin{aligned}
&\mathscr{S}_{\hat{\Phi}\Phi}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) \\
&\quad \equiv \int d\mathbf{r}\, dt \left\{ i\hat{\Phi}(\mathbf{r}, t)\, \dot{\Phi}(\mathbf{r}, t) + \frac{LT}{M}\, i\hat{\Phi}(\mathbf{r}, t)\, \hat{\Phi}(\mathbf{r}, t) \right. \\
&\quad + i\hat{\Phi}(\mathbf{r}, t)\, L \left[ \left( u_2 - \frac{g}{\beta} \right) \Phi(\mathbf{r}, t) + \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}')\, \Phi(\mathbf{r}', t) \right. \\
&\quad \left.\left. + u_4 \left( \Phi(\mathbf{r}, t)^3 + 3\Phi(\mathbf{r}, t) \frac{1}{M} \sum_a \varphi^a(\mathbf{r}, t)^2 + \frac{1}{M} \sum_a \varphi^a(\mathbf{r}, t)^3 \right) \right] \right\}
\end{aligned} \tag{7.19}$$

and

$$\begin{aligned}
\mathscr{S}_{\hat{\varphi}\varphi}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) \equiv \sum_a \int d\mathbf{r}\, dt\, i\hat{\varphi}^a(\mathbf{r}, t) \{ &\dot{\varphi}^a(\mathbf{r}, t) + L[u_2 \varphi^a(\mathbf{r}, t) \\
&+ \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}')\, \varphi^a(\mathbf{r}', t) + 3u_4 \Phi(\mathbf{r}, t)^2\, \varphi^a(\mathbf{r}, t) \\
&+ u_4 (3\Phi(\mathbf{r}, t)\, \varphi^a(\mathbf{r}, t)^2 + \varphi^a(\mathbf{r}, t)^3)] + LTi\hat{\varphi}^a(\mathbf{r}, t) \}
\end{aligned} \tag{7.20}$$

### 7.1.1. Elimination

Next we eliminate the variables $\varphi$ and $\hat{\varphi}$ to focus on dynamics of $\Phi$. This can best be done by integrating $\exp \mathscr{S}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M)$ out as $\int d[\hat{\varphi}]\, d[\varphi]\, \delta[\sum_{a=1}^{M} \varphi^a]\, \delta[\sum_{a=1}^{M} \hat{\varphi}^a] \cdots$. We can do this by retaining up to bilinear terms in $\varphi$, $\hat{\varphi}$. This is based on our expectation that the system will tend to a non-ergodic state characterzied by $\Phi(\mathbf{r}, t)$ and the amplitude of $\varphi$ will remain small. In this approximation MSR action including terms associated with the external fields $h^a, \hat{h}^a$ and Lagrangian multiplier terms resulting from Fourier representations of $\delta[\sum_{a=1}^{M} \varphi^a]\, \delta[\sum_{a=1}^{M} \hat{\varphi}^a]$ becomes

$$\begin{aligned} &\mathscr{S}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) + \sum_a \int d\mathbf{r}\, dt [h^a(\mathbf{r}, t)\, \phi^a(\mathbf{r}, t) + \hat{h}^a(\mathbf{r}, t)\, \hat{\phi}^a(\mathbf{r}, t) \\ &\qquad + i(\lambda(\mathbf{r}, t)\, \varphi^a(\mathbf{r}, t) + \hat{\lambda}(\mathbf{r}, t)\, \hat{\varphi}^a(\mathbf{r}, t))] \\ &\quad \cong \tilde{\mathscr{S}}_0([\Phi], [\hat{\Phi}]; [H][\hat{H}], M) + \tilde{\mathscr{S}}_2([\boldsymbol{\varphi}], [\hat{\boldsymbol{\varphi}}]; [\underline{\mathbf{h}}][\underline{\hat{\mathbf{h}}}][\lambda][\hat{\lambda}], M) \end{aligned} \tag{7.21}$$

where

$$\begin{gathered} \sum_a (h^a \phi^a + \hat{h}^a \hat{\phi}^a) = H\Phi + \hat{H}\hat{\Phi} + \sum_a (\underline{h}^a \varphi^a + \underline{\hat{h}}^a \hat{\varphi}^a) \\ H \equiv \sum_a h(\mathbf{r}, t)^a, \qquad \hat{H} \equiv \frac{1}{M} \sum_a \hat{h}^a \\ \underline{h}^a \equiv h_a - \frac{1}{M} \sum_b h_b, \qquad \underline{\hat{h}}^a \equiv \hat{h}_a - \frac{1}{M} \sum_b \hat{h}_b \end{gathered} \tag{7.22}$$

We give expressions for $\tilde{\mathscr{S}}_0$ and $\tilde{\mathscr{S}}_2$, the zeroth order and the sum of the first and second order terms in $\varphi$, $\hat{\varphi}$, respectively, as

$$\begin{aligned} &\tilde{\mathscr{S}}_0([\Phi], [\hat{\Phi}]; [H][\hat{H}], M) \\ &\quad \equiv \int d\mathbf{r}\, dt \left\{ i\hat{\Phi}(\mathbf{r}, t)\, \dot{\Phi}(\mathbf{r}, t) + \frac{LT}{M}\, i\hat{\Phi}(\mathbf{r}, t)\, i\hat{\Phi}(\mathbf{r}, t) \right. \\ &\qquad \left. + i\hat{\Phi}(\mathbf{r}, t)\, L \left[ \left( u_2 - \frac{g}{\beta} \right) \Phi(\mathbf{r}, t) + \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}')\, \Phi(\mathbf{r}', t) + u_4 \Phi(\mathbf{r}, t)^3 \right] \right\} \\ &\qquad + \int d\mathbf{r}\, dt (H(\mathbf{r}, t)\, \Phi(\mathbf{r}, t) + \hat{H}(\mathbf{r}, t)\, \hat{\Phi}(\mathbf{r}, t)) \end{aligned} \tag{7.23}$$

$$\begin{aligned} &\tilde{\mathscr{S}}_2([\boldsymbol{\varphi}], [\hat{\boldsymbol{\varphi}}]; [\Phi][\hat{\Phi}][\underline{\mathbf{h}}][\underline{\hat{\mathbf{h}}}][\lambda][\hat{\lambda}], M) \\ &\quad \equiv \sum_a \int d\mathbf{r}\, dt \left\{ -LT \hat{\varphi}^a(\mathbf{r}, t)^2 + \hat{\varphi}^a(\mathbf{r}, t)\, i\hat{\Lambda}^a(\mathbf{r}, t) \right. \\ &\qquad \left. + \frac{3Lu_4}{M}\, i\hat{\Phi}(\mathbf{r}, t)\, \Phi(\mathbf{r}, t)\, \varphi^a(\mathbf{r}, t)^2 + (i\lambda(\mathbf{r}, t) + \underline{h}^a(\mathbf{r}, t))\, \varphi^a(\mathbf{r}, t) \right\} \end{aligned} \tag{7.24}$$

where

$$\hat{\Lambda}^a(\mathbf{r}, t) \equiv D_t \varphi^a(\mathbf{r}, t) + \hat{\lambda}(\mathbf{r}, t) - i\underline{\hat{h}}^a(\mathbf{r}, t) \tag{7.25}$$

$$D_t(\mathbf{r}) \equiv \partial_t + L(u_2 + \hat{U}(\mathbf{r}) + 3u_4 \Phi(\mathbf{r}, t)^2) \tag{7.26}$$

$$\hat{U}(\mathbf{r})\, A(\mathbf{r}) \equiv \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}')\, A(\mathbf{r}') \tag{7.27}$$

$D_t, \hat{U}$ being both operators and $A$ an arbitrary function of space. We observe in passing that $D_t$ can be regarded as a symmetric matrix in the space of $\mathbf{r}$ and replica indices, which can be also written as

$$D_t = \mathbf{1}\partial_t + L\mathscr{H}(\{\Phi\}) \tag{7.28}$$

with $\mathbf{1}$ an identity matrix and $\mathscr{H}(\{\Phi\})$ the Hessian matrix of (5.17) with $M \to \infty$ and $\varphi = 0$.

We then integrate fuctionally over $\hat{\varphi}$ and find for $\tilde{\mathscr{S}}_\varphi$ defined by

$$e^{\tilde{\mathscr{S}}_\varphi} \equiv \int d[\hat{\varphi}]\, e^{\tilde{\mathscr{S}}_2} \tag{7.29}$$

the following:

$$\begin{aligned} \tilde{\mathscr{S}}_\varphi = \sum_a \int d\mathbf{r}\, dt \Bigg\{ & -\frac{1}{4LT} \hat{\Lambda}^a(\mathbf{r}, t)^2 + \frac{3Lu_4}{M} i\hat{\Phi}(\mathbf{r}, t)\, \Phi(\mathbf{r}, t)\, \varphi^a(\mathbf{r}, t)^2 \\ & + (i\lambda(\mathbf{r}, t) + \underline{h}^a(\mathbf{r}, t))\, \varphi^a(\mathbf{r}, t) \Bigg\} \end{aligned} \tag{7.30}$$

The effective action integral for $\Phi$, $\hat{\Phi}$ is given by

$$\begin{aligned} \tilde{\mathscr{S}}_{\text{eff}}([\Phi][\hat{\Phi}]; [\underline{\mathbf{h}}][\underline{\hat{\mathbf{h}}}], M) \equiv {} & \tilde{\mathscr{S}}_0([\Phi][\hat{\Phi}]; [H][\hat{H}], M) \\ & + \Delta\tilde{\mathscr{S}}_{\text{eff}}([\Phi][\hat{\Phi}]; [H][\hat{H}], M) \end{aligned} \tag{7.31}$$

$$e^{\Delta\tilde{\mathscr{S}}_{\text{eff}}} \equiv \int d[\lambda]\, d[\hat{\lambda}]\, e^{\Delta\tilde{\mathscr{S}}_{\lambda\hat{\lambda}}} \tag{7.32}$$

$$e^{\Delta\tilde{\mathscr{S}}_{\lambda\hat{\lambda}}} \equiv \int d[\varphi]\, e^{\tilde{\mathscr{S}}_\varphi} \tag{7.33}$$

In order to carry out integration with respect to $[\varphi]$ we rewrite (7.30) as follows:

$$\tilde{\mathscr{S}}_\varphi = \sum_a \int d\mathbf{r}\, dt \{ -\tfrac{1}{2} \varphi^a \cdot \mathscr{Q} \cdot \varphi^a - \mathscr{P}^a \cdot \varphi^a - \mathscr{T}^a \} \tag{7.34}$$

where $\mathscr{Q}$ and $\mathscr{P}$ are operators defined by (7.35) and (7.36), respectively, below and $\mathscr{T}$ is a quantity defined by (7.37) below. Center dots imply scalar products when $\mathscr{Q}$, $P$, and $\mathscr{T}$ are viewed as matrices which naturally involve space-time integrations. Here

$$\mathscr{Q} \equiv \frac{1}{2LT} D_t^\dagger D_t - \frac{6Lu_4}{M} i\hat{\Phi}\Phi \tag{7.35}$$

$$\mathscr{P}^a \equiv \frac{1}{2LT} (-i\hat{\underline{h}}^a + \hat{\lambda})\, D_t - \underline{h}^a - i\lambda \tag{7.36}$$

$$\mathscr{T}^a \equiv \frac{1}{4LT} (-i\hat{\underline{h}}^a + \hat{\lambda})^2 \tag{7.37}$$

Also $X^\dagger$ of any operator $X$ denotes an adjoint of $X$, but no complex conjugation. Therefore $D_t^\dagger = -\partial_t + L(u_2 + \hat{U} + 3u_4 \Phi(\mathbf{r}, t)^2)$ since $\hat{U}$ is self-adjoint. So is $\mathscr{Q}$ self-adjoint as well. With these notations we have, after carrying out integration with respect to $[\varphi]$ with its convergence assumed[16]

$$\Delta\tilde{\mathscr{S}}_{\lambda\hat{\lambda}} = \sum_a \int d\mathbf{r}\, dt \left\{ \frac{1}{2} \mathscr{P}^a \cdot \mathscr{Q}^{-1} \cdot \mathscr{P}^{a\dagger} - \mathscr{T}^a \right\} - \frac{M}{2} \ln \det \mathscr{Q} \tag{7.38}$$

where the last term is needed since $\mathscr{Q}$ depends on $\Phi$, $\hat{\Phi}$.

We are now left with performing $\int d[\lambda]\, d[\hat{\lambda}]$. To do this we write out (7.38) noting $\sum_a \underline{h}^a = \sum_a \hat{\underline{h}}^a = 0$. That is,

$$\begin{aligned} \Delta\tilde{\mathscr{S}}_{\lambda\hat{\lambda}} = {} & M \int d\mathbf{r}\, dt \left\{ -\frac{1}{4LT} \hat{\lambda}(\mathbf{r}, t)^2 + \frac{1}{2} \hat{\lambda} \cdot \mathscr{L} \cdot \hat{\lambda} + \frac{1}{2} \hat{\lambda} \cdot \mathscr{M} \cdot \lambda \right. \\ & \left. + \frac{1}{2} \lambda \cdot \mathscr{M}^\dagger \cdot \hat{\lambda} + \frac{1}{2} \lambda \cdot \mathscr{Q}^{-1} \cdot \lambda \right\} \\ & + \sum_a \int d\mathbf{r}\, dt \left\{ \frac{1}{4LT} \hat{\underline{h}}^a(\mathbf{r}, t)^2 - \frac{1}{2} \hat{\underline{h}}^a \cdot \mathscr{L} \cdot \hat{\underline{h}}^a - \frac{1}{2} \hat{\underline{h}}^a \cdot \mathscr{M} \cdot \underline{h}^a - \frac{1}{2} \underline{h}^a \cdot \mathscr{M}^\dagger \cdot \hat{\underline{h}}^a \right. \\ & \left. - \frac{1}{2} \underline{h}^a \cdot \mathscr{Q}^{-1} \cdot \underline{h}^a \right\} - \frac{M}{2} \ln \det \mathscr{Q} \end{aligned} \tag{7.39}$$

where

$$\mathscr{L} \equiv \frac{1}{(2LT)^2} D_t \cdot \mathscr{Q}^{-1} \cdot D_t^\dagger, \qquad \mathscr{M} \equiv -\frac{1}{2LT} D_t \cdot \mathscr{Q}^{-1} \tag{7.40}$$

---

[16] If this Gaussian integral and the one below are not convergent, the density profile $\Phi$ does not correspond to local free energy minma, and possibly contain saddle points and/or local maxima.

The integration $\int d[\lambda]\, d[\hat{\lambda}]$ involves only the first two lines of (7.39) which are written in the matrix form as

$$-\frac{1}{2} M(\lambda\ \hat{\lambda}) \cdot \Xi \cdot \begin{pmatrix} \lambda \\ \hat{\lambda} \end{pmatrix} \tag{7.41}$$

with

$$\Xi \equiv \begin{pmatrix} -\mathcal{Q}^{-1} & \mathcal{M}^{\dagger} \\ \mathcal{M} & \frac{1}{2LT}\mathbf{1} - \mathcal{L} \end{pmatrix} \tag{7.42}$$

Here $\hat{\Phi}$ enter only through $\mathcal{Q}$ and $\Phi$ through $\mathcal{Q}$ and $D_t$. Performing Gaussian integrations whose convergence is assumed again, we finally find

$$\Delta\tilde{\mathscr{S}}_{\text{eff}}([\Phi][\hat{\Phi}]; [\underline{\mathbf{h}}][\hat{\underline{\mathbf{h}}}], M)$$

$$= -\frac{M}{2} \ln \det \mathcal{Q} - \frac{1}{2} \ln \det \Xi + \sum_a \int d\mathbf{r}\, dt \left\{ \frac{1}{4LT} \underline{\hat{h}}^a(\mathbf{r}, t)^2 - \frac{1}{2} \underline{\hat{h}}^a \cdot \mathcal{L} \cdot \underline{\hat{h}}^a \right.$$

$$\left. - \frac{1}{2} \underline{\hat{h}}^a \cdot \mathcal{M} \cdot \underline{h}^a - \frac{1}{2} \underline{h}^a \cdot \mathcal{M}^{\dagger} \cdot \underline{\hat{h}}^a - \frac{1}{2} \underline{h}^a \cdot \mathcal{Q}^{-1} \cdot \underline{h}^a \right\} \tag{7.43}$$

Substitution of the above result into the second term of (7.31) completes the elimination of $\varphi$, $\hat{\varphi}$. Now, in this last form the causality is not evident. This apparent violation of causality comes from elimination of $\varphi$, $\hat{\varphi}$ over the entire time domain $(-\infty, \infty)$.

### *7.1.2. Special Case $M \to \infty$*

We consider the following limiting processes taken in the order indicated:

(1) the thermodynamic limit, (2) $M \to \infty$, and (3) $g \to 0$.

Here the degrees of freedom associated with $\Phi$ representing averages over $M$ replicas will literally freeze into the non-ergodic phase. Then the degrees of freedom associated with $\varphi$ represent rapid motion in the background of the frozen spatial heterogeneity.

Note $M$ enters the final result only through $\mathcal{Q}$. The limiting form for $\mathcal{Q}$ denoted as $\mathcal{Q}_{\infty}$ is

$$\mathcal{Q}_{\infty} = \frac{1}{2LT} D_t^{\dagger} D_t \tag{7.44}$$

which is independent of $\hat{\Phi}$. In view of the factor $M$ in front of the first term on r.h.s. of (7.43), we need to expand this term in $1/M$ and retain the first two terms. Thus putting

$$\begin{aligned} \mathscr{Q} &= \mathscr{Q}_\infty + \frac{1}{M}\mathscr{Q}_1 \\ \mathscr{Q}_1 &\equiv -6Lu_4 i\hat{\Phi}\Phi \end{aligned} \tag{7.45}$$

we find

$$\begin{aligned} \ln\det\mathscr{Q} &= \ln\det\mathscr{Q}_\infty + \ln\det\left(\mathbf{1} + \frac{1}{M}\mathscr{Q}_\infty^{-1}\cdot\mathscr{Q}_1\right) \\ &= \ln\det\mathscr{Q}_\infty + \frac{1}{M}\operatorname{Tr}\mathscr{Q}_\infty^{-1}\cdot\mathscr{Q}_1 + O(1/M^2) \end{aligned} \tag{7.46}$$

Here Tr actually implies $\int d\mathbf{r}\,dt$. Therefore, in this limit, we get for the first term on the r.h.s. of the action (7.43),

$$-\frac{M}{2}\ln\det\mathscr{Q} = -\frac{M}{2}\ln\det\mathscr{Q}_\infty - \frac{1}{2}\operatorname{Tr}\mathscr{Q}_\infty^{-1}\cdot\mathscr{Q}_1 + O(1/M) \tag{7.47}$$

Then the limit $M \to \infty$ suggests minimizing $\ln\det\mathscr{Q}_\infty$ with respect to $\Phi$, whose meaning is unclear at the moment.

Further results in this limit are, with suffices $\infty$ attached,

$$\mathscr{L}_\infty = \frac{\mathbf{1}}{2LT} \tag{7.48}$$

$$\mathscr{M}_\infty = (D_t^\dagger)^{-1}, \qquad \mathscr{M}_\infty^\dagger = D_t^{-1} \tag{7.49}$$

$$\Xi_\infty = \begin{pmatrix} -2LTD_t^{-1}(D_t^\dagger)^{-1} & D_t^{-1} \\ (D_t^\dagger)^{-1} & 0 \end{pmatrix} \tag{7.50}$$

## 7.2. SCSA Applied to Dynamics

Here we develop a formulation which uses an auxilliary field $\sigma$ which, in a sense behaves as $\phi^2$, to deal with the nonlinearity in (7.1)[17]

---

[17] A possible extenstion of this is to generalize $\sigma(\mathbf{r}) = \phi(\mathbf{r})^2$ into $\phi(\mathbf{r}_1)\,\phi(\mathbf{r}_2)$ with $|\mathbf{r}_1 - \mathbf{r}_2|$ of the order of $l$ or smaller. Then, writing $\mathbf{r}_1 = \mathbf{R} + \frac{1}{2}\mathbf{r}$ and $\mathbf{r}_2 = \mathbf{R} - \frac{1}{2}\mathbf{r}$ and expanding the product in $\mathbf{r}$, we obtain the expression of the form

$$\phi(\mathbf{r}_1)\,\phi(\mathbf{r}_2) = \sum_{lm}\sigma_{lm}(\mathbf{R}, |\mathbf{r}|)\,Q_{lm}(\hat{\mathbf{r}})$$

where $Q_{lm}(\hat{\mathbf{r}})$ with $\hat{\mathbf{r}} \equiv \mathbf{r}/|r|$ is the bond orientation parameter.(52) Here the $\sigma_{lm}$'s play the role of new copmposite fields.

MCA applied to this case is known to reproduce SCSA results.(45, 46)

The starting Langevin equations of motion for $M$ set of replicated variables $\phi^a$ and $\sigma^a$ with , $a = 1, 2,\dots, M$ are

$$\frac{\partial}{\partial t}\phi^a(\mathbf{r}, t) = -L\frac{\delta\hat{F}(\{\boldsymbol{\phi}(t)\}, \{\boldsymbol{\sigma}(t)\}; M)}{\delta\phi^a(\mathbf{r}, t)} + R_\phi^a(\mathbf{r}, t) \tag{7.51}$$

$$\frac{\partial}{\partial t}\sigma^a(\mathbf{r}, t) = -L_\sigma\frac{\delta\hat{F}(\{\boldsymbol{\phi}(t)\}, \{\boldsymbol{\sigma}(t)\}; M)}{\delta\sigma^a(\mathbf{r}, t)} + R_\sigma^a(\mathbf{r}, t) \tag{7.52}$$

with the following averages over noises:

$$\begin{aligned}\langle R_\phi^a(\mathbf{r}, t)\rangle = \langle R_\sigma^a(\mathbf{r}, t)\rangle = \langle R_\phi^a(\mathbf{r}, t)\, R_\sigma^b(\mathbf{r}', t')\rangle = 0\\ \langle R_\phi^a(\mathbf{r}, t)\, R_\phi^b(\mathbf{r}', t')\rangle = 2LT\delta_{ab}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\\ \langle R_\sigma^a(\mathbf{r}, t)\, R_\sigma^b(\mathbf{r}', t')\rangle = 2L_\sigma T\delta_{ab}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\end{aligned} \tag{7.53}$$

The free energy functional of the replicated system is similar to that of Section 5 except that now auxilliary fields $\boldsymbol{\sigma}$ are included.(46) That is,

$$\hat{F}(\{\boldsymbol{\phi}\}, \{\boldsymbol{\sigma}\}; M) = F_2(\{\boldsymbol{\phi}\}; M) + \sum_a \int d\mathbf{r}\left(\tfrac{1}{4}\sigma^a(\mathbf{r})^2 + \tfrac{1}{2}i\sqrt{u_4}\,\sigma^a(\mathbf{r})\,\phi^a(\mathbf{r})^2\right) \tag{7.54}$$

where

$$\begin{aligned}F_2(\{\boldsymbol{\phi}\}; M) \equiv \sum_a \int d\mathbf{r}\,\frac{u_2}{2}\,[\phi^a(\mathbf{r})]^2 + \sum_a \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\,U(\mathbf{r}-\mathbf{r}')\,\phi^a(\mathbf{r})\,\phi^a(\mathbf{r}')\\ -\frac{g}{2M\beta}\sum_{a,b}\int d\mathbf{r}\,\phi^a(\mathbf{r})\,\phi^b(\mathbf{r})\end{aligned} \tag{7.55}$$

Here $g(>0)$ is the weak field that pulls together a pair of replicated systems together so that they will evolve eventually in the same free energy minimum in nonergodic states. This plays a role similar to the symmetry-breaking field in an ordinary phase transition. We further remark a possiblity to make $g(>0)$ time-dependent to encompass larger classes of problems that can be dealt with by this formalism. For instance one can switch $g(t)$ on and off so that two replicated systems try to evolve into the same non-ergodic component for certain time periods.

In the above we have assumed that the inter-replica interactions arise solely from the free energy functional. However, one can think of interactions of other origins although we do not explore such possibilities at this time. In ref. 51 which deals with quantum glasses $g$ enters the quantum action integral in quite a different manner[18]

The MSR action is

$$\hat{\mathscr{S}}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}], [\boldsymbol{\sigma}], [\hat{\boldsymbol{\sigma}}]; M) = \mathscr{S}_2([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) + \Delta\hat{\mathscr{S}}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}], [\boldsymbol{\sigma}], [\hat{\boldsymbol{\sigma}}]; M) \tag{7.57}$$

with

$$\begin{aligned} \mathscr{S}_2([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}]; M) &\equiv \sum_a \int d\mathbf{r}\, dt\, i\hat{\phi}^a(\mathbf{r}, t) \Bigg[ \dot{\phi}^a(\mathbf{r}, t) \\ &\quad + L \frac{\delta F_2(\{\boldsymbol{\phi}(t)\}; M)}{\delta \phi^a(\mathbf{r}, t)} + LTi\hat{\phi}^a(\mathbf{r}, t) \Bigg] \\ &= \sum_a \int d\mathbf{r}\, dt\, i\hat{\phi}^a(\mathbf{r}, t) \Bigg[ \dot{\phi}^a(\mathbf{r}, t) + L \Bigg( u_2 \phi^a(\mathbf{r}, t) \\ &\quad + \int d\mathbf{r}\, U(\mathbf{r} - \mathbf{r}')\, \phi^a(\mathbf{r}', t) - \frac{g}{\beta} \Phi(\mathbf{r}, t) \Bigg) \\ &\quad + LTi\hat{\phi}^a(\mathbf{r}, t) \Bigg] \end{aligned} \tag{7.58}$$

$$\Delta\hat{\mathscr{S}}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}], [\boldsymbol{\sigma}], [\hat{\boldsymbol{\sigma}}]; M) \equiv \sum_a \Delta\hat{\mathscr{S}}([\phi^a], [\hat{\phi}^a], [\sigma^a], [\hat{\sigma}^a]) \tag{7.59}$$

$$\begin{aligned} \Delta\hat{\mathscr{S}}([\phi^a], [\hat{\phi}^a], [\sigma^a], [\hat{\sigma}^a]) &\equiv \int d\mathbf{r}\, dt \{ \hat{\phi}^a(\mathbf{r}, t)\, Li \sqrt{u_4}\, \sigma^a(\mathbf{r}, t)\, \phi^a(\mathbf{r}, t) \\ &\quad + i\hat{\sigma}^a(\mathbf{r}, t)\, L_\sigma [\tfrac{1}{2} (\sigma^a(\mathbf{r}, t) + i \sqrt{u_4}\, \phi^a(\mathbf{r}, t)^2) \\ &\quad + iT\hat{\sigma}^a(\mathbf{r}, t)]\} \end{aligned} \tag{7.60}$$

Here we have chosen $L_\sigma$ large enough that the l.h.s. of (7.52) can be dropped.$^{(53)}$

### 7.3. Schwinger–Dyson Equation

Derivation here follows the standard method described, for instance, in ref. 54, and is not limited to the zero (or constant) values of the field

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[18] It enters in the following form in the action integral appearing in quantum-mechanical calculation:

$$-g \sum_{ab} \int d\mathbf{r}\, dt\, dt'\, \phi^a(\mathbf{r}, t)\, \phi^b(\mathbf{r}, t') \tag{7.56}$$

variables. Thus it is applicable to the out-of-equilibrium problems as well. Note $\langle \sigma \rangle \neq 0$ even in equilibrium.

We first make use of the following identity:

$$
\begin{aligned}
&e^{-W^M([\mathbf{h}_\phi], [\hat{\mathbf{h}}_\phi], [\mathbf{h}_\sigma], [\hat{\mathbf{h}}_\sigma])} \int d[\boldsymbol{\phi}]\, d[\hat{\boldsymbol{\phi}}]\, d[\boldsymbol{\sigma}]\, d[\hat{\boldsymbol{\sigma}}]\, \frac{\delta}{\delta x_\alpha(\mathbf{r}, t)} \\
&\quad \times e^{\hat{\mathscr{S}}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}], [\boldsymbol{\sigma}], [\hat{\boldsymbol{\sigma}}]; M)} e^{i \int d\mathbf{r}'\, dt' (\mathbf{h}_\phi \cdot \boldsymbol{\phi} + \hat{\mathbf{h}}_\phi \cdot \hat{\boldsymbol{\phi}} + \mathbf{h}_\sigma \cdot \boldsymbol{\sigma} + \hat{\mathbf{h}}_\sigma \cdot \hat{\boldsymbol{\sigma}})} \\
&\quad \times e^{-\beta \hat{F}(\{\boldsymbol{\phi}(0)\}, \{\boldsymbol{\sigma}(0)\}; M)} = 0 \qquad (7.61)
\end{aligned}
$$

where

$$
\begin{aligned}
e^{W^M([\mathbf{h}_\phi], [\hat{\mathbf{h}}_\phi], [\mathbf{h}_\sigma], [\hat{\mathbf{h}}_\sigma])} &\equiv \int d[\boldsymbol{\phi}]\, d[\hat{\boldsymbol{\phi}}]\, d[\boldsymbol{\sigma}]\, d[\hat{\boldsymbol{\sigma}}]\, e^{\hat{\mathscr{S}}([\boldsymbol{\phi}], [\hat{\boldsymbol{\phi}}], [\boldsymbol{\sigma}], [\hat{\boldsymbol{\sigma}}]; M)} \\
&\quad \times e^{i \int d\mathbf{r}\, dt (\mathbf{h}_\phi \cdot \boldsymbol{\phi} + \hat{\mathbf{h}}_\phi \cdot \hat{\boldsymbol{\phi}} + \mathbf{h}_\sigma \cdot \boldsymbol{\sigma} + \hat{\mathbf{h}}_\sigma \cdot \hat{\boldsymbol{\sigma}})} e^{-\beta \hat{F}(\{\boldsymbol{\phi}(0)\}, \{\boldsymbol{\sigma}(0)\}; M)} \qquad (7.62)
\end{aligned}
$$

(7.61) and (7.62) incorporate the initial equilibrium condition at the time zero. For $t > 0$ this can be written as

$$
\left\langle \frac{\delta \hat{\mathscr{S}}}{\delta x_\alpha(\mathbf{r}, t)} + h_\alpha(\mathbf{r}, t) \right\rangle = 0 \qquad (7.63)
$$

Or,

$$
\left\langle \frac{\delta \hat{\mathscr{S}}}{\delta x_\alpha(\mathbf{r}, t)} \right\rangle = -h_\alpha(\mathbf{r}, t) \qquad (7.64)
$$

Using (7.57)–(7.60) we can find explicit forms of (7.64) as follows:

$$
\begin{aligned}
&-\langle \dot{\hat{\phi}}^a(\mathbf{r}, t) \rangle + u_2 \langle \hat{\phi}^a(\mathbf{r}, t) \rangle + \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}') \langle \hat{\phi}^a(\mathbf{r}', t) \rangle \\
&\qquad - \frac{g}{2\beta M} \langle \hat{\Phi}(\mathbf{r}, t) \rangle + iL \sqrt{u_4}\, \langle \sigma^a(\mathbf{r}, t) \rangle \\
&\qquad - L_\sigma \sqrt{u_4}\, [\mathscr{R}^{aa}_{\phi\sigma}(\mathbf{r}, t; \mathbf{r}, t) + \langle \hat{\sigma}^a(\mathbf{r}, t) > \phi^a(\mathbf{r}', t') \rangle] \\
&\quad = -h^a_\phi(\mathbf{r}, t) \qquad (7.65)
\end{aligned}
$$

$$
\begin{aligned}
&\langle \dot{\phi}^a(\mathbf{r}, t) \rangle + L \left[ u_2 \langle \phi^a(\mathbf{r}, t) \rangle + \int d\mathbf{r}'\, U(\mathbf{r} - \mathbf{r}') \langle \phi^a(\mathbf{r}', t) \rangle - \frac{g}{2\beta} \langle \Phi(\mathbf{r}, t) \rangle \right] \\
&\qquad - L \sqrt{u_4}\, [\mathscr{C}^{aa}_{\sigma\phi}(\mathbf{r}, t; \mathbf{r}, t) + \langle \sigma^a(\mathbf{r}, t) \rangle \langle \phi^a(\mathbf{r}, t) \rangle] = -\hat{h}^a_\phi(\mathbf{r}, t) \qquad (7.66)
\end{aligned}
$$

$$-L\sqrt{u_4}\,\hat{[}\mathscr{R}^{aa}_{\phi\phi}(\mathbf{r},t;\mathbf{r},t)+\langle\phi^a(\mathbf{r},t)\rangle\langle\hat{\phi}^a(\mathbf{r},t)\rangle]+\frac{L_\sigma}{2}\,i\langle\hat{\sigma}^a(\mathbf{r},t)\rangle=-h^a_\sigma(\mathbf{r},t) \tag{7.67}$$

$$iL_\sigma\left[\frac{1}{2}\langle\sigma^a(\mathbf{r},t)\rangle+\frac{i}{2}\sqrt{u_4}\,(\mathscr{C}^{aa}_{\phi\phi}(\mathbf{r},t;\mathbf{r},t)+\langle\phi^a(\mathbf{r},t)\rangle^2)+2iT\langle\hat{\sigma}^a(\mathbf{r},t)\rangle\right]=-\hat{h}^a_\sigma(\mathbf{r},t) \tag{7.68}$$

where we have introduced the response functions $\mathscr{R}$ and the correlation functions $\mathscr{C}$ by

$$\mathscr{R}^{ab}_{\phi\sigma}(\mathbf{r},t;\mathbf{r}',t')\equiv\langle\phi^a(\mathbf{r},t)\,\hat{\sigma}^b(\mathbf{r}',t')\rangle-\langle\phi^a(\mathbf{r},t)\rangle\langle\hat{\sigma}^b(\mathbf{r}',t')\rangle \tag{7.69}$$

$$\mathscr{C}^{ab}_{\sigma\phi}(\mathbf{r},t;\mathbf{r}',t')\equiv\langle\sigma^a(\mathbf{r},t)\,\phi^a(\mathbf{r},t)\rangle-\langle\sigma^a(\mathbf{r},t)\rangle\langle\phi^a(\mathbf{r}',t')\rangle \tag{7.70}$$

In order to proceed to derivation of the SD equation, we use a compact notation: let $x_\alpha(\mathbf{r},t)$ denote all the variables of the replicated systems, $\phi^a(\mathbf{r},t), \hat{\phi}^a(\mathbf{r},t), \sigma^a(\mathbf{r},t), \hat{\sigma}^a(\mathbf{r},t)$, $a=1,2,\ldots,M$. We then write $\mathbf{x}\equiv\{x_\alpha\}$ and the fields conjugate to $\mathbf{x}$ are denoted as $\mathbf{h}\equiv\{h_\alpha\}$. We first introduce the Legendre transform from $W([\mathbf{h}])$ to $\Gamma([\mathbf{x}])$ by

$$\Gamma([\mathbf{x}])=W([\mathbf{h}])-\mathbf{h}\cdot\mathbf{x} \tag{7.71}$$

where we suppressed space-time integration of the second term, and **h, x** are related by

$$x_\alpha(\mathbf{r},t)=\frac{\delta W([\mathbf{h}])}{\delta h_\alpha(\mathbf{r},t)},\qquad h_\alpha(\mathbf{r},t)=-\frac{\delta\Gamma([\mathbf{x}])}{\delta x_\alpha(\mathbf{r},t)} \tag{7.72}$$

The last member above is nothing but the general form of the averaged equations of motion obtained in (7.65)–(7.68) where average signs in $\langle\phi^a(\mathbf{r},t)\rangle$ etc. are suppressed.

In the following the inverse propagator and the vertex function play major roles which are formally defined, respectively, as (space-time coordinates are suppressed for simplicity),

$$\Gamma_{\alpha\beta}\equiv\frac{\delta h_\alpha}{\delta x_\beta}=-\frac{\delta^2\Gamma}{\delta x_\alpha\,\delta x_\beta},\qquad \Gamma_{\alpha\beta\gamma}\equiv-\frac{\delta^3\Gamma}{\delta x_\alpha\,\delta x_\beta\,\delta x_\gamma} \tag{7.73}$$

Here the propagator $\mathscr{G}_{\alpha\beta}$ (that is, response and correlation functions) is defined by

$$\mathscr{G}_{\alpha\beta}\equiv\frac{\delta^2 W([\mathbf{h}])}{\delta h_\alpha\,\delta h_\beta} \tag{7.74}$$

We naturally have $\Gamma_{\alpha\beta} = (\mathscr{G}^{-1})_{\alpha\beta}$ or

$$\mathscr{G}_{\alpha\gamma}\Gamma_{\gamma\beta} = \delta_{\alpha\beta} \tag{7.75}$$

where summation convention over repeated indices is implied. Taking the functional derivative of the above with respect to $x_\delta$ and noting $\delta\Gamma_{\gamma\delta}/\delta x_\beta = \Gamma_{\delta\gamma\beta}$ we find

$$\frac{\delta\mathscr{G}_{\alpha\gamma}}{\delta x_\delta} = -\mathscr{G}_{\alpha\beta}\Gamma_{\beta\epsilon\delta}\mathscr{G}_{\epsilon\gamma} \tag{7.76}$$

Here, to be more concrete, we introduce a cubic generating functional where again space-time integrals are suppressed:

$$\hat{\mathscr{S}}([\mathbf{x}]) = \gamma_\alpha x_\alpha + \frac{1}{2}\gamma_{\alpha\beta}x_\alpha x_\beta + \frac{1}{3!}\gamma_{\alpha\beta\gamma}x_\alpha x_\beta x_\gamma \tag{7.77}$$

were $x_\alpha$ etc. are not averaged values. This gives

$$\frac{\delta\hat{\mathscr{S}}([\mathbf{x}])}{\delta x_\alpha} = \gamma_\alpha + \gamma_{\alpha\beta}x_\beta + \frac{1}{2}\gamma_{\alpha\beta\gamma}x_\beta x_\gamma \tag{7.78}$$

We then take an average of the above to get (where average signs are again omitted), noting (7.64) and $-h_\alpha = \delta\Gamma/\delta x_\alpha$

$$\gamma_\alpha + \gamma_{\alpha\beta}x_\beta + \frac{1}{2}\gamma_{\alpha\beta\gamma}[\mathscr{G}_{\beta\gamma} + x_\beta x_\gamma] = \frac{\delta\Gamma}{\delta x_\alpha} \tag{7.79}$$

This is the explicit form of the averaged equation of motion. Now, the SD equation is obtained by taking functional derivative of the above with repect to $x_\beta$ as

$$\gamma_{\alpha\beta} + \frac{1}{2}\gamma_{\alpha\delta\gamma}\frac{\delta\mathscr{G}_{\delta\gamma}}{\delta x_\beta} + \gamma_{\alpha\beta\gamma}x_\gamma = -(\mathscr{G}^{-1})_{\alpha\beta} \tag{7.80}$$

This becomes using (7.76) as

$$\gamma_{\alpha\beta} - \tfrac{1}{2}\gamma_{\alpha\delta\gamma}\mathscr{G}_{\delta\epsilon}\Gamma_{\epsilon\alpha\beta}\mathscr{G}_{\alpha\gamma} + \gamma_{\alpha\beta\gamma}x_\gamma = -(\mathscr{G}^{-1})_{\alpha\beta} \tag{7.81}$$

This is again transformed into a more familiar Schwinger–Dyson form:

$$(\mathscr{G}^{-1})_{\alpha\beta} = (\mathscr{G}_0^{-1})_{\alpha\beta} + \Sigma_{\alpha\beta} \tag{7.82}$$

Now, there are two ways to define the two terms appearing on the r.h.s. of the above equation:

$$\begin{aligned}(\mathscr{G}_0^{-1})_{\alpha\beta} &\equiv -\gamma_{\alpha\beta} - \gamma_{\alpha\beta\gamma} x_\gamma \\ \Sigma_{\alpha\beta} &\equiv \tfrac{1}{2}\gamma_{\alpha\delta\gamma}\mathscr{G}_{\delta\epsilon}\mathscr{G}_{\mu\gamma}\Gamma_{\epsilon\mu\beta}\end{aligned} \tag{7.83}$$

$$\begin{aligned}(\mathscr{G}_0^{-1})_{\alpha\beta} &\equiv -\gamma_{\alpha\beta} \\ \Sigma_{\alpha\beta} &\equiv -\gamma_{\alpha\beta\gamma} x_\gamma + \tfrac{1}{2}\gamma_{\alpha\delta\gamma}\mathscr{G}_{\delta\epsilon}\mathscr{G}_{\mu\gamma}\Gamma_{\epsilon\mu\beta}\end{aligned} \tag{7.84}$$

The former is the way adopted by field theorists,(54) whereas the latter is the way chosen by condensed matter physicists.(7)

In the following we adopt the second choice. Note that $-\gamma_{\alpha\beta\gamma}x_\gamma$ appearing in the above gives rise to the Hartree term corresponding to (5.22) of static calculation.

If we note from (7.73), (7.84), and (7.82) that

$$\Gamma_{\alpha\beta\gamma} = \frac{\delta\Sigma_{\alpha\beta}}{\delta x_\gamma} \tag{7.85}$$

we get a functionally closed set of equations (7.82), (7.84), and (7.85).

Now, (7.84) and (7.85) give

$$\Gamma_{\alpha\beta\gamma} = -\gamma_{\alpha\beta\gamma} + \frac{\delta\, \Delta\Sigma_{\alpha\beta}}{\delta x_\gamma} \tag{7.86}$$

with

$$\Delta\Sigma_{\alpha\beta} \equiv \tfrac{1}{2}\gamma_{\alpha\delta\gamma}\mathscr{G}_{\delta\epsilon}\mathscr{G}_{\mu\gamma}\Gamma_{\epsilon\mu\beta} \tag{7.87}$$

A popular approximation scheme known under different names (DIA, MCA, BVA, etc.)[19] neglects the second term of (7.86). That is,

$$\Gamma_{\alpha\beta\gamma} \approx -\gamma_{\alpha\beta\gamma} \tag{7.88}$$

The error to this approximation is estimated as $O(n^{-1})$, $n$ being the number of components of the order parameter.(54) In this approximation we find the following self-consistent closed equation determining $\mathscr{G}$.

$$(\mathscr{G}^{-1})_{\alpha\beta} = -\gamma_{\alpha\beta} - \gamma_{\alpha\beta\gamma}x_\gamma + \tfrac{1}{2}\gamma_{\alpha\delta\gamma}\mathscr{G}_{\delta\epsilon}\mathscr{G}_{\mu\gamma}\gamma_{\epsilon\mu\beta} = (\mathscr{G}_0^{-1})_{\alpha\beta} + \Sigma_{\alpha\beta} \tag{7.89}$$

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[19] These abbreviate Direct Interaction Approximation,(30) Mode Coupling Approximation,(45) Bare Vertex Approximation,(54) respectively. Note that these namings originally refer to the cases when **x** do not contain the $\sigma$'s and the $\hat{\sigma}$'s.

Or

$$\mathscr{G}_{\alpha\beta} = \mathscr{G}_{0\alpha\beta} - \mathscr{G}_{0\alpha\gamma} \Sigma_{\gamma\delta} \mathscr{G}_{\delta\beta} \tag{7.90}$$

In our case this turns out to be the SCSA approximation. We give some technical details in applying the general results of this section to our problem in Appendix A.

## 8. MICROSCOPIC DERIVATION

In this section we attempt a microscopic derivation of our model system. In equilibrium problems there are many studies of the effects of long range forces on reference systems with short range interactions.(25, 55, 56) Our starting point is the following Liouville equation for the phase space distribution function $\hat{D}(\hat{x}, t)$ where $\hat{x}$ is the abbreviation for the entire set of the phase space variables. For particle systems these are the particle position vectors $\mathbf{r}_\mathrm{i}$ and momenta $\mathbf{p}_\mathrm{i}$ with $i = 1, 2,\ldots, N$, with $N$ the total number of particles, which are also denoted as $3N$-component vectors $\mathbf{r}^N$ and $\mathbf{p}^N$:

$$\frac{\partial}{\partial t} \hat{D}(\hat{x}, t) = \hat{\mathscr{L}}(\hat{x})\, \hat{D}(\hat{x}, t) \tag{8.1}$$

where the Liouville operator is given by

$$\hat{\mathscr{L}}(\hat{x}) = \hat{\mathscr{L}}_0(\hat{x}) + \hat{\mathscr{L}}_U(\hat{x}) \tag{8.2}$$

$$\hat{\mathscr{L}}_U(\hat{x}) \equiv \frac{\partial}{\partial \mathbf{p}^N} \cdot \mathbf{\nabla}^N \hat{U}^N(\hat{x}) \tag{8.3}$$

and $\mathscr{L}_0(\hat{x})$ is the Liouville operator for the reference system which need not be explicitly specified. Here $\hat{U}^N(\hat{x})$ is the total potential energy coming from long-range interaction and is given by

$$\hat{U}^N(\hat{x}) \equiv \sum_{j>l} U(\mathbf{r}_j - \mathbf{r}_l) \tag{8.4}$$

Our task is then to reduce this Liouville equation into those containing relevant hydrodynamic variables. Thus we introduce the reduced distribution function. Let us suppose that on a certain coarse-grained level time evolution of the system can be well described by a set of gross variables. The most familiar examples of the gross variables are the variables that enter the usual hydrodynamic equations which are dictated basically by conservation laws.$^{(6)}$ But, in reality the choice of the gross variables is far from clear-cut, especially for glassy sytems. We denote a set of these variables by $\{a\}$. This can be a discrete set, or a continuous set of variables like field variables. In many cases we can find expressions for them in terms of the phase space variables, which we denote by $\{\hat{a}(\hat{x})\}$. We can then express the (micro-canonical) reduced distribution function (or functional) for $\{a\}$ by

$$D(\{a\}, t) \equiv \int d\hat{x}\, \delta\{a-\hat{a}(\hat{x})\}\, \hat{D}(\hat{x}, t) \tag{8.5}$$

where $\delta\{a-a'\}$ expresses a product of delta functions for each of the set $\{a\}$ or what may be called a "delta functional." This has the following property for any functional $f(\{a\})$:

$$\int d\{a'\}\, f(\{a'\})\, \delta\{a-a'\} = f(\{a\}) \tag{8.6}$$

where the integral is the functional integral over the set $\{a'\}$.

The problem is to obtain an equation for $D(\{a\}, t)$. We may then write

$$\frac{\partial}{\partial t} D(\{a\}, t) = \mathscr{L}^{\mathrm{ref}} D(\{a\}, t) + \int d\hat{x} [\hat{\mathscr{L}}_U^{N\dagger}(\hat{x})\, \delta\{a-\hat{a}(\hat{x})\}]\, \hat{D}(\hat{x}, t) \tag{8.7}$$

where $\mathscr{L}^{\mathrm{ref}}$ is a Fokker–Planck type operator that takes care of time evolution of the reference fluid which is assumed to be known, and $\hat{\mathscr{L}}_U^{N\dagger}$ is the adjoint of $\hat{\mathscr{L}}_U^N$.

Now the momentum variables $\mathbf{p}^N$ are contained in the microscopic expression for the momentum density variable $\hat{\mathbf{j}}(\mathbf{r})$ and in that for the energy density variable $\hat{e}(\mathbf{r})$. These (hatted) variables are expressed as

$$\hat{\mathbf{j}}(\mathbf{r}; \hat{x}) \equiv \sum_j \mathbf{p}_j\, \delta(\mathbf{r}-\mathbf{r}_j) \tag{8.8}$$

$$\hat{e}(\mathbf{r}; \hat{x}) \equiv \sum_j \frac{p_j^2}{2m}\, \delta(\mathbf{r}-\mathbf{r}_j) + \cdots \tag{8.9}$$

where $\cdots$ represent terms coming from potential energies which do not contain momenta and will not be needed for the moment. The molecular kinetic energy exhibited in the second equation above goes partly to the thermal energy and partly to the kinetic energy of macroscopic motion of fluid elements.

We are particularly interested in the following expression:

$$[\hat{\mathscr{L}}_U^{N\dagger}(\hat{x})\,\delta\{a-\hat{a}(\hat{x})\}]=\int d\mathbf{r}\,\frac{\delta}{\delta a(\mathbf{r})}\,(\nabla^N\hat{U}^N)\cdot\frac{\partial\hat{a}(\mathbf{r};\hat{x})}{\partial\mathbf{p}^N}\,\delta\{a-\hat{a}(\hat{x})\} \tag{8.10}$$

where we assumed the gross variables to be a set of field variables $\{a(\mathbf{r})\}$ whose molecular expressions are $\{\hat{a}(\mathbf{r};\hat{x})\}$. We now work out $(\nabla^N\hat{U}^N)\cdot(\partial\hat{a}(\mathbf{r};\hat{x}))/\partial\mathbf{p}^N\}$ for the momentum and energy densities:

$$(\nabla^N\hat{U}^N)\cdot\frac{\partial\hat{\mathbf{j}}(\mathbf{r};\hat{x})}{\partial\mathbf{p}^N}=\sum_{jl}\delta(\mathbf{r}-\mathbf{r}_j)\,\frac{\partial U(\mathbf{r}_j-\mathbf{r}_l)}{\partial\mathbf{r}_j}$$

$$=\int d\mathbf{r}'\,[\nabla U(\mathbf{r}-\mathbf{r}')]\,\hat{\rho}(\mathbf{r};\hat{x})\,\hat{\rho}(\mathbf{r}';\hat{x}) \tag{8.11}$$

$$(\nabla^N\hat{U}^N)\cdot\frac{\partial\hat{e}(\mathbf{r};\hat{x})}{\partial\mathbf{p}^N}=\int d\mathbf{r}'\,\frac{\hat{\mathbf{j}}(\mathbf{r};\hat{x})}{m}\cdot[\nabla U(\mathbf{r}-\mathbf{r}')]\,\hat{\rho}(\mathbf{r}';\hat{x}) \tag{8.12}$$

We see that the r.h.s. are expressed solely in terms of hatted gross varaibles. In view of the delta functionals multiplied to the right in (8.7) and (8.10) these hats can be removed. Then we see that these extra terms arising from long-range force can appear as the following additional terms to the equation of motion for the momentum and energy densites:

$$\frac{\partial}{\partial t}\mathbf{j}(\mathbf{r},t)=\cdots-\rho(\mathbf{r},t)\int d\mathbf{r}'\,\nabla U(\mathbf{r}-\mathbf{r}')\,\rho(\mathbf{r}',t) \tag{8.13}$$

$$\frac{\partial}{\partial t}e(\mathbf{r},t)=\cdots-\frac{\mathbf{j}(\mathbf{r},t)}{m}\cdot\int d\mathbf{r}'\,\nabla U(\mathbf{r}-\mathbf{r}')\,\rho(\mathbf{r}',t) \tag{8.14}$$

where ellipsis stand for the terms in the absenc of long range forces. (8.13) give just the additional term of the model equation (2.4).

Since our model equations, in particular, (2.5), were expressed in terms of specific entropy $s$ instead of the energy density, we need to convert the variables in (8.14) using the thermodynamic relation as

$$\rho T\,\delta s=-\frac{1}{\rho}\,h\,\delta\rho+\delta e_I \tag{8.15}$$

where $e_I$ is the local internal energy density and $h\equiv e_I+p$ is the local enthalpy density with $p$ the local pressure. Thus $e$ is written as (arguments $\mathbf{r}$ and $t$ are often suppressed),

$$e=e_K+e_I \qquad \text{with} \quad e_K\equiv\frac{\mathbf{j}^2}{2m\rho}, \quad \text{(macroscopic kinetic energy)} \tag{8.16}$$

Now, we need to consider contributions of long range forces to this local enthalpy density. So we go back to (3.5) and (3.6). These equations give for the contribution to the total enthalpy $H = E + PV$ the value $V\rho_0^2 U_0$. We can infer from this result that this is translated into the additional local enthalpy density $h(\mathbf{r})$ as follows

$$\rho(\mathbf{r}) \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}')$$

Then we can find the equation for $s$ as

$$\begin{aligned} \rho(\mathbf{r}, t)\, T(\mathbf{r}, t) \frac{\partial}{\partial t} s(\mathbf{r}, t) &= -\frac{1}{\rho(\mathbf{r}, t)} h(\mathbf{r}, t) \frac{\partial}{\partial t} \rho(\mathbf{r}, t) + \frac{\partial}{\partial t} e_I(\mathbf{r}, t) \\ &= \frac{1}{\rho(\mathbf{r}, t)} h(\mathbf{r}, t)\, \nabla \cdot \frac{\mathbf{j}(\mathbf{r}, t)}{m} + \frac{\partial}{\partial t} e_I(\mathbf{r}, t) \\ &= \cdots + \left(\nabla \cdot \frac{\mathbf{j}(\mathbf{r}, \mathbf{t})}{m}\right) \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}', t) + \frac{\partial}{\partial t} e_I(\mathbf{r}, t) \end{aligned} \tag{8.17}$$

where ellipsis is for the reference fluid contributions.

We still need to compute the last term $\frac{\partial}{\partial t} e_I(\mathbf{r}, t)$. For this purpose let us consider a change $\delta_U e_I(\mathbf{r}, t)$ in $e_I(\mathbf{r}, t)$ coming from the long-range force when the density at $\mathbf{r}$ is changed by $\delta\rho(\mathbf{r}, t)$:

$$\delta_U e_I(\mathbf{r}, t) = \delta\rho(\mathbf{r}, t) \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}', t) \tag{8.18}$$

where we noted that $\int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}', t)$ behaves like an external field acting at a molecule located at $\mathbf{r}$ at time $t$. This immediately gives

$$\begin{aligned} \frac{\partial}{\partial t} e_I(\mathbf{r}, t) &= \cdots + \left(\frac{\partial}{\partial t} \rho(\mathbf{r}, t)\right) \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}', t) \\ &= \cdots - \frac{\nabla \cdot \mathbf{j}(\mathbf{r}, t)}{m} \int d\mathbf{r}'\, U(\mathbf{r}-\mathbf{r}')\, \rho(\mathbf{r}', t) \end{aligned} \tag{8.19}$$

If we substitute this into (8.17) we find that the long-range force contributions cancel and we finally obtain

$$\rho(\mathbf{r}, t)\, T(\mathbf{r}, t) \frac{\partial}{\partial t} s(\mathbf{r}, t) = \cdots + (\text{no long-range force term}) \tag{8.20}$$

This result confirms our model equation for the entropy, (2.5).

## 9. CONCLUDING REMARKS

We have introduced a model which embodies the spirit of van der Waals, but permits us to deal with non-equilibrium situations as well. The difficult many body problems of dense liquids are replaced by the problems of nonlinear fields. The capacity to handle the nonequilibrium aspects is crucial for a thorough understanding of problems as complex as glassy behavior. This model was used to derive the MCT equation in a manner that shows a way to examine the uncontrolled approximations entering it.

An attempt to tackle glassy behavior in the continuum framework has recently been put forward for stripe glasses(7) as was touched upon in Sections 5 and 6. Despite apparent differences between the stripe glasses and the van der Waals glasses, a striking similarity was found as far as emergence of glassiness is concerned. In fact we argued that glassiness results from existence of two competing length scales. For our model system, a "short" length scale is the range $l$ of long-range interactions and a "long" length scale appears near the mean field spinodal of the model system between liquid and crystalline phases (not liquid-gas). For the stripe glass considered in ref. 7 glassyness is assoicated with occurrence of correlated stripe structure, where correlation length exceeds the stripe spacing[20] It is interesting that even for fluid with apparent short range interactions such as the Lennard-Jones mixture the emergence of glassiness is closely related to appearence of ramified long-lived frustrated clusters of the sort found in spinodal nucleation.(57) There also the intrinsic length scale of the ramified strucutre and the average cluster size will naturally be identified as the two length scales.

On the other hand, haziness of these criteria for emergence of glassiness seems to be due to lack of explicit kinetic considerations in these arguments because glassiness can also appear by purely kinetic reasons even for systems with trivial free energy.(4, 5, 42)

The real replica approach is very appealing in the sense that it stems from the idea of symmetry-breaking field familiar in usual phase transitions. As a first step toward better understanding of glassy dynamics we

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[20] Lou–Colonno–Romero, Yuval Clejan, Harvey Gould, and William Klein found a stripe phase for Ising models with repulsive long range interactions.(58) Lattice effects on stripe glassses are discussed in ref. 59 as well.

One can argue that the glassy behavior tends to disappear as the force range $l$ increases (J. Jäckle, private communication, and also ref. 47). Since the effects of our interest occur over length scales of the order of $l$ and longer in our model, one needs to scale up the system size by a factor proportional to $l^d$ to prevent glassy behavior from disappearing as $l$ increases.

have presented our attempt to formulate real replica treatment of nonequilibrium problems. So far our results are mainly formal. Real values of this approach in deciphering mysteries of glassiness are yet to be seen. The analogous development of real replica dynamics for our van der Waals fluid model introduced in Section 2 has to be left for future. These problems with more concrete results will require years of major endeavor.

In this work as well as in refs. 7 and 47 the SCSA approximation of Bray[(28)] was extensively applied because it appears to be the only existing approxiamtion that permits field-theoretical treatment of complex free energy landscape although the approximation is uncontrolled. Examination of this approximation in the context of our van der Waals model is an important future research subject.

After this manuscript was submitted for publication, J. Jäckle informd the author about a related work,[(60)] where a model with a Kac-type potential was used to study glassy behavior. Two comments are appropriate for this work.

1. The replica method employed in this work is of the old Edwards type where the number of replicas is taken to be zero at the end.

2. $U_k$, a Fourier transform of the Kac pothetial, was chosen to be always positive. Hence no glassy behavior was found. This is consistent with the conclusion of our work.

## APPENDIX A: SOME TECHNICAL DETAILS OF SCSA APPLIED TO REPLICATED DYNAMICS

Here we apply the general SCSA approximation scheme described in the last half of Section 7 to the model system of Section 7.2. This serves to illustrate what would be involved in implementing the general scheme of Section 7 to concrete problems. At this stage we can only treat the time-dependent Ginzburg–Landau type model of Section 7.2.

The first task is to consider a possible form of $\mathscr{G}_{\alpha\beta}$. We can then in general write as follows,

$$\mathscr{G}_{\alpha\beta} = \mathscr{G}^{ab}_{\lambda\mu}, \quad a, b = 1, 2, \ldots, M; \quad \lambda, \mu = \phi, \hat{\phi}, \sigma, \hat{\sigma} \tag{A.1}$$

Following the symmetry argument of ref. 7, we only need the following two types:

$$\begin{aligned} \mathscr{G}^{d}_{\lambda\mu}(\mathbf{r}, t; \mathbf{r}', t') &\equiv \mathscr{G}^{aa}_{\lambda\mu}(\mathbf{r}, t; \mathbf{r}', t') \\ \mathscr{G}^{f}_{\lambda\mu}(\mathbf{r}, t; \mathbf{r}', t') &\equiv \mathscr{G}^{ab}_{\lambda\mu}(\mathbf{r}, t; \mathbf{r}', t') \qquad \text{with} \quad a \neq b \end{aligned} \tag{A.2}$$

The corresponding response and correlation functions $\mathscr{R}$, $C$ can have superfixes $a=b$ or $a\neq b$. Then we can write when $\hat{\mathbf{h}}=0$,

$$\mathscr{G}^{d}_{\lambda\mu}(\mathbf{r},t;\mathbf{r}',t') = \begin{pmatrix} \mathscr{C}^{d}_{\phi\phi}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{C}^{d}_{\phi\sigma}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{R}^{d}_{\phi\phi}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{R}^{d}_{\phi\sigma}(\mathbf{r},t;\mathbf{r}',t') \\ \mathscr{C}^{d}_{\sigma\phi}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{C}^{d}_{\sigma\sigma}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{R}^{d}_{\sigma\phi}(\mathbf{r},t;\mathbf{r}',t') & \mathscr{R}^{d}_{\sigma\sigma}(\mathbf{r},t;\mathbf{r}',t') \\ \{\mathscr{R}\}^{d}_{\phi\phi}(\mathbf{r}',t';\mathbf{r},t) & \mathscr{R}^{d}_{\phi\sigma}(\mathbf{r}',t';\mathbf{r},t) & 0 & 0 \\ \{\mathscr{R}\}^{d}_{\sigma\phi}(\mathbf{r}',t';\mathbf{r},t) & \mathscr{R}^{d}_{\sigma\sigma}(\mathbf{r}',t';\mathbf{r},t) & 0 & 0 \end{pmatrix} \tag{A.3}$$

The zeros are due to the causality which prohibits correlations of two hatted variable.$^{(36)}$ There are similar expressions for $\mathscr{G}^{f}_{\lambda\mu}(\mathbf{r},t;\mathbf{r}',t')$.

Next we turn to the matrix $\gamma$:

$$(\gamma)_{\alpha\beta} \equiv \gamma_{\alpha\beta} = -(\mathscr{G}_0^{-1})_{\alpha\beta} \tag{A.4}$$

There are two types:

$$\gamma^{d}_{\lambda\mu} \equiv \gamma^{aa}_{\lambda\mu}, \qquad \text{and} \qquad \gamma^{f}_{\lambda\mu} \equiv \gamma^{ab}_{\lambda\mu}, \qquad \text{with} \quad a \neq b \tag{A.5}$$

Since this part is linear, it is simpler to Fourier transform with wave vector $\mathbf{k}$. Then we can write

$$(\gamma^{d}_{\lambda\mu})_{tt'}(\mathbf{k}) = \begin{pmatrix} 0 & 0 & -iD^{0}_{t}(\mathbf{k}) & 0 \\ 0 & 0 & 0 & \frac{i}{2}L_\sigma \\ -iD^{0}_{t'}(\mathbf{k}) & 0 & -LT & 0 \\ 0 & \frac{i}{2}L_\sigma & 0 & -L_\sigma T \end{pmatrix} \tag{A.6}$$

$$(\gamma^{f}_{\lambda\mu})_{tt'}(\mathbf{k}) = \begin{pmatrix} 0 & 0 & -iL\frac{g}{M\beta} & 0 \\ 0 & 0 & 0 & 0 \\ -iL\frac{g}{M\beta} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \tag{A.7}$$

where

$$D^{0}_{t}(\mathbf{k}) \equiv \partial_t + L\left(u_2 + U_{\mathbf{k}} - \frac{g}{M\beta}\right) \tag{A.8}$$

Next we consider bare vertices $\gamma_{\alpha\beta\gamma}$ which are symmentric with respect to their indices. The only nonvanishing type is of the form:

$$\gamma_{\alpha\beta\gamma} = \gamma^{aaa}_{\lambda\mu\nu} = \gamma_{\lambda\mu\nu}\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t'), \qquad \lambda, \mu, \nu = \phi, \hat{\phi}, \sigma, \hat{\sigma} \tag{A.9}$$

Here we list only the nonvanishing coefficients $\gamma_{\lambda\mu\nu}$ :

$$\gamma_{\hat{\phi}\sigma\phi} = Li\sqrt{u_4}\,, \qquad \gamma_{\hat{\sigma}\phi\phi} = -L\sigma\sqrt{u_4} \tag{A.10}$$

and those formed by interchanging indices.

We next find explicit forms for elements of $\mathscr{G}_0 = -\gamma^{-1}$ which we write

$$\mathscr{G}_0 = -\gamma^{-1} = (\mathscr{G}^d_0 - \mathscr{G}^f_0)\,\mathbf{1} + \mathscr{G}^f_0 \mathbf{E} \tag{A.11}$$

Forming $-\gamma \cdot \mathscr{G}_0 = \mathbf{1}$ we obtain the equations for $\mathscr{G}^d_0$, $\mathscr{G}^f_0$ in terms of $\gamma^d$, $\gamma^f$. After some algebra we obtain

$$\mathscr{G}^f_0 = (\gamma^d + (M-1)\gamma^f)^{-1} \cdot \gamma^f \cdot (\gamma^d - \gamma^f)^{-1} \tag{A.12}$$

$$\mathscr{G}^d_0 = (\gamma^d + (M-1)\gamma^f)^{-1} \cdot [\gamma^f - (\gamma^d + (M-1)\gamma^f)] \cdot (\gamma^d - \gamma^f)^{-1} \tag{A.13}$$

In view of (A.12), (A.13), (A.6), and (A.7) we shall need inverses of the matrices of the form:

$$X \equiv \begin{pmatrix} 0 & 0 & A & 0 \\ 0 & 0 & 0 & B \\ A' & 0 & C & 0 \\ 0 & B & 0 & D \end{pmatrix} \tag{A.14}$$

Although some of the elements of matrix are operators, they commute and can be treated as c-numbers. We find the following whose correctness can be directly verified:[21]

$$X^{-1} = \begin{pmatrix} -\frac{C}{AA'} & 0 & \frac{1}{A'} & 0 \\ 0 & -\frac{D}{B^2} & 0 & \frac{1}{B} \\ \frac{1}{A} & 0 & 0 & 0 \\ 0 & \frac{1}{B} & 0 & 0 \end{pmatrix} \tag{A.15}$$

---

[21] A way to find this inverse is to notice that $X$ consists of four $2 \times 2$ submatrices of the forms $x\mathbf{1} + y\sigma_z$ where $x$, $y$ are some numbers and $\mathbf{1}$ and $\sigma_z$ are, respecitvely, $2 \times 2$ unit matrix and the Pauli spin matrix. The inverse of $X$ has the same form with the coefficients determined by the condition $X \cdot X^{-1} = (4 \times 4$ unit matrix).

Putting all these results together and with further algebra we arrive at the following expressions for the $\mathscr{G}_0$'s:

$$(\mathscr{G}_0^f)_{tt'}\,(\mathbf{k}) = \begin{pmatrix} \mathscr{U}_{\mathbf{k}}(tt') & 0 & \mathscr{V}_{\mathbf{k}}(t') & 0 \\ 0 & 0 & 0 & 0 \\ \{\mathscr{V}\}_{\mathbf{k}}(t) & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \delta(t-t') \tag{A.16}$$

$$(\mathscr{G}_0^d)_{tt'}(\mathbf{k}) = \begin{pmatrix} LT[\mathscr{U}_{\mathbf{k}}(tt') - G_{\mathbf{k}}^1(t)\, G_{\mathbf{k}}^1(t')] & 0 & \mathscr{W}_{\mathbf{k}}(t') & 0 \\ 0 & \frac{4T}{L\sigma} & 0 & \frac{2i}{L\sigma} \\ \mathscr{W}_{\mathbf{k}}(t) & 0 & 0 & 0 \\ 0 & \frac{2i}{L\sigma} & 0 & 0 \end{pmatrix} \delta(t-t') \tag{A.17}$$

where

$$\mathscr{U}_{\mathbf{k}}(tt') \equiv \frac{1}{M}\,(G_{\mathbf{k}}^1(t)\, G_{\mathbf{k}}^1(t') - G_{\mathbf{k}}^2(t)\, G_{\mathbf{k}}^2(t')), \qquad \mathscr{V}_{\mathbf{k}}(t) \equiv \frac{1}{M}\,(G_{\mathbf{k}}^1(t) - G_{\mathbf{k}}^2(t))$$

$$G_{\mathbf{k}}^1(t) \equiv (-i\partial_t - iL(u_2 + U_{\mathbf{k}}))^{-1},$$

$$G_{\mathbf{k}}^2(t) \equiv \left(-i\partial_t - iL\left(u_2 + U_{\mathbf{k}} - \frac{g}{M\beta}\right)\right)^{-1}$$

$$\mathscr{W}_{\mathbf{k}}(t) \equiv -G_{\mathbf{k}}^1(t) + \mathscr{V}_{\mathbf{k}}(t) \tag{A.18}$$

Now, the Hartree term $-\gamma_{\alpha\beta\gamma} x_\gamma$ mentioned in connection with (7.84) arises in our context by the fact that the average $\langle\sigma\rangle$, which is written simply as $\sigma$ does not in general vanish. From (7.60) we can infer that for large $L_\sigma$ the unaverged $\sigma$ obeys the Langevin like equation as

$$\sigma(\mathbf{r}, t) = -i\sqrt{u_4}\;\phi(\mathbf{r}, t)^2 + (\text{Gaussian noise}) \tag{A.19}$$

Hence we can deduce if $\langle\phi\rangle = 0$

$$\langle\sigma(\mathbf{r}, t)\rangle = -i\sqrt{u_4}\langle\phi(\mathbf{r}, t)^2\rangle = -i\sqrt{u_4}\;\mathscr{C}^{aa}(\mathbf{r}, t; \mathbf{r}, t) \tag{A.20}$$

which produces the Hartree term.

Now there are two ways to get SCSA equations:

1. Use (7.89). Since we know the forms of $\mathscr{G}^d$, (A.3), and the corresponding expression for $\mathscr{G}^f$, we need to know the form of $\mathscr{G}^{-1}$ appearing on

the l.h.s. of (7.89) to obtain SCSA equations. For this purpose we can use general relationships connecting $\mathscr{G}^d$, $\mathscr{G}^f$ on one hand, and $\Gamma^d$, $\Gamma^f$ appearing in the definition (A.21) below.

2. Use (7.90). Here we need to know the expressions for $\mathscr{G}_0$ appearing on the r.h.s. of (7.90). These are just given by (A.16) and (A.17).

As was said above we can find the $\Gamma$'s defined through

$$\mathscr{G}^{-1}=(\Gamma^d-\Gamma^f)\,\mathbf{1}+\Gamma^f\mathbf{E} \tag{A.21}$$

Formally this is the same as the relationship between $\mathscr{G}_0$ and $\mathscr{G}_0^{-1}=-\gamma$ obtained before. See (A.12) and (A.13). Sructural similarity to the earlier case for the bare correlation and response functions is closer if we can assume that

$$\mathscr{R}^{d,f}_{\phi\sigma}=\mathscr{R}^{d,f}_{\sigma\phi}=\mathscr{C}^{d,f}_{\phi\sigma}=\mathscr{C}^{d,f}_{\sigma\phi}=0 \tag{A.22}$$

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