Does Fluctuating Nonlinear Hydrodynamics Support an Ergodic-Nonergodic Transition?

Shankar P. Das\textsuperscript{1} and Gene F. Mazenko\textsuperscript{2}

\textsuperscript{1} School of Physical Sciences, Jawaharlal Nehru University  
New Delhi 110067, India

\textsuperscript{2} The James Franck Institute and the Department of Physics  
The University of Chicago  
Chicago, Illinois 60637, USA

Abstract

Despite its appeal, real and simulated glass forming systems do not undergo an ergodic-nonergodic (ENE) transition. We reconsider whether the fluctuating nonlinear hydrodynamics (FNH) model for this system, introduced by us in 1986, supports an ENE transition. Using non-perturbative arguments, with no reference to the hydrodynamic regime, we show that the FNH model does not support an ENE transition. Our results support the findings in the original paper. Assertions in the literature questioning the validity of the original work are shown to be in error.
I. INTRODUCTION

It is appealing to associate the vitrification of the liquid into a frozen glassy state as an ergodic nonergodic (ENE) transition. Unfortunately there is strong evidence against the ENE transition scenario in physical and numerical experiments. This is in agreement with the results we found twenty years ago in Ref. [1] (hereafter mentioned as DM) when we introduced the model of fluctuating nonlinear hydrodynamics (FNH). We present here a nonperturbative analysis of the FNH model and the possibility of an ENE transition. In the end our results here agree with those in Ref. [1]. There is no sharp ENE transition in the FNH model. Recent reservations [2] concerning our results in are shown to be unfounded. We also address some misrepresentations [3] of our work.

In the theory of Classical Liquids, a new approach to studying the complex behavior of the supercooled state started with the introduction of the self consistent mode-coupling theory (MCT) [4, 5]. The model referred to here is based on a nonlinear feedback mechanism due to the coupling of the slowly decaying density fluctuations in the supercooled liquid. The feedback effects at metastable densities strongly enhance the transport properties of the liquid. In the simple version proposed initially [6, 7, 8] a sharp ergodic to non-ergodic (ENE) transition of the liquid into a glassy phase was predicted. This transition occurs at a critical density (or at the corresponding values of other controlling thermodynamic parameters) beyond which the density auto correlation function freezes at a nonzero value over long times. Soon afterward it was demonstrated that this sharp ENE transition is [1] rounded. The absence of a sharp ENE transition in the supercooled liquids was supported by work [9, 10] using similar theoretical models. Two recent works [2, 3] has called these conclusions into question. The purpose of the present paper is to show that our previous analysis withstands careful scrutiny and to reassert that the results of ref. [1] are correct and captures the right phenomena for the removal of the ENE transition.

We organize this paper as follows. In the next section we briefly introduce the FNH model. This is followed by an analysis of whether this model supports an ENE transition. In section III we compare our findings here to those in DM. Next we comment on the works which question the conclusions in DM. We end the paper with a short discussion.
II. THE FLUCTUATING NONLINEAR HYDRODYNAMIC MODEL

In Ref. [1] a model for the long time relaxation behavior of the supercooled liquid was constructed using fluctuating nonlinear hydrodynamics. The dynamics of collective modes in the liquid was formulated with nonlinear Langevin equations involving bare transport coefficients. These nonlinear stochastic equations for the time evolution of the conserved densities are plausible generalizations of the macroscopic hydrodynamic laws. The set of collective variables \( \{\psi_i\} \) for the liquid we considered consists of mass and momentum densities \( \{\rho(\mathbf{r},t), \mathbf{g}(\mathbf{r},t)\} \). The construction of the equations of motion [11] for the slow variables involve a driving free energy functional \( F \) which is expressed in terms of the hydrodynamic fields, i.e., \( \rho \) and \( \mathbf{g} \). The corresponding equilibrium distribution for the system is \( \exp(-\beta F) \).

The free energy functional \( F \) is separated in two parts, \( F = F_K[\mathbf{g},\rho] + F_U[\rho] \). The dependence of \( F \) on \( \mathbf{g} \) is entirely in the kinetic part \( F_K \) in the form [12] constrained by galilean invariance:

\[
F_K[\mathbf{g},\rho] = \int d\mathbf{x} \frac{g^2(\mathbf{x})}{2\rho(\mathbf{x})}. \tag{1}
\]

The potential part \( F_U \) is treated as a functional of the density only. The density \( \rho \) follows the continuity equation

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{g} = 0, \tag{2}
\]

having the flux as the momentum density \( \mathbf{g} \) which itself is a conserved property. The nonlinear equation for the momentum current density \( g_i \) is a generalized form of the Navier-Stokes equation [1],

\[
\frac{\partial g_i}{\partial t} = -\sum_j \nabla_j \left[ \frac{g_j g_i}{\rho} \right] - \rho \nabla_i \frac{\delta F_U}{\delta \rho} - \sum_j L_{ij}^o \frac{g_j}{\rho} + \theta_i. \tag{3}
\]

The noise \( \theta_i \) is assumed to be Gaussian following the fluctuation dissipation relation to the bare damping matrix \( L_{ij}^o \). For compressible liquids, the \( 1/\rho \) non-linearity appear in two terms in the generalized Navier-Stokes equation. These are respectively the convective term coupling two flow fields and the dissipative term involving the bare viscosity of the liquid. The appearance of this non linearity in the hydrodynamic equations is formally avoided in Ref. [1] by introducing the local velocity field \( \mathbf{V}(\mathbf{x},t) \),

\[
\mathbf{g}(\mathbf{x},t) = \rho(\mathbf{x},t) \mathbf{V}(\mathbf{x},t). \tag{4}
\]
The set of fluctuating variables in terms of which the renormalized field theory is constructed in our analysis therefore consists of the set $\psi_i \equiv \{\rho, g, V\}$.

The consequences of the nonlinearities in the equations of motion, i.e., renormalization of bare transport coefficients, are obtained using graphical methods of field theory \[13\]. The correlation of the hydrodynamic fields involve averages defined in terms of the action $A$ which is a functional of the field variables $\{\psi_i\}$ and the corresponding conjugate hatted fields $\{\hat{\psi}_i\}$ introduced in the MSR formalism. Using the equations of motions (2) and (3) respectively for $\rho$ and $g$ the action functional is obtained as \[1\],

$$
A = \int dt \int d\mathbf{x} \left\{ \sum_{ij} \hat{g}_i \beta^{-1} L_{ij}^o \hat{g}_j + i \sum_i \hat{g}_i \left[ \frac{\partial g_i}{\partial t} + \rho \nabla_i \delta F_u \delta \rho + \sum_j \nabla_j (\rho V_i V_j) - \sum_j L_{ij} V_j \right] + i \hat{\rho} \left[ \frac{\partial \rho}{\partial t} + \nabla \cdot g \right] + i \sum_i \hat{V}_i [g_i - \rho V_i] \right\},
$$

(5)

The theory is developed in terms of the correlation functions,

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

(6)

and the response functions,

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

(7)

The averages here are functional integrals over all the fields weighted by $e^{-A}$.

The nonlinearities in the equations of motion (3) and (4) give rise non-gaussian terms in the action (5) involving products of three or more field variables. The role of the non gaussian parts of the action $A$ on the correlation functions are quantified in terms of the self energy matrix which show up in the equation satisfied by the response functions and that satisfied by the correlation functions. We begin with the response functions which satisfy:

$$
\left[ (G_0^{-1})_{\hat{\alpha}\mu}(13) - \Sigma_{\hat{\alpha}\mu}(13) \right] G_{\mu\hat{\beta}}(32) = \delta(12)\delta_{\hat{\alpha}\hat{\beta}},
$$

(8)

with self energies $\Sigma_{\hat{\alpha}\mu}$ which can be expressed in perturbation theory in terms of the two-point correlation and response functions. Using the explicit polynomial form of the action (5), the response functions are expressed in the general form,

$$
G_{\alpha\hat{\mu}} = \frac{N_{\alpha\hat{\mu}}}{D}
$$

(9)
where the matrix $N$ is given in table I and the determinant $D$ in the denominator is given by

$$D = \rho L(\omega^2 - q^2 c^2) + i L(\omega + iq^2\gamma) \quad .$$  (10)

The various quantities are defined such that $\rho_L$, $c^2$ and $L$ are identified as the corresponding renormalized quantities respectively for the bare density $\rho_0$, speed of sound squared $c^2_0$ and longitudinal viscosity $L_0$. We have in terms of single-hatted or response self-energies:

$$\rho_L = \rho_0 - i \Sigma_{VV} \quad (11)$$

$$L = L_0 + i \Sigma_{gV} \quad (12)$$

$$qc^2 = qc^2_0 + \Sigma_{g\rho} \quad (13)$$

and $\gamma$ is defined in terms of the self energy element $\Sigma_{\bar{V}\rho} \equiv q\gamma$. One can also show that the correlation functions of the physical un-hatted field variables are given by,

$$G_{\alpha\beta} = - \sum_{\mu\nu} G_{\alpha,\mu} C_{\mu\nu} G_{\nu\beta} \quad (14)$$

where Greek letter subscripts take values $\rho, g, V$, and the self energy matrix $C_{\mu\nu}$ is given by,

$$C_{\mu\nu} = 2\beta^{-1} L_0 \delta_{\mu\bar{\nu}} \delta_{\bar{\mu}\bar{\nu}} - \Sigma_{\mu\bar{\nu}} \quad .$$  (15)

The double-hatted self-energies $\Sigma_{\mu\nu}$ vanish if either index corresponds to the density. This model does not have a complete set of FDR linearly relating correlation and response functions.

However, using the time translational invariance properties of the action (5), we obtained in DM the following fluctuation dissipation relation between correlation and response functions involving the field $g$ in the form :

$$G_{V_i\alpha}(q, \omega) = -2\beta^{-1}\text{Im}G_{\bar{g}_i\alpha}(q, \omega) \quad (16)$$

where $\alpha$ indicates any of the fields $\{\rho, g, V\}$. 

4
III. ERGODIC-NONERGODIC TRANSITION AND FNH

Does this model have an ENE transition? To answer this question we first pose the conditions for such a transition. Suppose, due to a nonlinear feedback mechanism, the self-energy $\Sigma_{\bar{g}\bar{g}}$ blows up at small frequencies:

$$\Sigma_{\bar{g}\bar{g}} = -A\delta(\omega).$$  \hspace{1cm} (17)

This is presumed to result from a persistent time dependence of the density correlation function. This hypothesis is motivated by the one-loop contribution and the physics of the viscosity blowing up as one enters the glass. Is this assumption compatible with the set of Dyson equations? What we mean by a nonergodic phase is that $G_{\rho\rho}$ shows a $\delta$-function peak at zero frequency. Putting Eq.$(17)$ back into Eq.$(14)$ we obtain a $\delta(\omega)$ peak in $G_{\rho\rho}$ as long as the response function $G_{\rho\bar{g}}$ is not zero in the $\omega \to 0$ limit. We assume, with no reason to expect otherwise, that the $\omega \to 0$ limits of $\rho_L$, $\gamma$, $c^2$ and $L$ are nonzero. With these assumptions $D$ is not infinite in the low frequency limit and $G_{\rho\bar{g}}$, and $G_{V\bar{g}}$ are nonzero in the low frequency limit. Then from Eq.$(14)$ we find that $G_{\rho\rho}$, $G_{\rho V}$, and $G_{V V}$ show a $\delta(\omega)$ component. Since $G_{\bar{g}\bar{g}}$ vanishes as $\omega \to 0$ as long as $D(\omega = 0) \neq 0$, the correlation functions involving a momentum density index do not show a $\delta$-function peak at zero frequency. So it is necessary for an ENE transition that $G_{\rho\bar{g}}$ not vanish as $\omega \to 0$. This requires that $\rho_L$ goes to a nonzero value in the zero frequency limit and the determinant $D$ not blow up as $\omega \to 0$.

If, as expected, the self-energy contribution $\gamma(\omega = 0) \neq 0$ then the correlation functions $G_{\rho V}$, and $G_{V V}$ show a $\delta(\omega)$ component. Now we apply the FDT $(16)$. Since $G_{V\rho}$ and $G_{V V}$ blow up, it then follows from the FDT that the imaginary parts of the response functions $G_{\bar{g}\rho}$ and $G_{\bar{g} V}$ also blow up. However we also require simultaneously that $D^*D$ is bounded, and imaginary parts of both $\rho_L q D^*$ and $(\omega + iq^2\gamma)D^*$ diverge. But since both $D'$ and $D''$ denoting the real and imaginary parts of $D$ are bounded so $\rho_L$ and $\gamma$ must diverge. However, if these quantities blow up then from $(10)$ it follows that $D$ must also blow up and we have a contradiction. The obvious conclusion is that the original assumption of a nonergodic phase is not supported in the model. The key self-energy contribution is $\gamma$. If for some reason this quantity vanishes at zero frequency then $G_{\rho\bar{g}}$, and $G_{V V}$ vanish as $\omega$ goes to zero. Then $G_{\rho\rho}$, and $G_{V V}$ do not show a $\delta(\omega)$ component and one does not have the constraints on $\rho_L$, 

γ, and D. In this case one may have an ENE transition in this model.

IV. RELATION TO DM RESULTS

The argument we give in the hydrodynamic regime in Ref. [1] is completely consistent with the results presented above. The simplest way of understanding the argument in the previous section is to look at the response function

$$G_{\rho\dot{\rho}} = \frac{\omega \rho_L + iL}{\rho_L(\omega^2 - q^2c^2) + iL(\omega + iq^2\gamma)}.$$  \hspace{1cm} (18)

The renormalization of the longitudinal viscosity $L$ is computed, see Eq. (12) in terms of the longitudinal part $\Sigma_{\hat{g}V}^L$ of the corresponding self-energy matrix $\Sigma_{\hat{g}_iV_j}$ of the isotropic liquid,

$$L(q,z) = L_0 + \frac{\beta}{2} \Sigma_{\hat{g}V}^L(q,z).$$  \hspace{1cm} (19)

If we ignore the self energy $\Sigma_{\hat{V}\rho}$, the expression (18) is identical to the conventional expression for the density correlation function with the generalized memory function or the renormalized transport coefficient $L(q,z)$. The dependence of $G_{\rho\dot{\rho}}$ on the self energy $\Sigma_{\hat{V}\rho}$ in the renormalized theory is a consequence of the non-linear term involving the $\hat{V}$ field in the MSR action (5) and is originating from the nonlinear constraint (4) introduced to deal with the $1/\rho$ nonlinearity in the hydrodynamic equations. Analyzing the expression (14) for the correlation functions and the FDT relation (16) we obtain in the hydrodynamic limit the following nonperturbative relation between the two types of self energies contributing alternatively to the renormalization of the longitudinal viscosity,

$$\gamma_{\hat{g}\hat{g}}(0,0) = 2\beta^{-1} \left[ \gamma'_{\hat{g}V}(0,0) + \lim_{\omega \to 0} \frac{\gamma''_{\rho\dot{\rho}}(0,\omega)}{\omega} \right]$$  \hspace{1cm} (20)

where we have used in the above following definitions, in the isotropic limit, $\Sigma_{\hat{g}\hat{g}}^L \sim -q^2\gamma_{\hat{g}\hat{g}}$, $\Sigma_{\hat{g}V}^L \sim -iq^2\gamma_{\hat{g}V}$, and $\Sigma_{\rho\dot{\rho}} \sim q\gamma_{\rho\dot{\rho}}$. The relation (20) which is obtained from the FDT relation (16) only, implies that both the self energies $\Sigma_{\hat{g}\hat{g}}$ and $\Sigma_{\hat{g}V}$ have the same diverging contribution in the low frequency limit. In the simplified model it is this contribution in terms of density correlation function which constitute the feed back mechanism of MCT and leads to the dynamic transition beyond a critical density. The singular contribution to the renormalized transport coefficient $L$ in (18) is now obtained in terms of the self energy $\Sigma_{\hat{g}V}$. As a consequence of (20) it also follows that the response function $G_{\rho\dot{\rho}}$ is equal to the
corresponding density correlation function $G_{\rho\rho}$ in the hydrodynamic limit. It is important to note here that (contrary to the assertion in Ref. [2]) this relation is not forced by us, rather it follows as a natural consequence of the relations (20) linking the response to correlation self energies.

The asymptotic behavior of the density correlation function is inferred from $G_{\rho\hat{\rho}}$. The denominator of (18) for the response functions contain the self energy $\Sigma_{V\rho}$ which in this case is crucial for the long time dynamics and understanding how the ENE transition is cutoff. The density correlation function (in the small $q, \omega$ limit) only freeze due to the feedback mechanism if the self energy matrix elements $\Sigma_{V\rho}$ is zero - a result obtained in the earlier section. In this regard it is useful to note that for the $\omega \to 0$ limit the quantity $L(\omega + i\gamma q^2)$ in $D$ does not diverge even when $L \sim 1/\omega$ is getting large, since $L\gamma q^2$ remains finite in the non hydrodynamic regime $\omega \sim q^2$. To leading order in wave numbers $q\Sigma_{V\rho}(q,0) \equiv q^2\gamma$, is expressed in terms of the self energy $\Sigma_{VV}^L$ using the nonperturbative relation

$$\gamma_{\hat{V}V}(0,0) = \frac{2\rho\beta^{-1}}{c^2}\gamma_{V\rho}'(0,0),$$

(21)

where $c$ is the sound speed introduced in (13). Note that the relation (21) is also obtained from the same fluctuation-dissipation relation (16).

V. ABL AND CR

We now address the criticisms made in Ref. [2] on our work. ABL imply that we misapplied the FDT relating $G_{\rho\rho}$ and $G_{\rho\hat{\rho}}$ in the hydrodynamical limit. These authors offer that we assumed a linear FDT from the beginning. DM clearly discusses the consequences of not having a complete set of FD relations. On the other hand the introduction of the $\theta = \delta F/\delta \rho$ field by ABL for obtaining a FDT in the linear form has not yet been shown to be useful. The nonlinear contribution in $\theta$ comes from the part $\delta F_K/\delta \rho$, which actually gives rise [1] to the term $\nabla_j (g_i g_j / \rho)$ in the generalized Navier-Stokes equation. The latter is essentially the $1/\rho$ nonlinearity which we address in our model through the introduction of the variable $V$. In this regard we believe that the importance of linear FDT in the MSR formulation has been overemphasized by ABL. Indeed in the absence of a linear FDT the response functions lose their physical meaning and become mere computational tools. From a physical point of view however what is important is that the correlation functions are time
invariant which is maintained as can be directly seen from the above equation (14).

In Ref. [3] the cut off mechanism of Ref. [1] has been questioned by treating the highly nonlinear model described above using a rather naive approach. CR basically make some phenomenological manipulations on a Newtonian dynamics model [14], ending with a memory function description they claim, without proof, is related to our model. All subsequent discussion of our work made by these authors are based on this claim. Our model, as shown on examination of table I, satisfies at all stages the density conservation law. The memory function proposed in CR to represent our work, Eq.(5) there, does not satisfy this conservation law. Therefore the analysis of CR does not apply to the model we studied. None of their conclusions concerning our work have any validity. CR concede that there is no error in our calculation, rather they offer vaguely that our model itself is the problem! Our model, as shown on examination of table I, satisfies at all stages the density conservation law. The source of their error appears to be the naive assumption that this model can be represented in terms of a single memory function [15]. This work represents a fundamental misunderstanding of the problem.

Though the authors of both papers, ABL and CR, seem to agree that finally the ENE transition does not survive they disagree with our analysis of the problem. The arguments put forward in Ref. [3] to rediscover that the transition is finally cutoff are rather vague and of descriptive nature. These authors only seem to conjecture that the transition will be cutoff nonperturbatively citing other recent works [16].

VI. DISCUSSION

The basic feedback mechanism of MCT is a consequence of simple quadratic nonlinearities in density fluctuations (arising from purely dynamic origin) that is present in the pressure term of the generalized Navier-Stokes equation. The ergodicity restoring mechanism goes beyond this. The description in terms of coupling to currents is a physically appealing way of explaining the nature of the FNH equations (expressed in a form which can be sensibly related to the hydrodynamics of liquids). It is in fact the full implications of the density nonlinearities in the dynamics that cuts off the sharp transition to nonergodicity. This is also reflected in the fact that the basic conclusions of Ref. [1] follow even if the relevant nonlinearity is considered in a different manner. In fact by formulating the model [17] only
in terms of the fields \( \{ \rho, g \} \) the same conclusions implying the absence of the dynamic transition is reached as in Ref. [1]. The \( 1/\rho \) nonlinearity mentioned above is treated here in terms of a series of density nonlinearities. The self energy matrix elements \( \Sigma_{gV} \) and \( \Sigma_{V\rho} \) are absent from the theoretical formulation in this case and the cutoff kernel is obtained here from a different self energy element \( \Sigma_{g\rho} \).

Twenty years ago we had predicted that the feedback effects from mode-coupling of density fluctuations, when properly analyzed keeping consistency with concepts of basic hydrodynamics, results in a qualitative crossover in the dynamics. We presented here a selfcontained nonperturbative proof that FNH does not support an ENE transition. This new analysis is completely compatible with the results of DM, simulations and experiment.

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\[
\begin{array}{|c|c|c|}
\hline
\ & \hat{\rho} & \hat{g} & \hat{V} \\
\hline
\rho & \omega \rho L + iL & \rho L q & L q \\
g & q(\rho_L c^2 + L \gamma) & \rho L \omega & L \omega \\
V & q(c^2 + i \omega \gamma) & \omega + iq^2 \gamma & i(\omega^2 - q^2 c^2) \\
\hline
\end{array}
\]

TABLE I: The matrix of the coefficients $N_{\alpha \beta}$ in the numerator on the RHS of eqn. (9) for the response functions

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