Fluctuating hydrodynamics and diffusion in amorphous solids

Shankar P. Das*
Saha Institute of Nuclear Physics, 1/AF Bidhannagar, Calcutta 700 064, India

Rolf Schilling
Institut fur Physik, Johannes Gutenberg Universität, D55099 Mainz, Germany
(Received 15 September 1993; revised manuscript received 11 April 1994)

The fluctuating hydrodynamic description for an isotropic fluid is extended to include the displacement field **u**, reflecting the freezing of the local structures in an amorphous solid. The fluctuating nonlinear equations for the set of hydrodynamic variables including **u** has been obtained. The role of **u** is manifested through its longitudinal part, i.e., $\nabla\cdot\mathbf{u}$, in terms of which we define the variable $c(\mathbf{x},t)$. It refers to the diffusion of the free volume or vacancies, signifying configurational rearrangements in the amorphous solid. The analysis here shows that one recovers the earlier result obtained by Das and Mazenko [Phys. Rev. A **34**, 2265 (1986)] for mode coupling models of a glass transition, where the time scales associated with the relaxation of the density fluctuations and that of the vacancy diffusion can be comparable.

PACS number(s): 64.70.Pf, 05.60.+w, 64.60.Cn

I. INTRODUCTION

Use of mode coupling theory has been very useful in understanding its slow relaxation behavior at supercooled densities. The basic mechanism [1–3] producing long time scales at supercooled densities is related to the coupling of the density fluctuations causing a nonlinear feedback to the viscosity of the supercooled liquid. For reviews the reader is referred to [4]. For an idealized model it was shown that this feedback mechanism results in a freezing of the liquid to a nonergodic glassy state above a critical density ρ_c or below a critical temperature T_c. In this state the long time limit of the density correlation function remains nonzero for all values of the wave vector q and the viscosity diverges. This transition is essentially a dynamic one and the static properties of the liquid are not affected by it. In subsequent works [5] a more careful treatment of the dynamical equations for the compressible fluid was done by Das and Mazenko. The analysis involved the development of a Martin-Siggia-Rose-type field theory [6] for the dynamics of the fluctuating variables, constrained by the nonlinear relation $\mathbf{g}=\rho\mathbf{v}$, where **g** is the momentum density, ρ is the mass density, and **v** is the velocity for the fluid. It showed that the sharp transition of the liquid to the nonergodic phase is cut off and the dynamic correlation of density fluctuations over very long times actually decays through a diffusive process. Thus ergodicity is maintained at all densities. More recent works [7] have confirmed that the dynamical behavior for the fluid remains the same when the theory is formulated in terms of the two fields **g** and ρ without the nonlinear constraint. It was demonstrated that the cutoff mechanism discovered by Das and Mazenko is actually a consequence of the $1/\rho$ nonlinearity appearing in the hydrodynamic equations for the compressible fluid.

The dynamic instability in the mode coupling model is characterized by the long time behavior of the density autocorrelation function. The analysis of Ref. [5] showed that the Laplace transform of the density correlation function behaves like $1/(z+i\gamma q^2)$, for all values of density, instead of a $1/z$ pole, which would have implied a freezing of the dynamic correlation function. Thus the diffusive process by which the density correlation decays happens over a time scale $\tau=(\gamma q^2)^{-1}$. Such a diffusive decay in the hydrodynamic limit has also been obtained in the recent work by Schmitz, Dufty, and De [8]. The mode coupling contribution to the viscosity now remains finite for all densities. Subsequent works [9] including realistic structural effects and one-loop approximations for the cutoff function have provided good agreement with results obtained from computer simulations [10] of simple fluids. However, the simple approximate forms for the cutoff function were not enough to explain the extremely slow relaxation seen in some of the supercooled liquids. There were also suggestions [11] of making this time scale arbitrarily long by an *ad hoc* introduction of a lower cutoff time in calculating the mode coupling integrals.

Similar results demonstrating smoothing off the sharp transition at T_c were also reported by Götze and Sjögren [12]. But in contrast to the diffusive decay, the relaxation time of the ergodicity restoring processes, interpreted as hopping processes by these authors, does *not* diverge for $q\to 0$. Since, e.g., light scattering experiments above and below T_c exhibit an α peak (Mountain peak [13]) with a width which remains finite for $q\to 0$ (see also the discussion by Fuchs and Latz [14]), one must conclude that the hopping processes are dominant at least in the immediate neighborhood of T_c. Most recent work [15] even demonstrates that consideration of the hopping processes described by a single parameter δ can remove significant deviations between experimental data and the idealized mode coupling theory. Nevertheless, there might be a

*Present address: School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India.

diffusive part, but with a relaxation strength which is too small to be resolved. Of course, this strength might increase with decreasing temperature.

Since both cutoff mechanisms discussed above were obtained from a mode coupling approach starting from the *liquid side* it is tempting to use such an approach from the *glass side*. To do this is the purpose of the present contribution; thereby ideas will be adopted that have been used to make a hydrodynamic description for systems with solidlike properties.

Earlier, attempts have been made to understand the diffusive transport in an amorphous solid using the free volume [16] approach. This mainly involved associating some free volume or void with the individual units in the cluster structure of the liquids. The movement of the free volumes was considered crucial for the transport in the liquid. The idea of liquidlike and solidlike clusters was introduced from a phenomenological picture of the supercooled liquid and percolation theory used to predict that at some critical density the transport process gets completely frozen, giving rise to very long relaxation times. The consideration of the dynamics of the vacancies in a crystal from a unified hydrodynamic approach was first proposed by Martin, Parodi, and Pershan [17]. This was further developed by Cohen and co-workers to describe the linear transport in a crystal [18] and also an amorphous solid [19]. Usually the transport properties of the isotropic fluid are considered through the dynamical equations for the conserved modes which arise as a direct consequence of the microscopic conservation laws in the system. The hydrodynamic description for crystals as well as liquid crystals involved extending the set of hydrodynamic variables in the system to include the Nambu-Goldstone modes due to symmetry breaking. The additional hydrodynamic variable that was introduced was the displacement vector for the different lattice sites. Thus in a cubic crystal the number of conserved variables was increased to eight as compared to five in an isotropic fluid. The theory included the existence of transverse sound modes as well as the vacancy diffusion in the crystal. In the case of liquid crystals, study of the nonlinear fluctuating equations has proved to have nontrivial consequences for its transport properties [20]. Very recently it was also used to study [21] the glass transition in similar systems from a mode coupling approach. In the hydrodynamic approach the new bare transport coefficients referring to the dissipative parts in the dynamics of the Goldstone modes enter the theory as a parameter. Expressions for the viscosity coefficients and elastic constants for a crystal were obtained [23] starting from the revised Enskog equation [22]. However, this work ignored the process of vacancy diffusion in the crystal. A full microscopic consideration of this problem is still lacking [24].

In an amorphous solid the same idea used in formulating the hydrodynamic description of the crystal is extended. Although in this case there is no broken symmetry producing a long-range order similar to a crystal, it is assumed that there one can define a displacement field $\mathbf{u}(\mathbf{x})$ about the local metastable positions of the atoms which remain unaltered for a long time in the glassy state. This idea is supported by the fact that in an experiment one sees the transverse sound modes in the glass. Thus the translational symmetry is broken over length scales related to local structures, although it is assumed to be valid over long distances. The attempt of the present approach is to focus on how, through the introduction of the u fields, one can take into account rearrangements other than that of actual mass motion. In an amorphous system the idea of vacancy is not very appropriate and the term "vacancy diffusion" is used here for referring to diffusion of free volumes in it. Recently a similar approach of fluctuating hydrodynamics was used by Kim [25] to demonstrate that the introduction of the displacement fields led to stretched exponential behavior in glassy relaxation. In the present work our focus is on the vacancy diffusion process in the glassy system.

In order to keep the analysis simple we ignore the energy fluctuations in the present formulation. The dynamical equations for the fluctuating variables are obtained with the standard techniques used by Ma and Mazenko [26] in the study of dynamics of critical phenomena. In the first part of the analysis we keep the treatment general using a rather standard form of the effective Hamiltonian that determines the equilibrium behavior in the system. To focus on the diffusive motion of the free volume we introduce a variable $c(\mathbf{r},t)$ for the amorphous solid similar to Cohen, Fleming, and Gibbs [19] which follows a diffusive dynamics. We develop a Martin-Siggia-Rose-type field theory for the dynamical set of equations obtained for the slow variables following the formalism developed in Ref. [5]. The importance of the coupling of the density fluctuations with the vacancy mode is demonstrated from a nonperturbative analysis.

The paper is organized as follows. In the next section we develop the fluctuating nonlinear hydrodynamic equations for the extended set of slow variables and obtain the diffusive equation for the variable $c(\mathbf{x},t)$ discussed above. In Sec. III we sketch briefly the field theoretic analysis necessary for computing the time correlation functions and in Sec. IV we analyze the renormalization of the theory. In Sec. V we consider the implications of the model for glass transition. We end the paper in Sec. VI with a short discussion.

II. NONLINEAR FLUCTUATING HYDRODYNAMICS OF AMORPHOUS SOLID

The dynamical equations for the slow variables are obtained using the standard techniques developed by Ma and Mazenko [26]. This is given by the generalized form of the Langevin equation for the fluctuating variable $\psi_i(t)$, where the label i stands for the different hydrodynamic variables in the system,

$$\frac{\partial \psi_i}{\partial t} = V_i[\psi] - \sum_j \Upsilon_{ij} \frac{\delta F}{\delta \psi_j} + \zeta_i \ . \tag{2.1}$$

The reversible part of the equation of motion given by $V_i[\psi]$ is called the streaming velocity and is represented by

$$V_i[\psi]=\sum_j\{\psi_i,\psi_j\}\frac{\delta F}{\delta\psi_j}\ , \tag{2.2}$$

where $\{\psi_i,\psi_j\}$ is the Poisson bracket [27] between the slow variables. $F[\psi]$ is the effective Hamiltonian that determines their equilibrium correlations for the fluid and in a Fokker-Planck description corresponding to Eq. (2.1) the stationary probability distribution is given by e^{-F}. The noise ζ_i is assumed to be Gaussian with the following fluctuation dissipation relation to the bare damping matrix Υ_{ij}:

$$\langle\zeta_i(t)\zeta_j(t')\rangle=2k_BT\Upsilon_{ij}\delta(t-t')\ , \tag{2.3}$$

where $k_BT=\beta^{-1}$ is the Boltzmann factor.

In evaluating the Poisson bracket mentioned above we need the microscopic variables $\hat{\psi}_i(\mathbf{x})$ corresponding to fields $\psi_i(\mathbf{x})$. We assume the system to be a collection of N classical particles each of mass m. $\mathbf{r}_\alpha(t)$ and $\mathbf{p}_\alpha(t)$ are, respectively, the position and the momentum of the αth particle at time t. For the density $\hat{\rho}$ and the momentum density $\hat{\mathbf{g}}$ we use the standard prescription [30]

$$\begin{aligned}\hat{\rho}(\mathbf{x},t)&=m\sum_{\alpha=1}^{N}\delta(\mathbf{x}-\mathbf{r}_\alpha(t))\ ,\\ \hat{g}_i(\mathbf{x},t)&=\sum_{\alpha=1}^{N}p_\alpha^i(t)\delta(\mathbf{x}-\mathbf{r}_\alpha(t))\ .\end{aligned} \tag{2.4}$$

The extra slow mode that is to be added to the set of conserved densities takes into account the broken symmetry of the solid state with elastic properties. We define here a fluctuating variable $\mathbf{u}(\mathbf{x},t)$, by considering the displacements of the individual particles $\mathbf{u}_\alpha(t)$ $(\alpha=1-N)$ from their respective mean position denoted by $\mathbf{r}_\alpha^0$,

$$\hat{\rho}(\mathbf{x},t)\hat{u}_i(\mathbf{x},t)=m\sum_{\alpha=1}^{N}u_\alpha^i(t)\delta(\mathbf{x}-\mathbf{r}_\alpha(t))\ , \tag{2.5}$$

such that $\mathbf{r}_\alpha(t)=\mathbf{r}_\alpha^0+\mathbf{u}_\alpha(t)$. Using the canonical Poisson bracket relation $\{r_\alpha^i,p_\beta^j\}=\delta_{ij}\delta_{\alpha\beta}$ we obtain the following relations for the hydrodynamic variables:

$$\begin{aligned}\{\hat{\rho}(\mathbf{x}),\hat{g}_i(\mathbf{x}')\}&=-\nabla_x^i[\delta(\mathbf{x}-\mathbf{x}')\hat{\rho}(\mathbf{x})]\ ,\\ \{\hat{g}_i(\mathbf{x}),\hat{g}_j(\mathbf{x}')\}&=-\nabla_x^j[\delta(\mathbf{x}-\mathbf{x}')\hat{g}_i(\mathbf{x})]\\ &\quad+\nabla_{x'}^i[\delta(\mathbf{x}-\mathbf{x}')\hat{g}_j(\mathbf{x})]\ ,\\ \{\hat{g}_i(\mathbf{x}),\hat{u}_j(\mathbf{x}')\}&=-\delta(\mathbf{x}-\mathbf{x}')[\delta_{ij}-\nabla_{x'}^i u_j(\mathbf{x}')]\ ,\\ \{\hat{u}_i(\mathbf{x}),\hat{u}_j(\mathbf{x}')\}&=0\ ,\\ \{\hat{u}_i(\mathbf{x}),\hat{\rho}(\mathbf{x}')\}&=0\ .\end{aligned} \tag{2.6}$$

The effective Hamiltonian has two parts, namely, the kinetic and the potential one: $F=F_K+F_U$. The kinetic energy part, in terms of the coarse grained fields, has the standard form (for a deduction from a microscopic point of view we refer to Langer and Turski [28])

$$F_K=\tfrac{1}{2}\int d^3x\, g^2(\mathbf{x})/\rho(\mathbf{x})\ . \tag{2.7}$$

Using Eqs. (2.6) and (2.7), we obtain the following forms for the streaming velocity terms:

$$\begin{aligned}V_\rho&=-\nabla\cdot\mathbf{g}\ ,\\ V_g^i&=-\nabla_j\left[\frac{g_ig_j}{\rho}\right]-\rho\nabla_x^i\frac{\delta F_U}{\delta\rho}-[\delta_{ij}-\nabla_x^i u_j(\mathbf{x})]\frac{\delta F_U}{\delta u_j(\mathbf{x})}\ ,\\ V_u^i&=\frac{g_i}{\rho}-\frac{\mathbf{g}}{\rho}\cdot[\nabla_x u_i]\ .\end{aligned} \tag{2.8}$$

Next, we consider the dissipative parts of the dynamical equations. For the momentum density equation, the irreversible part has the standard form with the dissipative coefficient in Eq. (2.1) given by

$$\Upsilon_{g_ig_j}\equiv L_{ij}=-\eta_0(\tfrac{1}{3}\nabla_i\nabla_j+\delta_{ij}\nabla^2)-\zeta_0\nabla_i\nabla_j\ , \tag{2.9}$$

where ζ_0 is the bare bulk viscosity and η_0 is the bare shear viscosity. We define the longitudinal viscosity as $L_0=\zeta_0+4\eta_0/3$. The dissipative term in the equation for the u field is assumed to have the simple time-dependent Ginzburg-Landau form

$$\Upsilon_{u_iu_j}\equiv\Gamma_{ij}=\bar{\Gamma}_0\delta_{ij} \tag{2.10}$$

and we require all other $\Upsilon_{ij}=0$. In the fluctuating hydrodynamic description the bare transport coefficients ζ_0, η_0, and Γ_0 act as external parameters. For the bare viscosities Green-Kubo-type relations can be evaluated in a kinetic theory approach using suitable models. The time scale for the quantity Γ_0 that relates to the vacancy diffusion is, however, much longer.

In order to compute the nonlinear fluctuating hydrodynamic equations we need the explicit form of the effective Hamiltonian F. In addition to the usual terms that appear in the free energy functional for an isotropic liquid, we include the energy cost due to distortion in the elastic solid, as well as the coupling of the density fluctuations to the displacement field $\mathbf{u}$. We use here an isotropic approximation for the solid. Thus the potential energy part $F_U[\rho,u]$ is constructed preserving the rotational and translational invariance of the system,

$$F_U=\tfrac{1}{2}\int d\mathbf{x}\left[A\left[\frac{\delta\rho}{\rho_0}\right]^2+2B\frac{\delta\rho}{\rho_0}S+\lambda S^2+2\mu(s_{ij}s_{ji})\right]\ . \tag{2.11}$$

Here S is the trace of the strain tensor field s_{ij} in the solid and the latter is defined in terms of the gradient of the field $\mathbf{u}(\mathbf{x})$ in the following form:

$$s_{ij}=\tfrac{1}{2}(\nabla_iu_j+\nabla_ju_i)-\tfrac{1}{2}\nabla_iu_m\nabla_ju_m\ . \tag{2.12}$$

λ and μ are related to the bulk and shear elastic moduli of the amorphous solid. The longitudinal modulus is given by $\vartheta=\lambda+\mu$. The couplings A and B in the effective Hamiltonian relate to the static structure factor for the system. Using Eq. (2.11) the following set of fluctuating nonlinear equations are obtained:

$$\frac{\partial \rho}{\partial t}+\nabla\cdot\mathbf{g}=0 ,$$
$$\frac{\partial g_i}{\partial t}+\sum_j \nabla_j \sigma_{ij}=0\theta_i , \tag{2.13}$$
$$\frac{\partial u_i}{\partial t}+\mathbf{v}\cdot\nabla u_i=v_i-\Gamma_{ij}\frac{\delta F}{\delta u_j}+f_i ,$$

together with the nonlinear constraint

$$\mathbf{g}=\rho\mathbf{v} . \tag{2.14}$$

The stress-energy tensor σ_{ij} has the reversible and the dissipative (irreversible) parts, respectively, denoted by σ_{ij}^R and σ_{ij}^D such that

$$\sigma_{ij}=\sigma_{ij}^R+\sigma_{ij}^D , \tag{2.15}$$

where

$$\sigma_{ij}^R=\frac{g_i g_j}{\rho}+\left[A\frac{\delta\rho}{\rho_0}+\frac{A}{2}\left[\frac{\delta\rho}{\rho_0}\right]^2 +BS-\frac{1}{2}\lambda S^2-\frac{\mu}{2}(s_{lm}s_{ml})\right]\delta_{ij}$$
$$-\left[\lambda S+B\frac{\delta\rho}{\rho_0}\right]\frac{\partial S}{\partial\nabla_i u_m}\frac{\partial S}{\partial\nabla_j u_m}-\mu[s_{ij}-2s_{im}s_{jm}] \tag{2.16}$$

and

$$\sigma_{ij}^D=-\eta_0[\nabla_i v_j+\nabla_j v_i-\tfrac{2}{3}\delta_{ij}(\nabla\cdot\mathbf{v})]-\zeta_0\delta_{ij}(\nabla\cdot\mathbf{v}) . \tag{2.17}$$

In the present formulation we have $\sigma_{ij}=\sigma_{ji}$, which guarantees conservation of angular momentum. The random parts in these equations are Gaussian noises and are related to the bare transport coefficients as

$$\langle\theta_i(\mathbf{x},t)\theta_j(\mathbf{x}',t')\rangle=2\rho_0 k_B T L_{ij}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') ,$$
$$\langle f_i(\mathbf{x},t)\theta_j(\mathbf{x}',t')\rangle=0 , \tag{2.18}$$
$$\langle f_i(\mathbf{x},t)f_j(\mathbf{x}',t')\rangle=2k_B T\Gamma_{ij}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') .$$

The set of fluctuating equations obtained above gives the dynamics of the slow modes for the solid with elastic properties. Calculation of correlation functions from these sets of equations provides [25] the existence of the transverse modes and the vacancy diffusion mode. In the present context we want to focus on the situation where freezing has occurred at the scale of local structure in the solid, but overall translational invariance is maintained over longer distances.

In order to include the motion of free volumes or vacancies in the amorphous solid, we introduce, following Cohen, Fleming, and Gibbs [19] the variable

$$c(\mathbf{x},t)=\delta\rho(\mathbf{x},t)+\rho_0(\nabla\cdot\mathbf{u}) , \tag{2.19}$$

where ρ_0 is the equilibrium density [29]. In the case where each lattice site is rigidly fixed to the molecules and the diffusion of vacancies is omitted from the formulation, the density fluctuation is simply given by $-\rho_0(\nabla\cdot\mathbf{u})$. The dynamical equation for $c(\mathbf{x},t)$ can be obtained using Eqs. (2.13) for the slow modes. We consider first the linearized dynamics of the fluctuating set of hydrodynamic variables $\{\rho(\mathbf{x},t),\mathbf{g}(\mathbf{x},t),\mathbf{v}(\mathbf{x},t),c(\mathbf{x},t)\}$. The linearized equations for ρ and $\mathbf{g}$ can immediately be obtained from Eqs. (2.13). The equation for $c(\mathbf{x},t)$ is obtained as

$$\frac{\partial c(\mathbf{x},t)}{\partial t}-\Gamma_0\nabla^2 c(\mathbf{x},t)+\Gamma_0'\nabla^2\delta\rho(\mathbf{x},t) +\nabla\cdot\mathbf{g}(\mathbf{x},t)-\rho_0\nabla\cdot\mathbf{v}(\mathbf{x},t)=\bar{f} , \tag{2.20}$$

where for notational convenience we make the substitutions

$$\Gamma_0=\vartheta\bar{\Gamma}_0 , \quad \Gamma_0'=\vartheta'\bar{\Gamma}_0 , \tag{2.21}$$

with $\vartheta'=\vartheta-B$ and the noise $\bar{f}=\rho_0\nabla\cdot\mathbf{f}$. The variable $c(\mathbf{x},t)$ is related to the longitudinal part of $\mathbf{u}$ through the quantity $\nabla\cdot\mathbf{u}$. In the present analysis we do not deal with the transverse part of u, which is related to the transverse sound modes in the amorphous solid. The motivation for focusing on u_l is that we want to investigate the dynamics of the structural rearrangements other than that associated with actual mass motion, namely, the diffusive motion of the vacancies given by the variable $c(\mathbf{x},t)$ in the amorphous solid.

III. FIELD THEORETIC FORMULATION

In order to calculate the dynamic correlation functions for the set of fields $\{\rho,\mathbf{g},\mathbf{v},c\}$ we develop a field theoretic description of the Martin-Siggia-Rose type. The formalism is standard in literature and we follow here closely the one developed in Ref. [5]. For each of the fields ψ_α we introduce a corresponding caretted field $\hat{\psi}_\alpha$ and let $\Psi\equiv\{\psi_\alpha,\hat{\psi}_\alpha\}$ be the field vector where α runs over the set of hydrodynamic modes. The correlation functions are obtained from the generating functional Z_U as

$$G(12)=\frac{\delta}{\delta U(2)}\frac{\delta}{\delta U(1)}\ln Z_U=\langle\delta\Psi(1)\delta\Psi(2)\rangle , \tag{3.1}$$

where $\delta\Psi(1)=\Psi(1)-\langle\Psi(1)\rangle$. The quantity $Z_U[\psi]$ is defined in terms of the action A_U by

$$Z_U[\psi]=\int D(\Psi)e^{-A_U[\Psi]} . \tag{3.2}$$

The action A_U can in general be obtained as a polynomial of the fields ψ_α. The construction of the action A_U for a given set of stochastic dynamical equations is discussed in detail in Ref. [5]. The linearized equations of motion give rise to a quadratic or Gaussian action $A_U^0[\Psi]$. Here we assume isotropic symmetry and separate the different correlation functions into their longitudinal and transverse parts as

$$G_{\psi_\alpha^i\psi_\beta^j}(\mathbf{q},\omega)=\hat{q}_i\hat{q}_j G^L_{\psi_\alpha\psi_\beta}(\mathbf{q},\omega)+(\delta_{ij}-\hat{q}_i\hat{q}_j)G^T_{\psi_\alpha\psi_\beta}(\mathbf{q},\omega) , \tag{3.3}$$

where $\hat{q}$ is the unit vector in the direction of $\mathbf{q}$. As argued in Sec. II, our focus here is on the longitudinal part of the correlation functions between the hydrodynamic

modes since this contains the relevant variable $c(\mathbf{x})$. The Gaussian action together with the so-called current term proportional to $U(1)$ can be formally written as

$$A_U^0[\psi]\equiv\sum_{1,2}\tfrac{1}{2}\Psi(1)G_0^{-1}(1,2)\Psi(2)-\sum_1\Psi(1)U(1)\ . \tag{3.4}$$

The inverse matrix G_0^{-1} can be split into longitudinal and transverse parts as described in Eq. (3.3) and our focus here is on the former. We follow the procedure for constructing G_0^{-1} described in Ref. [5], using the following form for the longitudinal part of the equations of motion, in the $(\mathbf{q},\omega)$ space:

$$\begin{aligned}
&\omega\rho-qg_l=0\ ,\\
&\omega g_l-qc_0^2\delta\rho-\frac{\vartheta'}{\rho_0}qc+iL_0\nu_l=\theta_l\ ,\\
&(\omega+i\Gamma_0q^2)c-i\Gamma_0'q^2\delta\rho-qg_l+q\rho_0\nu_l=\bar{f}_l\ ,\\
&g_l-\rho_0v_l=0\ ,
\end{aligned} \tag{3.5}$$

where the subscript l denotes the longitudinal part and $c_0^2=(A+\vartheta')/\rho_0$ is the sound speed. The correlation function in the Gaussian theory, denoted by a subscript 0, satisfies the trivial relation

$$\sum_\gamma[G_0^{-1}(\mathbf{q},\omega)]_{\alpha\gamma}[G_0(\mathbf{q},\omega)]_{\gamma\beta}=\delta_{\alpha\beta}\ , \tag{3.6}$$

where we have dropped the superscript L for the sake of brevity. The G_0^{-1} matrix can be identified from the action functional (3.5) through the definition given in Eq. (3.4). In general inversion of this 8×8 matrix will be rather involved, but the task here is simpler due to the fact $[G_0^{-1}]_{\alpha\beta}=0$ if α and β are both uncaretted variables. The inverted matrix thus splits into blocks with the part $[G_0]_{\hat{\alpha}\hat{\beta}}=0$. The zeroth order response functions $G^0_{\psi\hat{\psi}}$ thus obtained are given in Table I. The results for the zeroth order correlation functions $G^0_{\psi\psi}$ are listed in Table II in terms of the three quantities F_1, F_2, and F_3 defined as

$$\begin{aligned}
F_1&=(\omega^2+\Gamma_0^2q^4)L_0+\vartheta'\Gamma_0'q^4\ ,\\
F_2&=\bar{\Gamma}_0[L_0q^4(\omega^2L_0+\vartheta'\Gamma_0'q^2)+\rho_0^2(\omega^2-q^2c_0^2)^2]\ ,\\
F_3&=\Gamma_0'q^2[L_0q^4\Gamma_0-\rho_0(\omega^2-q^2c_0^2)]\ .
\end{aligned} \tag{3.7}$$

The poles in the correlation functions [30] are signatures of the hydrodynamic modes in the system. Thus zeros of the denominator D_0 give the dispersion relations for the longitudinal sound modes in the solid. However, now there is also an extra diffusive mode given by the dispersion relation

$$\omega=-iq^2\Gamma_0\ , \tag{3.8}$$

which corresponds to the vacancy diffusion mode. Similarly the poles of the transverse part of the correlation functions show the existence of the transverse sound modes [25].

Corrections to the Gaussian theory due to the nonlinearities in the equations of motion can be obtained by using the standard Feynman graph techniques. The full nonlinear equations given by (2.13) cannot in general be written in terms of the density fluctuations and the c fields only; rather it will involve the couplings to the transverse component of $\mathbf{u}$ as well. In the present calculation we will ignore such couplings to the transverse fields since the vacancy diffusion is primarily represented through the c field. A more rigorous approach, however, will necessitate taking into account the coupling to the transverse parts of $\mathbf{u}$. In the present analysis our treatment of the nonlinearities in the equations of motion will be nonperturbative and hence the explicit form of the nonlinear vertex functions is not considered here. The inverse of the matrix containing the full Green's functions with the nonlinear equations of motion included in the action can be expressed in terms of the self-energy matrix Σ as

$$G^{-1}(12)=G^{0-1}(12)-\Sigma(12)\ . \tag{3.9}$$

TABLE I. The zeroth-order matrix $G^0_{\alpha\beta}$ for the response function. $\bar{c}_0^2=A/\rho_0$ and $D_0=(\omega+i\Gamma_0q^2)[\rho_0(\omega^2-q^2c_1^2)+i\omega L_0q^2]+i\vartheta'\Gamma_0'q^4$.

	$\hat{\rho}$	$\hat{g}_l$	$\hat{v}_l$	$\hat{c}$
ρ	$\dfrac{(\omega+iq^2\Gamma_0)(\rho_0\omega+iq^2L_0)}{D_0}$	$\dfrac{\rho_0q(\omega+iq^2\Gamma_0)}{D_0}$	$\dfrac{q^3\left[L_0(\omega+iq^2\Gamma_0)+i\dfrac{\vartheta'}{\rho}\right]}{D_0}$	$\dfrac{-q^2\vartheta'}{D_0}$
g_l	$\dfrac{q[\rho_0c_1^2(\omega+iq^2\Gamma_0)-iq^2\vartheta'\Gamma_0']}{D_0}$	$\dfrac{\rho_0\omega(\omega+iq^2\Gamma_0)}{D_0}$	$\dfrac{\omega q^2\left[L_0(\omega+iq^2\Gamma_0)+i\dfrac{\vartheta'}{\rho_0}\right]}{D_0}$	$\dfrac{-\omega q\vartheta'}{D_0}$
ν_l	$\dfrac{q\left[c_1^2(\omega+iq^2\Gamma_0)-iq^2\Gamma_0'\dfrac{\vartheta'}{\rho_0}\right]}{D_0}$	$\dfrac{\omega(\omega+iq^2\Gamma_0)}{D_0}$	$\dfrac{1}{D_0}\left[\dfrac{B}{\rho_0}\Gamma_0'q^4+i(\omega+iq^2\Gamma_0)(\omega^2-q^2\bar{c}_0^2)\right]$	$\dfrac{-\omega q\vartheta'/\rho_0}{D_0}$
c	$\dfrac{iq^2\Gamma_0'(\rho_0\omega+iq^2L_0)}{D_0}$	$\dfrac{iq^3\Gamma_0'\rho_0}{D_0}$	$\dfrac{q}{D_0}[L_0q^2(\omega+iq^2\Gamma_0')-i\rho_0(\omega^2-q^2c_0^2)]$	$\dfrac{\rho_0(\omega^2-q^2c_0^2)+iq^2L_0\omega}{D_0}$

TABLE II. The zeroth-order matrix $G^0_{\alpha\beta}$ for the correlation functions, where the expressions for F_1, F_2, and F_3 are listed in Eq. (3.7).

	ρ	g_l	v_l	c
ρ	$\frac{2\beta^{-1}q^4F_1\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q^3\omega F_1\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q^3\omega F_1\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}q^2F_3\rho_0^2}{D_0D_0^*}$
g_l	$\frac{2\beta^{-1}q^3\omega F_1\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q^2\omega^2F_1\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q^2\omega^2F_1\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}q\omega F_3\rho_0^2}{D_0D_0^*}$
v_l	$\frac{2\beta^{-1}q^3\omega F_1\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}q^2\omega^2F_1\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}q^2\omega^2F_1}{D_0D_0^*}$	$\frac{2\beta^{-1}q\omega F_3\rho_0}{D_0D_0^*}$
c	$\frac{2\beta^{-1}q^2\omega F_3\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q\omega F_3\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}q\omega F_3\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}\bar{\Gamma}_0q^2F_2\rho_0^2}{D_0D_0^*}$

A graphical expansion for Σ as a power series in the nonlinear vertices in the equations of motion that generates the non-Gaussian terms in the action can be obtained. In Ref. [5] such an expansion to lowest order was used to obtain some quantitative results about how much slowing down can occur in the relaxation behavior due to the mode coupling effect treated in a self-consistent manner. Our main goal here is to investigate the implications for the diffusive mechanism by which density autocorrelation decays in the long time limit in the supercooled liquid. The role of the self-energy matrix is to renormalize the transport coefficients that appear in the linear theory. Due to the large number of fields in the present analysis the structure gets quite complicated. Following Ref. [5] we will use a fluctuation dissipation result as well as the isotropy of the system over long length scales to investigate how much the different self-energies interrelate with each other and how they contribute to the renormalization of the transport coefficients.

IV. NONPERTURBATIVE ANALYSIS

We make use of a special relation between the response functions $G_{\psi\hat{\psi}}$ and the correlation functions $G_{\psi\psi}$ to simplify the structure of the theory and demonstrate how it can be renormalized by using proper self-energy functions . Following Ref. [5], in the present case, with the extended set of slow modes the same fluctuation-dissipation relation remains valid. Thus for any uncaretted variable α we have

$$G_{v\alpha}(\mathbf{q},\omega)=-2\beta^{-1}\mathrm{Im}[G_{\hat{g}\alpha}(\mathbf{q},\omega)] \ . \tag{4.1}$$

At the zeroth order this relation can be easily checked using Tables I and II. In order to investigate the consequences of the nonlinearities we consider the full Green's functions $G_{\alpha\beta}$. This involves inverting the G^{-1} matrix obtained using Eq. (3.9). Similar to the zeroth order case, the correlation function $G_{\hat{\psi}\hat{\psi}}$ between the caretted fields vanishes [31] and the response functions $G_{\psi\hat{\psi}}$ can be suitably expressed in the form

$$G_{\alpha\hat{\beta}}(\mathbf{q},\omega)=\frac{N_{\alpha\hat{\beta}}}{D} \ . \tag{4.2}$$

The denominator D is obtained as

$$D=(\omega+iq^2\Gamma_R)[\rho_0(\omega^2-q^2c_R^2)+iq^2L_R(\omega+iq\Sigma_{\hat{\gamma}\rho})] +iq^2(\Gamma_0'q^2-q\Sigma_{\hat{\gamma}\rho})(\vartheta_R\rho_0-L_R\Sigma_{\hat{\gamma}c}) \ , \tag{4.3}$$

where the subscript R on the various symbols generally refers to the corresponding renormalized quantities

$$q\Gamma_R(q,\omega)=q\Gamma_0+\Sigma_{\hat{\gamma}c}(q,\omega) \ , \tag{4.4}$$

$$q^2L_R(q,\omega)=q^2L_0+i\Sigma_{\hat{g}\hat{v}}(q,\omega) \ , \tag{4.5}$$

$$qc_R^2(q,\omega)=qc_0^2-\Sigma_{\hat{g}\rho}(q,\omega) \ , \tag{4.6}$$

$$q\vartheta_R(q,\omega)=q\vartheta'-\Sigma_{\hat{g}c}(q,\omega) \ . \tag{4.7}$$

The numerator N is a 4×4 matrix involving the various elements of the matrix Σ. It satisfies the general relation $N_{\alpha\hat{\beta}}=(N_{\hat{\beta}\alpha})^*$, where

$$G_{\hat{\beta}\alpha}(\mathbf{q},\omega)=\frac{N_{\hat{\beta}\alpha}}{-D^*} \ . \tag{4.8}$$

We list in Appendix B the elements of the $N_{\alpha\hat{\beta}}$ matrix that are useful for the analysis of the fluctuation dissipation relations discussed below. The correlation functions $G_{\alpha\beta}$ for the hydrodynamic fields can be expressed in the form

$$G_{\alpha\beta}=-G^{-1}_{\alpha\hat{\gamma}}C_{\hat{\gamma}\hat{\alpha}}G^{-1}_{\hat{\alpha}\beta} \ , \tag{4.9}$$

where the summations of the repeated variables are over the set $\{\rho,\mathbf{g},\mathbf{v},c\}$ and the matrix C is given by

$$C_{\hat{\alpha}\hat{\beta}}=2k_BT[L_0\delta_{\alpha\hat{g}}\delta_{\beta\hat{g}}+\delta_{\alpha\hat{c}}\delta_{\beta_{\hat{c}}}\hat{\Gamma}_0\rho_0^2]q^2-\Sigma_{\hat{\alpha}\hat{\beta}} \ . \tag{4.10}$$

The matrix $\Sigma_{\hat{\alpha}\hat{\beta}}=0$, if $\hat{\alpha}$ or $\hat{\beta}$ equal to $\hat{\rho}$. This is because the continuity equation do not contain any nonlinear term in it. In order to simplify the analysis we also consider the dynamical equation for the variable $c(\mathbf{x},t)$ in the linear form. This is reasonable since our main goal here is to investigate the relevance of the hydrodynamic diffusive mode for correlation of density relaxation to the motion of vacancies in the amorphous solid through the

dynamics of $c(\mathbf{x},t)$. With this we have $\Sigma_{\hat{\alpha}\hat{\beta}}=0$ if either of the caretted indices is equal to $\hat{c}$. In Appendix A we show that using Eqs. (4.9) and (4.2) leads to the following form of the fluctuation-dissipation relation (4.1):

$$i\beta N_{\nu\hat{\alpha}}C_{\hat{\alpha}\hat{\nu}}=[D\delta_{\hat{\nu}\hat{g}}+N_{\beta\hat{g}}G^{-1}{}_{\beta\hat{\nu}}] . \tag{4.11}$$

For $\nu=\hat{g}$ and $\hat{v}$ the relation (4.11) reduces to Eqs. (6.38)–(6.40) obtained in Ref. [5] for the set of conserved variables $\{\rho,\mathbf{g},\mathbf{v}\}$ in the case of isotropic liquid. Here we use Eq. (4.11) to obtain some useful relations between elements of the matrix Σ. Although, in general, these relations are very complicated for the arbitrary value of the wave vector q and frequency ω, some simple results can be obtained in the hydrodynamic limit of small q and ω.

Hydrodynamic limit

First, we extract certain explicit factors of the wave number from the various self-energy matrices using conservation laws and symmetry,

$$\Sigma_{\hat{g}v}=-iq^2\gamma_{\hat{g}v} , \tag{4.12}$$

$$\Sigma_{\hat{g}\hat{g}}=-q^2\gamma_{\hat{g}\hat{g}} , \tag{4.13}$$

$$\Sigma_{\hat{g}\rho}=q\gamma_{\hat{g}\rho} , \tag{4.14}$$

$$\Sigma_{\hat{v}\rho}=q\gamma_{\hat{v}\rho} , \tag{4.15}$$

$$\Sigma_{\hat{v}c}=q\gamma_{\hat{v}c} , \tag{4.16}$$

$$\Sigma_{\hat{g}c}=q\gamma_{\hat{g}c} . \tag{4.17}$$

Using these results and the expressions for the different elements of the $N_{\psi\hat{\psi}}$ matrix listed in Appendix B in the relation (4.11) we obtain a set of interrelations between the self-energies in the hydrodynamic limit. Thus, for $\nu=\hat{g}$ in Eq. (4.11), we obtain the relation useful for the renormalization of the viscosity,

$$\gamma_{\hat{g}\hat{g}}(0,0)-2\beta^{-1}\gamma'_{\hat{g}v}(0,0)=\lim_{\omega\to 0}\left[2\beta^{-1}\frac{\gamma''_{\rho\hat{g}}(0,\omega)}{\omega}\right] , \tag{4.18}$$

where the single and double primes, respectively, stand for the real and imaginary parts. From Eq. (4.5) it then follows that the viscosity L_R can be renormalized by analyzing the self-energy $\Sigma_{\hat{g}\hat{g}}$. At the one loop order this gives rise to the well-known quadratic [5] term involving a convolution of the density correlation functions essential for the feedback mechanism causing the dynamic instability in the mode coupling model. The relation between $\gamma_{\rho\hat{v}}$ and $\Sigma_{\hat{v}\hat{v}}$ remains the same as that found in Ref. [5], i.e.,

$$C_{\hat{v}\hat{v}}(0,0)=-2\beta^{-1}\rho_0\gamma'_{\rho\hat{v}}/c^2 , \tag{4.19}$$

with c being the renormalized sound speed in the hydrodynamic limit. This is also in agreement with the one-loop results found by Kim [25] for the cutoff function in similar circumstances. Setting $\hat{v}=\hat{c}$ in the relation Eq. (4.11) and taking the hydrodynamic limit we obtain the following relation for the self-energy $\Sigma_{\hat{v}c}$ that renormalizes the diffusion constant Γ_0 in Eq. (4.3):

$$\gamma_{\hat{v}c}(0,0)=\frac{B}{\rho_0c^2}\gamma_{\hat{v}\rho}(0,0) , \tag{4.20}$$

together with

$$\gamma'_{\hat{g}c}(0,0)=0 . \tag{4.21}$$

In the present context Eq. (4.20) is the important new relation that gives the renormalization of the bare transport coefficient Γ_0 due to the nonlinear couplings in the hydrodynamic equations for the density and current fluctuations. The self-energy matrix element $\gamma_{\hat{v}\rho}$ is responsible for the diffusive mode that restores ergodicity in the supercooled liquid [5]. We explore the consequences of this further in the next section.

V. THE GLASS INSTABILITY AND ERGODICITY-RESTORING MECHANISM

In the present section we discuss how the diffusive process that cuts off the sharp transition predicted in the simple mode coupling model can be interpreted through the introduction of th slow variable $c(\mathbf{x},t)$ which corresponds to vacancy diflusion in the amorphous solid. To analyze the long time behavior of the density correlation function we note that in the small q and ω limit the response function $G_{\rho\hat{\rho}}$ is obtained as

$$G_{\rho\hat{\rho}}(q,\omega)=\frac{(\omega+iq^2L_R)(\omega+iq^2\Gamma_R)}{(\omega+iq^2\Gamma_R)[\rho_0(\omega^2-q^2c_R^2)+iq^2L_R(\omega+iq^2\gamma)]+i\Delta q^4} , \tag{5.1}$$

where $\Delta=\vartheta'(\Gamma'_0-\gamma_{\hat{v}\rho})$ and $\gamma\equiv\gamma_{\rho\hat{v}}$. The renormalized viscosity can be obtained in the form

$$L_R(\mathbf{q},\omega)=L_0+\int_0^\infty dt\, e^{izt}L_{\mathrm{MC}}(\mathbf{q},t) , \tag{5.2}$$

where L_{MC} denotes the feedback to the viscosity of the supercooled liquid from the slowly decaying mode coupling contribution. Now, as the system becomes denser the viscosity L_R becomes very large due to very slow decay of L_{MC}. At first, if we ignore the contributions coming from $\gamma_{\rho\hat{v}}$, then for a fixed wave number, the frequency ω becomes small compared to L_Rq^2 at high enough density. It follows easily then that $G_{\rho\hat{\rho}}$ tends to develop a $1/\omega$ type frequency dependence. This slows down the relaxation and hence the mode coupling contribution to the viscosity for intermediate time becomes large. However, it was pointed out in Ref. [5] on retaining the nonhydrodynamic correction due to γ that the density correlation

in the long time limit decays through a diffusive mode. In the present analysis, due to the presence of the term proportional to Δ in the denominator of Eq. (5.1), the quantity γ has an additional contribution, thus obtaining

$$\bar{\gamma}=\gamma+\frac{\Delta}{(\omega+q^2\Gamma_R)\tilde{L}_R(q,\omega)}\ , \tag{5.3}$$

where $\tilde{L}_R$ is the singular part of L_R due to the mode coupling contribution. In the present theoretical framework the quantity Γ_0' is an external parameter and in general the correction due to inclusion of the extra diffusion mode can be complicated. We propose here the simpler ansatz $\Delta=0$, which is equivalent to

$$\gamma_{\hat{v}_\rho}(0,0)=\Gamma_0'\ , \tag{5.4}$$

in which case the density correlation function has the long time diffusive behavior

$$G_{\rho\hat{\rho}}=\frac{1}{\omega+iq^2\gamma} \tag{5.5}$$

while the dynamic correlation of the slow variable $c(\mathbf{x},t)$ relaxes through a diffusive mechanism, the response function $G_{c\hat{c}}$ being given by

$$G_{c\hat{c}}=\frac{1}{\omega+iq^2\Gamma_R}\ . \tag{5.6}$$

Here the quantity Γ_R is the renormalized diffusion constant related to the motion of the vacancies in the system. Comparing Eqs. (4.20), (4.4), and (5.4) we obtain

$$\Gamma_R=\gamma\left[1+\frac{B}{\vartheta'}+\frac{B}{\rho_0 c^2}\right]\ . \tag{5.7}$$

For small B, i.e., weak coupling between the mass density and the defect density c, the time scales of the two diffusive processes are comparable. This simple ansatz thus provides possible insight into the ergodicity restoring process in the supercooled amorphous state. Note that the result (5.5) has been derived under the assumption that vacancy diffusion is the only ergodicity-restoring process. Therefore the corresponding relaxation strength appears to be one. Taking the hopping processes into account will reduce this strength.

VI. DISCUSSION

In the present work we have extended the fluctuating hydrodynamic description for the fluid to include the displacement field $\mathbf{u}$ reflecting the freezing of the local structures in the amorphous solid. This represents the displacements of the particles from their average positions in the metastable state. We work with an effective Hamiltonian for the system that is compatible with the elastic properties of the amorphous solid. The full set of fluctuating nonlinear equations for the set of hydrodynamic variables including u has been obtained. The symmetry of the stress energy tensor ensures the angular momentum conservation. The role of $\mathbf{u}$ is manifested through its longitudinal part, i.e., $\nabla\cdot\mathbf{u}$, in terms of which we define the variable $c(\mathbf{x})$ that follows a diffusive dynamics. Our focus here is on the diffusion of the free volume or vacancies signifying configurational rearrangements in the amorphous solid. Although the u fields signify the freezing on a local length scale, isotropy is assumed for the system for long length scales. The analysis here shows that one recovers the earlier results [5] obtained by Das and Mazenko for mode coupling models of glass transition if the time scales associated with the diffusion of density fluctuations and that of the vacancy diffusion are comparable. It should be pointed out here that the quantity γ is obtained in the analysis beyond the linear level. Thus to the lowest order it is explicitly of $O(k_BT)$. On the other hand, in the present paper we demonstrate that the associated time scales can be interpreted in a consistent manner through (5.4). It also means that the diffusion constant $\bar{\Gamma}$ will tend to vanish as T approaches zero.

In the present analysis we demonstrate how the diffusive mode which also restores ergodicity in the mode coupling models can be linked with the diffusion of vacancies or free volumes in the amorphous solids. Indeed the introduction of the displacement field $\mathbf{u}$ in the case of the amorphous solid requires the reference to a rigid lattice and, on the other hand, the ergodicity-restoring process in the system invalidates the existence of any such rigid structure. The crossover between these two situations requires a self-consistent treatment with possible dynamic connections between the elastic and viscous behavior of the system. Here we take the simpler approach where effect of the local displacement field $\mathbf{u}$ is manifested through its longitudinal part only, i.e., $(-\nabla\cdot\mathbf{u})$, which in the case of a strictly rigid lattice will be simply related to the density fluctuations. Thus $\mathbf{u}$ is not an order parameter in the amorphous solid and translational symmetry is maintained over long length scales. $(-\nabla\cdot\mathbf{u})$, which is well defined on not too large time and length scales, is just used to define the vacancy concentration $c(\mathbf{x},t)$ via (2.19). It is assumed that the equations for ρ, $\mathbf{g}$, and c from (2.20) constitute a reasonable set of equations incorporating vacancy diffusion at least below T_c.

Let us finally stress again that the diffusive type of the ergodicity-restoring process by vacancy diffusion has not been experimentally found in the immediate neighborhood of T_c up to today. This could be due to a small relaxation strength. Nevertheless we think that vacancy diffusion could play a role for the relaxation in glasses, maybe at lower temperatures compared to T_c.

ACKNOWLEDGMENTS

S.P.D. acknowledges the hospitality and financial support of the Institute of Physik in Mainz. R.S. gratefully acknowledges helpful discussions with G. Götze.

APPENDIX A

Here we prove the form given in Eq. (4.11) for the fluctuation-dissipation relation. We start from the general identity

$$\int D(\psi)\frac{\delta}{\delta\psi_{\hat{\alpha}}(1)}[\psi_{\hat{\beta}}(2)e^{-A}]=0\ . \tag{A1}$$

Noting that the variable $c(\mathbf{x},t)$ behaves in the same way as $\delta\rho(\mathbf{x},t)$ under time reversal, the steps given in Appendix B of Ref. [5] follow here as well and the same result

$$G_{V\alpha}(\mathbf{q},\omega)=-2\beta^{-1}\mathrm{Im}[G_{\hat{g}\alpha}(\mathbf{q},\omega)] \tag{A2}$$

holds in the present case. Now, using Eqs. (4.9) and (4.2) in this relation obtains

$$N_{v\hat{\alpha}}C_{\hat{\alpha}\hat{\nu}}=\frac{2\beta^{-1}}{D^*}\mathrm{Im}[N_{\hat{g}\beta}D]G^{-1}_{\beta\hat{\nu}}$$
$$=\frac{2\beta^{-1}}{D^*}\frac{1}{2i}[N_{\hat{g}\beta}D-N^*_{\hat{g}\beta}D^*]G^{-1}_{\beta\hat{\nu}}\ . \tag{A3}$$

However, from the definition of the matrix $N_{\psi\hat{\psi}}$ it follows that

$$N_{\hat{g}\beta}G^{-1}_{\beta\hat{\nu}}=C_{\beta\hat{g}}[G^{-1}]_{\beta\hat{\nu}}=\delta_{\hat{\nu}\hat{g}}D^*\ , \tag{A4}$$

where $C_{\beta\hat{g}}$ denotes the cofactor of the $(\beta\hat{g})$th element of the matrix G^{-1}. Using this in Eq. (A3), the result (4.11) easily follows. This result is true independent of the detailed nature of the fields as long as the relation (4.1) holds.

APPENDIX B

Here we list the necessary elements of the $N_{\alpha\hat{\beta}}$ matrix that are used in the analysis of Secs. IV and V:

$$N_{\rho\hat{\rho}}=(\omega+iq^2\Gamma_R)[\rho_0\omega+iq^2L_R]\ , \tag{B1}$$

$$N_{\rho\hat{g}}=\rho_0 q(\omega+iq^2\Gamma_R)\ , \tag{B2}$$

$$N_{g\hat{g}}=\rho_0\omega(\omega+iq^2\Gamma_R)\ , \tag{B3}$$

$$N_{v\hat{g}}=(\omega+iq^2\Gamma_R)(\omega+iq\Sigma_{\hat{v}\rho})+q\Sigma_{\hat{v}c}(q\Sigma_{\hat{v}\rho}-\Gamma_0')\ , \tag{B4}$$

$$N_{c\hat{g}}=-iq\rho_0(q\Sigma_{\hat{v}\rho}-\Gamma_0')\ , \tag{B5}$$

$$N_{v\hat{c}}=i(\omega^2-q^2c_R^2)\Sigma_{\hat{v}c}-q\vartheta'(\omega+iq\Sigma_{\hat{v}\rho})\ , \tag{B6}$$

$$N_{v\hat{v}}=i(\omega+iq^2\Gamma_R)(\omega^2-q^2c_R^2)+iq^2\vartheta'(\rho_0\omega+iq^2\Gamma_0')\ . \tag{B7}$$

[1] U. Bengtzelius, W. Götze, and A. Sjölander, J. Phys. C **17**, 5915 (1984).
[2] E. Leutheusser, Phys. Rev. A **29**, 2765 (1984).
[3] S. P. Das, G. F. Mazenko, S. Ramaswamy, and J. Toner, Phys. Rev. Lett. **54**, 118 (1985).
[4] W. Götze, in *Liquids, Freezing and the Glass Transition,* edited by D. Levisque, J. P. Hansen, and J. Zinn-Justin (Elsevier, New York, 1991); W. Götze and L. Sjögren, Rep. Prog. Phys. **55**, 241 (1992); B. Kim and G. F. Mazenko, Adv. Chem. Phys. **78**, 129 (1990); R. Schilling, in *Disorder Effects on Relaxational Processes,* edited by R. Richert and A. Blumen (Springer, New York, 1994).
[5] S. P. Das and G. F. Mazenko, Phys. Rev. A **34**, 2265 (1986).
[6] P. C. Martin, E. D. Siggia, and H. A. Rose, Phys. Rev. A **8**, 423 (1973); U. Deker and F. Haake, *ibid.* **11**, 2043 (1975); R. V. Jensen, J. Stat. Phys. **25**, 183 (1981).
[7] G. F. Mazenko and J. Yeo (unpublished).
[8] R. Schmitz, J. W. Dufty, and P. De, Phys. Rev. Lett. **71**, 2066 (1993).
[9] S. P. Das, Phys. Rev. A **36**, 211 (1987); **42**, 6116 (1990).
[10] J. J. Ullo and S. Yip, Phys. Rev. Lett. **54**, 1509 (1985); Phys. Rev. A **39**, 5877 (1989).
[11] L. Sjögren, Z. Phys. B **79**, 5 (1990).
[12] W. Götze and L. Sjögren, Z. Phys. B **65**, 415 (1987).
[13] R. D. Mountain, J. Res. Natl. Bur. Stand. Sect. A **70**, 207 (1966).
[14] M. Fuchs and A. Latz, J. Chem. Phys. **95**, 7074 (1991).
[15] M. Fuchs, W. Götze, S. Hildebrand, and A. Latz, J. Phys. Condens. Matter **4**, 7709 (1992); H. Z. Cummins, W. Du, M. Fuchs, W. Götze, S. Hildebrand, A. Latz, G. Li, and N. Tao, Phys. Rev. E **47**, 4223 (1993).
[16] M. H. Cohen and G. S. Grest, Phys. Rev. B **20**, 1077 (1979).
[17] P. C. Martin, O. Parodi, and P. S. Pershan, Phys. Rev. A **6**, 2401 (1972).
[18] P. D. Fleming III and C. Cohen, Phys. Rev. B **13**, 500 (1976).
[19] C. Cohen, P. D. Fleming III, and J. H. Gibbs, Phys. Rev. B **13**, 866 (1976).
[20] G. F. Mazenko, S. Ramaswamy, and J. Toner, Phys. Rev. Lett. **49**, 51 (1982); Phys. Rev. A **28**, 1618 (1983).
[21] P. De, R. A. Pelcovits, E. Vogel, and J. Vogel, Phys. Rev. E **47**, 1824 (1993).
[22] H. van Beijeren and M. H. Ernst, Physica **68**, 437 (1973).
[23] T. R. Kirkpatrick, S. P. Das, M. H. Ernst, and J. Piacheski, J. Chem. Phys. **92**, 3768 (1990).
[24] M. H. Ernst and G. Szammel, Phys. Rev. B **48**, 112 (1993).
[25] B. Kim, Phys. Rev. A **46**, 1992 (1992).
[26] S. K. Ma and G. F. Mazenko, Phys. Rev. B **11**, 4077 (1975).
[27] I. E. Dzyloshinskii and G. E. Volvick, Ann. Phys. (N.Y.) **125**, 67 (1980).
[28] J. Langer and L. Turski, Phys. Rev. A **8**, 3230 (1973).
[29] ρ_0 is the quiescent (equilibrium) density for the system and in principle it should be included in the theory in a self-consistent manner.
[30] D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, Reading, MA, 1975).
[31] Since the response functions are time ordered and the diagrams for the self-energy elements with both the indices caretted contain a close loop of response functions, the latter vanish due to causality.