PHYSICAL REVIEW A VOLUME 34, NUMBER 3 SEPTEMBER 1986

Fluctuating nonlinear hydrodynamics and the liquid-glass transition

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(Received 23 December 1985)

We study the fluctuating nonlinear hydrodynamics of compressible fluids. Development of the appropriate field-theoretical description for this problem requires treatment of nonlinearities which arise through the relationship $\mathbf{g}=\rho\mathbf{V}$, where $\mathbf{g}$ is the momentum density, ρ is the mass density, and $\mathbf{V}$ is the velocity field. We show how this constraint can be naturally included in a field theory of the Martin-Siggia-Rose type. We analyze the structure of the resulting field theory using the available fluctuation-dissipation theorem. We also develop the perturbation-theory expansion in powers of the temperature and evaluate the contributions from the nonlinearities to one-loop order. We show that the theory is renormalizable in the hydrodynamic limit. This field-theoretical model is used to systematically investigate the origins and viability of the nonlinear density feedback mechanism first identified by Leutheusser as a source of the liquid-glass transition. While we find that the nonlinear couplings driving this mechanism are present, we also find contributions, arising from the nonlinear constraint relating $\mathbf{g}$, ρ, and $\mathbf{V}$, which cut off the mechanism. The cutoff arises from a nonhydrodynamic correction not treated in previous work. While we find that there is no sharp transition, we do find evidence for a rounded version of the transition.

I. INTRODUCTION

The development of an understanding of the nature of the liquid-glass transition has been elusive. This is not surprising considering the nonequilibrium nature of the problem and the very strongly varying time scales. There have been suggestions that glassy relaxation is universal.[1] More recent experimental evidence[2] suggests that glassy relaxation should be classified along a continuum ranging from "weakly" to "strongly" coupled glasses. The weakly coupled systems[3] show a much weaker power-law temperature dependence for the viscosity than the more strongly coupled systems[4] which show an Arrhenius behavior. The underlying physical distinctions among these systems are not yet understood. There appears to be some evidence[5] that some of the more weakly coupled systems correspond to simpler, structurally less hindered systems. In this paper we use fluctuating nonlinear hydrodynamics to investigate the nature of the glass transition in these simpler systems. Our main new result is that the dynamical transition predicted in earlier work[6–9] is eventually cut off when a careful analysis is carried out for the full nonlinear problem.

Recently hope has been raised that a theory of the glass transition in simple systems is evolving. The first step in this evolution was the bold step by Leutheusser[6] of assuming that a very simple model, abstracted from previous very involved work in kinetic theory,[10] could be applied to a very dense and low-temperature fluid. A similar model was developed independently by Bengtzelius *et al.*[7] Analysis of this model indicates that it has an intrinsically dynamic transition at higher densities with many of the characteristics of the liquid-gas transition. While transport properties vary strongly near the transition, the thermodynamic properties are insensitive to the transition. Thus the viscosity diverges as one approaches the transition, while the structure factor and the isothermal compressibility are very insensitive to the transition. The large viscosity is only one of several features of this transition which identifies it with glass formation. One also finds that transverse sound propagates in the "glass," and there is strong viscoelastic behavior.

While this model is extremely suggestive (and some of the results appear to be quantitative[7]), it has no firm theoretical basis and one does not know how to investigate for corrections. In Ref. 8 it was suggested that the model developed in Refs. 6 and 7 could be understood in terms of the nonlinear fluctuating hydrodynamics (NFH) governing compressible fluids. Let us first make clear the point that the glass transition is not intrinsically a hydrodynamic—long-distance and long-time—phenomenon. As one approaches the glass transition the freezing occurs first at small or intermediate length scales. However, the effects of freezing should eventually propagate out to scales[11] governed by hydrodynamics. Thus the hydrodynamic regime should be influenced by the glass transition. At least the hydrodynamic regime will be pushed out to much longer distances and times. An important question is whether, as one approaches the liquid-glass transition, the hydrodynamic equations must be supplemented through the introduction of additional "slow" variables (like for example the staggered magnetization in an antiferromagnet[12] as one approaches the Néel transition or the layer field displacement in a smectic-A crystal[13]) or whether the freezing is reflected in the equations of NFH without modification. The identification of any new slow variable is more subtle than in previous examples since one expects any such variable will not show any strong equilibrium behavior as one approaches the glass transition and there will, therefore, be no associated symmetry change in the system. While it seems likely that there are additional variables which might be important in more complicated systems,[14] we restrict ourselves here to the simplest case where we include only the hydrodynamic variables and ask whether this simplest description is sensitive to the hindered dynamics appropriate to the glass transition.

In this paper we present the detailed calculations following up on the work reported in Ref. 8. We find some significant differences from the conclusions drawn there. These differences arise from a more careful treatment of the model developed in Ref. 8. A full appreciation of the origins of our new results requires considerable formal development:

(i) In the next section we construct the equations of motion appropriate for the fluctuating nonlinear hydrodynamics of compressible fluids in the absence of energy fluctuations. Our results are similar to those found previously by Enz and Turski.[15]

(ii) We develop the appropriate field theoretic formulation for this problem in Sec. III. This nontrivial step involves a generalization of the Martin-Siggia-Rose[16] (MSR) method to a case involving nonlinear constraints.

(iii) The results of Sec. II can be used to construct a formal perturbation theory expansion in powers of $k_B T$, where T is the temperature, for the correlation functions of interest.

Using these results we can investigate the influence of the nonlinearities on the transport properties of the system. Our main new results are:

(i) If one ignores all nonhydrodynamic corrections (terms of higher order in wave number) then one easily makes contact with the results of Ref. 8 and the earlier works in Refs. 6 and 7. In this case there is a sharp dynamic transition at a particular temperature or density where the viscosity diverges with a power-law behavior, the system becomes nonergodic for higher densities "in the glass" and the hydrodynamic structure of the system is modified significantly.

(ii) We have found, however, that there are nonhydrodynamic corrections, which have been ignored in previous work, which, when taken into account, cut off the sharp nature of the transition. The new picture, which results when a complete analysis is carried out, is that the system is ergodic for all values of the density and temperature and, if one goes to long enough length and time scales, hydrodynamics is recovered in its conventional form. However, the role of the Leutheusser mechanism is not insignificant. Density fluctuations do tend to drive the viscosity to larger values in a manner consistent with the experimental results of Taborek *et al.*[17] As the viscosity becomes large, however, one does not find, as speculated in the previous work,[6-8] that the Laplace transform of the density-density correlation function goes as $1/\omega$, where ω is a small frequency. Instead one finds, arising from nonhydrodynamic corrections, that the correlation function is proportional to $1/(\omega+i\gamma q^2)$. Thus one obtains a diffusive mode, the previous nonergodic behavior is destroyed and correlation functions decay to zero with a lifetime $\tau=(\gamma q^2)^{-1}$. It is important to realize that this result holds only outside the true hydrodynamic regime which has now been pushed to much longer times and distances.[18] We find, in keeping with previous findings,[6,7] that the longitudinal viscosity increases with a power-law behavior $\Gamma \sim (\lambda^* - \lambda)^{-2}$ for λ less than λ^* where λ^* is the critical value of the coupling in the Leutheusser transition. However, as λ approaches λ^* there is a crossover to a behavior $\Gamma \sim (\lambda - \lambda^*)$ which holds for λ greater than λ^*. Γ is smooth for λ near λ^*. This behavior is discussed in more detail in Sec. VIII.

In our analysis here we ignore the role of energy fluctuations. This does not mean that we believe there is nothing interesting[19] going on with the heat transport in systems near the glass transition. Rather, we believe that any strong dependences on the heat transport as one approaches the glass transition are driven by the density fluctuations. We intend to include the heat variable in future work.

While, from a formal point of view, we have been able to set up a sensible perturbation-theory expansion, from a practical point of view things are not completely satisfactory. As we shall see, the theory is more complicated than one might guess *a priori*. Indeed even at second order in $k_B T$ things are too complicated to be worked out in detail. More sobering is the fact that while the expansion is systematic, it is in terms of a dimensionless expansion parameter which is not small. Thus while things are under control in a formal sense, they are not controlled (quantitatively accurate) in a practical sense. Therefore, while our conclusions are consistent, they cannot be considered definitive.

Much attention has been paid to the Kauzman paradox[20] which is associated with the hypothetical temperature where the entropy of the supercooled liquid becomes lower than that of the associated crystal. It should be pointed out that the identification of such a temperature and its relationship to any glass transition temperature would require inclusion of terms in the effective Hamiltonian which could lead to crystallization. Since we assume crystallization can be avoided, we do not include such terms here, and can, therefore, make no estimate of this temperature in our model.

While we set up the equations of NFH rather generally in the next section, we restrict most of our analysis to the simplest possible situation where the effective Hamiltonian controlling the density fluctuations is quadratic and where there are no gradient terms. While this will simplify the analysis a great deal and we will be able to get a feeling for the structure of the theory, we do not believe this will give a good approximation for real dense liquids where the structure factor has substantial structure away from $q=0$. Indeed, as demonstrated by the work of Kirkpatrick,[9] one expects the slowness of fluctuations for wave numbers near the peak in the structure factor to enhance any instability. Thus a quantitative treatment of this problem must include a more realistic treatment of the wave-number dependence of the equilibrium correlations than we give here.

II. EQUATIONS OF NONLINEAR FLUCTUATING HYDRODYNAMICS FOR COMPRESSIBLE FLUIDS

While the Navier-Stokes equations[21] have been well established for over a century, the appropriate nonlinear equations in the presence of thermal noise have only been developed[22,23] recently. The case of incompressible fluids has been extensively studied. As we shall see, the generalization to compressible fluids introduces some new complexities which have not, to our knowledge, been dealt with previously.

The first stage in our analysis is straightforward and follows the development in Ma and Mazenko.[24] We assume that the set of slow variables includes the mass density $\rho(\mathbf{x})$ and the momentum density $\mathbf{g}(\mathbf{x})$. In principle, we should also include the energy density, but, for the reasons mentioned in the Introduction, we neglect it here. $\rho(\mathbf{x})$ and $\mathbf{g}(\mathbf{x})$ are components of a vector ψ_i (where i labels the type of field, the coordinate label, and the vector index on $\mathbf{g}$) which satisfy the generalized Langevin equation

$$\frac{\partial \psi_i}{\partial t}=\bar{V}_i[\psi]-\sum_j \Gamma_{ij}\frac{\delta F}{\delta \psi_j}+\Theta_i \ , \tag{2.1}$$

where $\bar{V}_i[\psi]$ is the "streaming velocity" representing the reversible part of the equation of motion and given by

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

and $\{\psi_i,\psi_j\}$ is the Poisson bracket between the "slow" variables (see following for a more careful definition). In (2.1) and (2.2) $F[\psi]$ is the effective Hamiltonian governing the equilibrium behavior of the ψ_i. Thus equilibrium averages of the fields ψ_i at equal times are given by

$$\langle \psi_i\psi_j\rangle=\int D(\psi)e^{-\beta F[\psi]}\psi_i\psi_j/Z \tag{2.3}$$

where

$$Z=\int D(\psi)e^{-\beta F[\psi]} \tag{2.4}$$

is the partition function, $\beta=(k_BT)^{-1}$, and $D(\psi)$ indicates a functional integral over the fields ψ_i. The Γ_{ij} in (2.1) form a "bare" damping matrix and the Θ_i are Gaussian noise sources satisfying

$$\langle \Theta_i(t)\Theta_j(t')\rangle=2k_BT\Gamma_{ij}\delta(t-t') \ . \tag{2.5}$$

Evaluation of the Poisson brackets in (2.2) means in practice[24,25] identifying the fields $\psi_i(\mathbf{x})$ with microscopic variables $\tilde{\psi}_i(\mathbf{x})$, evaluating the Poisson brackets in the usual way, and then replacing $\tilde{\psi}_i$ by the $\psi_i(\mathbf{x})$ in the result. For a classical fluid with N particles specified by the phase-space coordinates $\mathbf{R}_\alpha$ and $\mathbf{P}_\alpha$, $\alpha=1,2,\ldots,N$, the mass density is given by

$$\tilde{\rho}(\mathbf{x},t)=m\sum_{\alpha=1}^{N}\delta(\mathbf{x}-\mathbf{R}_\alpha(t)) \ , \tag{2.6}$$

assuming the particles have mass m, while the momentum density is given by

$$\tilde{g}_i(\mathbf{x},t)=\sum_{\alpha=1}^{N}P_\alpha^i(t)\delta(\mathbf{x}-\mathbf{R}_\alpha(t)) \tag{2.7}$$

where i is a vector label. It is straightforward to show that $\tilde{\rho}$ and $\tilde{g}$ satisfy the Poisson bracket relations

$$\{\tilde{\rho}(\mathbf{x}),\tilde{g}_i(\mathbf{x}')\}=-\nabla_{\mathbf{x}}^i[\delta(\mathbf{x}-\mathbf{x}')\tilde{\rho}(\mathbf{x})] \ , \tag{2.8}$$

$$\{\tilde{g}_i(\mathbf{x}),\tilde{\rho}(\mathbf{x}')\}=\nabla_{\mathbf{x}'}^i[\delta(\mathbf{x}-\mathbf{x}')\tilde{\rho}(\mathbf{x})] \ , \tag{2.9}$$

$$\{\tilde{g}_i(\mathbf{x}),\tilde{g}_j(\mathbf{x}')\}=-\nabla_{\mathbf{x}}^j[\delta(\mathbf{x}-\mathbf{x}')\tilde{g}_i(\mathbf{x})]+\nabla_{\mathbf{x}'}^i[\delta(\mathbf{x}-\mathbf{x}')\tilde{g}_j(\mathbf{x})] \ . \tag{2.10}$$

In order to complete the determination of the streaming velocities, we must specify the effective Hamiltonian $F[\psi]=F[\rho,\mathbf{g}]$. The momentum dependence of F is governed by the requirement that $\rho(\mathbf{x},t)$ satisfy the continuity equation

$$\frac{\partial \rho}{\partial t}=-\nabla\cdot\mathbf{g} \ . \tag{2.11}$$

Comparing (2.1) and (2.11), one finds that $\Gamma_{\rho j}=0$, and since

$$\begin{aligned}\bar{V}_\rho(\mathbf{x})&=\sum_j\int d^3x'\{\rho(\mathbf{x}),g_j(\mathbf{x}')\}\frac{\delta F}{\delta g_j(\mathbf{x}')}\\&=-\sum_j\int d^3x'\,\nabla_{\mathbf{x}}^j[\delta(\mathbf{x}-\mathbf{x}')\rho(\mathbf{x}')]\frac{\delta F}{\delta g_j(\mathbf{x}')}\\&=-\sum_j\nabla_{\mathbf{x}}^j\left[\rho(\mathbf{x})\frac{\delta F}{\delta g_j(\mathbf{x})}\right] \ ,\end{aligned} \tag{2.12}$$

we require

$$\frac{\delta F}{\delta g_j(\mathbf{x})}=\frac{g_j(\mathbf{x})}{\rho(\mathbf{x})} \ . \tag{2.13}$$

This leads to the natural assumption that F is the sum of a kinetic energy term, F_K, and a potential energy term, F_u:

$$F=F_K+F_u \tag{2.14}$$

where

$$F_K=\tfrac{1}{2}\int d^3x\, g^2(\mathbf{x})/\rho(\mathbf{x}) \tag{2.15}$$

and

$$F_u=F_u[\rho(\mathbf{x})] \ . \tag{2.16}$$

The derivation of the kinetic energy term starting from a microscopic point of view is discussed by Langer and Turski.[26]

The streaming velocity for the current is given by

$$\bar{V}_g^i(\mathbf{x})=\int d^3x'\left[\{g_i(\mathbf{x}),\rho(\mathbf{x}')\}\frac{\delta F}{\delta \rho(\mathbf{x}')}+\sum_j\{g_i(\mathbf{x}),g_j(\mathbf{x}')\}\frac{\delta F}{\delta g_j(\mathbf{x}')}\right] . \tag{2.17}$$

Using (2.13),

$$\frac{\delta F}{\delta \rho(\mathbf{x}')}=-\frac{g^2(\mathbf{x}')}{2\rho^2(\mathbf{x}')}+\frac{\delta F_u}{\delta \rho(\mathbf{x}')} \ ,$$

and the Poisson bracket relations, we obtain

$$\bar{V}_g^i(\mathbf{x}) = -\rho(\mathbf{x})\nabla_i \frac{\delta F_u}{\delta\rho(\mathbf{x})} - \sum_j \nabla_j (g_i g_j/\rho) . \tag{2.18}$$

The only quantity left to specify is the damping or dissipative matrix Γ_{ij}. We choose the Γ_{ij} to give the appropriate linearized theory. Therefore

$$\Gamma_{\rho g_1}(\mathbf{x}) = \Gamma_{g_i\rho}(\mathbf{x}) = 0 , \tag{2.19}$$

while

$$\Gamma_{g_i g_j}(\mathbf{x}) = L_{ij}(\mathbf{x}) \tag{2.20a}$$

$$= -\eta_0(\tfrac{1}{3}\nabla_i\nabla_j + \delta_{ij}\nabla^2) - \zeta_0\nabla_i\nabla_j \tag{2.20b}$$

where η_0 is the bare shear viscosity and ζ_0 the bare bulk viscosity. We define the bare longitudinal viscosity $\Gamma_0 = \zeta_0 + 4\eta_0/3$.

Our generalized Langevin equations then reduce to the continuity equation (2.11) and

$$\frac{\partial g_i}{\partial t} = -\rho\nabla_i \frac{\delta F_u}{\delta\rho} - \sum_j \nabla_j(g_i g_j/\rho) - \sum_j L_{ij}(g_j/\rho) + \Theta_i , \tag{2.21}$$

where the noise is Gaussian and satisfies

$$\langle \Theta_i(\mathbf{x},t)\Theta_j(\mathbf{x}',t')\rangle = 2k_B T L_{ij}(\mathbf{x})\delta(\mathbf{x}-\mathbf{x}')\delta(t-t') . \tag{2.22}$$

Each term on the right-hand side of (2.21) should be familiar. If we assume that $F_u[\rho]$ is a local functional of $\rho(x)$ and $\nabla_i\rho(x)$,

$$F_u[\rho] = \int d^3x\, f(\rho(\mathbf{x}), \nabla\rho(\mathbf{x})) \tag{2.23}$$

and define

$$\frac{\partial f}{\partial(\nabla_j\rho)} = (\nabla_j\rho)H(\rho(\mathbf{x}), \nabla\rho(\mathbf{x})) , \tag{2.24}$$

then it is straightforward to show that

$$\rho\nabla_i\delta F_u/\delta\rho = \nabla_i P + \sum_j \nabla_j[H(\nabla_i\rho)(\nabla_j\rho)] \tag{2.25}$$

where

$$P = \rho\frac{\partial f}{\partial\rho} - f + H(\nabla\rho)^2 - \nabla\cdot\left[\frac{H}{2}\nabla\rho^2\right] . \tag{2.26}$$

In the absence of spatial fluctuations, $\nabla\rho = 0$, we see that (2.26) reduces to the usual thermodynamic expression relating the pressure P to the free-energy density f. Thus (2.26) serves to define the "fluctuating" pressure and the first term on the right-hand side of (2.25) gives the usual force term in the Navier-Stokes equation.

The second term on the right-hand side of (2.25) gives a contribution which depends explicitly on the spatial fluctuations in the system. The second term on the right-hand side of (2.21) is just the usual convective term as it appears normally in the equation for $\mathbf{g}$. We return to this in the following.

We note that the streaming velocity for $\mathbf{g}$ can be written in the form

$$\bar{V}_g^i(\mathbf{x}) = -\sum_j \nabla_j \sigma_{ij}^R(\mathbf{x}) \tag{2.27}$$

where

$$\sigma_{ij}^R(\mathbf{x}) = \delta_{ij}P + H\,\nabla_i\rho\,\nabla_j\rho + g_i g_j/\rho \tag{2.28}$$

is the reversible part of the stress tensor and is manifestly symmetric in i and j.

While the equation of motion for g_i should be familiar once we identify $\rho\nabla_i\delta F_u/\delta\rho$ with the force term, the particular form of (2.21) looks a bit unpleasant because of the explicit factors of ρ^{-1} in the convective and dissipative terms. Typically such terms are not evident because one works with the velocity field $\mathbf{V}(\mathbf{x},t)$ which is defined by

$$\mathbf{g} = \rho\mathbf{V} . \tag{2.29}$$

For the incompressible case, where $\rho(\mathbf{x},t) = \rho_0$, this relationship is trivial, but in the compressible case this nonlinear relation must be handled with care. In the next section we discuss one way of dealing with this technical problem. If we use (2.29) in (2.21) then we are dealing with the set of equations given by the continuity equation (2.11),

$$\frac{\partial g_i}{\partial t} = -\nabla_i P - \sum_j \nabla_j[(H\,\nabla_i\rho\,\nabla_j\rho) + \rho V_i V_j + L_{ij}V_j] + \Theta_i \tag{2.30}$$

and the nonlinear constraint (2.29).

III. FIELD-THEORETICAL FORMULATION

The model we have developed is strongly nonlinear in its structure and we want to study the perturbative corrections to the linearized theory due to the various nonlinearities. The approach we will follow is in its general structure, now standard and first described by MSR.[16] The development allows one to develop the theory in the standard field-theoretical form and to carry out perturbation theory in a manner that facilitates conventional renormalization methods.

While the general method is standard,[27] it is also a bit involved. We therefore give a quick review of the basic steps in the formulation, which allows us to set up some definitions and conventions.

Let us, for the moment, return to the general case where we have an equation of motion of the type

$$\frac{\partial\psi(1)}{\partial t_1} = H_1[\psi] + \Theta(1) . \tag{3.1}$$

where the index 1 labels space, time, and any other index carried by the field ψ. Averages of the form $\langle\psi(1)\psi(2)\cdots\psi(N)\rangle$ correspond to averages over the noise source $\Theta(1)$

$$\langle G(\psi)\rangle = \frac{1}{I_0}\int D(\Theta)e^{-A_0(\Theta)}G(\psi) , \tag{3.2}$$

where $G(\psi)$ is some function of ψ, which, in turn, is a function of Θ. I_0 is defined such that $\langle 1\rangle = 1$, and the Gaussian nature of the noise requires that the weighting is given by

$$A_0(\Theta)=\frac{\beta}{4}\int d1\int d2\,\Theta(1)\Gamma^{-1}(1,2)\Theta(2)\ . \tag{3.3}$$

Using the identity

$$\int D(\Theta)\frac{\delta}{\delta\Theta(1)}[\Theta(2)e^{-A_0(\Theta)}]=0 \tag{3.4}$$

and, assuming $\Gamma^{-1}(12)$ is symmetric, it is straightforward to show that the autocorrelation of the noise is given by

$$\langle\Theta(1)\Theta(2)\rangle=2k_BT\Gamma(1,2)\ , \tag{3.5}$$

as required by (2.5). The generating functional for all of the correlation functions of interest is of the form

$$Z_U=I_1\int D(\Theta)e^{-A_0(\Theta)}\exp\left[\int d1\,U(1)\psi(1)\right]\ , \tag{3.6}$$

where I_1 is some normalization constant, and, for example, an N-point correlation function is generated from Z_U using

$$\langle\psi(1)\psi(2)\cdots\psi(N)\rangle=\frac{1}{Z_U}\frac{\delta}{\delta U(1)}\frac{\delta}{\delta U(2)}\cdots\frac{\delta}{\delta U(N)}Z_U\ . \tag{3.7}$$

Z_U, given by (3.6), is not very convenient for generating the perturbation-theory expansion. We can transform it into a more manageable form by using the identity

$$e^{-A_0(\Theta)}=I_2\int D(\hat\psi)\exp\left[-\int d1\int d2[\hat\psi(1)\beta^{-1}\Gamma(12)\hat\psi(2)+i\Theta(1)\hat\psi(2)\delta(12)]\right]\ . \tag{3.8}$$

The left-hand side is obtained by completing the square in the $\hat\psi$ integration and choosing I_2 to cancel the constant coming from the resulting Gaussian integration. We have seen that

$$Z_U=I_1I_2\int D(\Theta)\int D(\hat\psi)\exp\int d1\,U(1)\psi(1)\exp\left[-\int d1\int d2[\hat\psi(1)\beta^{-1}\Gamma(1,2)\hat\psi(2)+i\Theta(1)\delta(1,2)\hat\psi(2)]\right]\ . \tag{3.9}$$

The final step in transforming Z_U is to change variables[28] of integration from Θ to ψ. We will use a "causal" continuation from discrete to continuous time where the Jacobian of the transformation is a constant and obtain

$$Z_U=I_3\int D(\psi)\int D(\hat\psi)e^{-A_U[\psi,\hat\psi]} \tag{3.10}$$

where I_3 is a constant, and

$$A_U[\psi,\hat\psi]=\int d1\int d2\,\hat\psi(1)\beta^{-1}\Gamma(1,2)\hat\psi(2)+i\int d1\,\Theta(1)\hat\psi(1)-\int d1\,U(1)\psi(1)\ . \tag{3.11}$$

We then replace the noise, using (3.1) in the form

$$\Theta(1)=\frac{\partial\psi(1)}{\partial t_1}-H_1[\psi]\ , \tag{3.12}$$

so that

$$A_U[\psi,\hat\psi]=\int d1\int d2\,\hat\psi(1)\beta^{-1}\Gamma(1,2)\hat\psi(2)+i\int d1\,\hat\psi(1)\left[\frac{\partial}{\partial t_1}\psi(1)-H_1[\psi]\right]-\int d1\,U(1)\psi(1)\ . \tag{3.13}$$

Specializing to the fluid case, where $1=(\mathbf{x}_1,t_1)$, we note that the density variable is a bit peculiar since there is no dissipative coupling ($\Gamma_{\rho j}=0$) associated with it. Therefore the only appearance of $\hat\rho$ in the action is in the form

$$i\int d1\,\hat\rho(1)\left[\frac{\partial\rho(1)}{\partial t_1}+\nabla\cdot\mathbf{g}(1)\right]\ , \tag{3.14}$$

and the functional integral over $\hat\rho$ reduces to a δ functional enforcing the continuity equation.

Similarly the relation $\mathbf{g}=\rho\mathbf{V}$ is enforced via a δ functional constraint. One has then, in Z_U, the construction

$$e^{-i\int d1\,\hat\psi(1)H_1[\psi]}\int D(V)\prod_{\mathbf{x}}\delta(\mathbf{g}-\rho\mathbf{V})\ .$$

At this point, after integrating by parts, so that L_{ij} and $-\nabla_j$ operate on $\hat g_i$ in $\hat\psi(1)H_1[\psi]$, one can set $g_i\to\rho V_i$ in the "equation of motion" $H_1[\psi]$. Finally we introduce the additional field $\hat V_i(\mathbf{x},t)$ in the integral representation for the δ functional $\delta(\mathbf{g}-\rho\mathbf{V})$ which generates the term in the action

$$i\int d1\sum_i\hat V_i(1)[g_i(1)-\rho(1)V_i(1)]\ . \tag{3.15}$$

The action, in the absence of the source term proportional to U, is given then by

$$\begin{aligned}A[\psi,\hat\psi]=\int d1\Bigg[&\sum_{i,j}\hat g_i(1)\beta^{-1}L_{ij}(1)\hat g_j(1)+i\hat\rho(1)\left[\frac{\partial\rho(1)}{\partial t_1}+\nabla_1\cdot\mathbf{g}(1)\right]\\&+i\sum_i\hat g_i(1)\left[\frac{\partial}{\partial t_1}g_i(1)+\rho(1)\nabla_1^i\frac{\delta F u}{\delta\rho(1)}+\sum_j\{\nabla_1^j[\rho(1)V_i(1)V_j(1)]\}+L_{ij}(1)V_j(1)\right]\\&+i\sum_i\hat V_i(1)[g_i(1)-\rho(1)V_i(1)]\Bigg]\ .\end{aligned} \tag{3.16}$$

A key reason for changing over to this formalism is that A_U is a polynomial in the ψ and $\hat{\psi}$'s, and standard field-theoretical methods can be used. If we carry out the rescaling

$$\hat{\psi}(1)\to\beta^{-1/2}\hat{\psi}'(1)\ ,$$

$$\psi(1)\to\frac{1}{\beta^{-1/2}}\psi'(1)\ ,$$

we see that the quadratic components are of $O(1)$ and the higher-order terms are of $O[(k_BT)^{n/2-1}]$ where n is the power of ψ in the nonlinear term. Thus we can systematically compute corrections to the Gaussian theory in powers of k_BT.

As discussed in the Introduction, we study here the simplest case where the potential part of the effective Hamiltonian is quadratic and local:

$$F_u[\rho]=\frac{1}{2}\int d^3x\,\chi^{-1}(\delta\rho)^2\ , \tag{3.17}$$

where $\delta\rho(\mathbf{x})=\rho(\mathbf{x})-\rho_0$. The potential energy density then reduces to $f=\frac{1}{2}\chi^{-1}(\delta\rho)^2$ and the pressure is given by[29]

$$P=\rho_0\chi^{-1}\delta\rho+\frac{1}{2}\chi^{-1}(\delta\rho)^2\ . \tag{3.18}$$

As pointed out in Ref. 8, it is the nonlinear term in (3.18) which drives the density feedback mechanism proposed to explain the liquid-glass transition. Putting the quadratic form for F_u into the action (3.16) generates a cubic term of the form $i\sum_i\hat{g}_i(1)\frac{1}{2}\chi^{-1}\nabla_i(\delta\rho)^2$.

In characterizing our field theory it is useful to introduce some notation. Let $\psi_\alpha(1)$ be a vector where α runs over ρ, $\hat{\rho}$, g_i, $\hat{g}_i$, V_i, and $\hat{V}_i$ and 1 labels space $\mathbf{x}_1$ and time t_1. We also use the notation $\hat{\alpha}$ to indicate a set including the hatted variables $\hat{\rho}$, $\hat{g}_i$, and $\hat{V}_i$. The action can then be written in the symmetrized form

$$A[\psi]=\frac{1}{2}\int d1\int d2\sum_{\alpha,\beta}\psi_\alpha(1)[G^{-1}_{\alpha\beta}(1,2)]^0\psi_\beta(2)+\frac{1}{3}\int d1\int d2\int d3\sum_{\alpha,\beta,\gamma}V_{\alpha\beta\gamma}(1,2,3)\psi_\alpha(1)\psi_\beta(2)\psi_\gamma(3)$$
$$+\frac{1}{4}\int d1\int d2\int d3\int d4\sum_{\alpha,\beta,\gamma,\mu}V_{\alpha\beta\gamma\mu}(1,2,3,4)\psi_\alpha(1)\psi_\beta(2)\psi_\gamma(3)\psi_\mu(4)\ , \tag{3.19}$$

where the $[G^{-1}(12)]^0$ are given explicitly in the next section. The symmetrized cubic vertices are given by

$$\begin{aligned}V_{\alpha\beta\gamma}(1,2,3)=\tfrac{1}{2}[&\tilde{V}_{\alpha\beta\gamma}(1,2,3)+\tilde{V}_{\beta\alpha\gamma}(2,1,3)\\&+\tilde{V}_{\gamma\beta\alpha}(3,2,1)+\tilde{V}_{\alpha\gamma\beta}(1,3,2)\\&+\tilde{V}_{\beta\gamma\alpha}(2,3,1)+\tilde{V}_{\gamma\alpha\beta}(3,1,2)]\ ,\end{aligned} \tag{3.20}$$

where

$$\tilde{V}_{\alpha\beta\gamma}(1,2,3)=\sum_{i=1}^{3}\tilde{V}^{(i)}_{\alpha\beta\gamma}(1,2,3) \tag{3.21}$$

with

$$\tilde{V}^{(1)}_{\alpha\beta\gamma}(1,2,3)=i\sum_i\delta_{\alpha,\hat{g}_i}\nabla_1^i\frac{\chi^{-1}}{2}\delta(1,2)\delta(1,3)\delta_{\beta,\rho}\delta_{\gamma,\rho}$$

from the pressure term (3.22)

$$\tilde{V}^{(2)}_{\alpha\beta\gamma}(1,2,3)=i\rho_0\sum_{i,j}\delta_{\alpha,\hat{g}_i}\nabla_1^j\delta_{\beta,V_i}\delta_{\gamma,V_j}\delta(1,2)\delta(1,3) \tag{3.23}$$

from the convective term, and

$$\tilde{V}^{(3)}_{\alpha\beta\gamma}(1,2,3)=-i\sum_i\delta_{\alpha,\hat{V}_i}\delta_{\beta,\rho}\delta_{\gamma,V_i} \tag{3.24}$$

from the nonlinear constraint $\mathbf{g}=\rho\mathbf{V}$. The symmetrized quartic vertex is $\frac{1}{6}$ the sum of all pair-wise permutations of the set of variables $(\alpha,1)$, $(\beta,2)$, $(\gamma,3)$, and $(\mu,4)$ labeling the unsymmetrized vertex $\tilde{V}_{\alpha\beta\gamma\mu}(1,2,3,4)$ where

$$\tilde{V}_{\alpha\beta\gamma\mu}(1,2,3,4)=i\sum_{i,j}\delta_{\alpha,\hat{g}_i}\delta_{\beta,\rho}\delta_{\gamma,V_i}\delta_{\mu,V_j}\nabla_1^i[\delta(1,2)\delta(1,3)\delta(1,4)]\ . \tag{3.25}$$

IV. LINEARIZED THEORY

The linearized theory is generated by neglecting the cubic and quartic couplings in the action $A[\psi]$. Defining the correlation functions

$$G_{\alpha\beta}(1,2)=\langle\psi_\alpha(1)\psi_\beta(2)\rangle\ , \tag{4.1}$$

it is trivial to show for the Gaussian theory, denoted by the superscript 0, that

$$\sum_\gamma\int d3[G^{-1}(1,3)]^0_{\alpha\gamma}G^0_{\gamma\beta}(3,2)=\delta_{\alpha\beta}\delta(1,2)\ . \tag{4.2}$$

After Fourier transformation over space and time, (4.2) reduces to

$$\sum_\gamma[G^{-1}(\mathbf{q},\omega)]^0_{\alpha\gamma}G^0_{\gamma\beta}(\mathbf{q},\omega)=\delta_{\alpha\beta}\ , \tag{4.3}$$

where the matrix elements $[G^{-1}(\mathbf{q},\omega)]^0_{\alpha\beta}$ are given in Table I.

The inversion of the matrix $(G^{-1})^0$ to obtain G^0 is facilitated by the realization that the transverse and longitudinal components of $\mathbf{g},\mathbf{V}$ and their hatted counterparts can be treated separately. The transverse components, $\mathbf{q}\cdot\mathbf{g}_t=0$, do not couple into the density and its hatted conjugate $\hat{\rho}$ and one easily finds the various correlation functions given in Table II.

We note several properties, valid for the G^0, which hold quite generally:

TABLE I. The inverse of the zeroth-order matrix $G^0_{\alpha\beta}$.

	ρ	g_j	V_j	$\hat{\rho}$	$\hat{g}_j$	$\hat{V}_j$
ρ	0	0	0	$-\omega$	$q_j c_0^2$	0
g_i	0	0	0	q_i	$-\omega\delta_{ij}$	$i\delta_{ij}$
V_i	0	0	0	0	iL_{ij}	$-i\rho_0\delta_{ij}$
$\hat{\rho}$	ω	$-q_j$	0	0	0	0
$\hat{g}_i$	$-q_i c_0^2$	$\omega\delta_{ij}$	iL_{ij}	0	$2\beta^{-1}L_{ij}$	0
$\hat{V}_i$	0	$i\delta_{ij}$	$-i\rho_0\delta_{ij}$	0	0	0

$$G_{\hat{\psi}_i\hat{\psi}_j}(\mathbf{q},\omega)=0 \tag{4.4}$$

and the $G_{\hat{\psi}_i\psi_j}$ and $G_{\psi_j\hat{\psi}_i}$ act like, and we shall refer to them as, response functions. Note that they are either retarded or advanced. $G_{\hat{\psi}_i\psi_j}(\mathbf{q},\omega)$ is analytic in the lower half of the complex ω plane.

We also note, for example, that the transverse current correlation function,

$$G_{gg}^{T,0}=\frac{2\beta^{-1}\eta_0 q^2}{\omega^2+(\eta_0 q^2/\rho_0)^2} \tag{4.5}$$

has the usual hydrodynamic form.[12]

The longitudinal correlation functions can also be worked out explicitly and are given in Table III. The analytic structure is the same as in the transverse case, but the spectrum now involves the traveling waves associated with sound propagation.

V. PERTURBATION-THEORY EXPANSION

Finding the corrections to the linearized theory due to the nonlinearities amounts to the development of standard Feynman-graph methods for handling a theory with an action

$$A_U[\Psi]=\sum_{1,2}\tfrac{1}{2}\Psi(1)G_0^{-1}(1,2)\Psi(2)+\tfrac{1}{3}\sum_{1,2,3}V(1,2,3)\Psi(1)\Psi(2)\Psi(3)+\tfrac{1}{4}\sum_{1,2,3,4}V(1,2,3,4)\Psi(1)\Psi(2)\Psi(3)\Psi(4)-\sum_{1}\Psi(1)U(1) \tag{5.1}$$

where we have included a source term, and, in this section only, have incorporated the index α_1 with $\mathbf{x}_1$ and t_1 into the single index 1. Defining the generating functional

$$Z_U=\int D(\Psi)e^{-A_U[\Psi]}, \tag{5.2}$$

the correlation functions of interest are given by

$$G(1)=\frac{\delta}{\delta U(1)}\ln Z_U=\langle\Psi(1)\rangle, \tag{5.3}$$

which vanishes by construction as $U\to 0$ if we include $\delta\rho(1)=\rho(1)-\langle\rho(1)\rangle$ in our vector Ψ, and

$$G(12)=\frac{\delta}{\delta U(2)}G(1)=\langle\delta\Psi(1)\delta\Psi(2)\rangle \tag{5.4}$$

where $\delta\Psi(1)=\Psi(1)-\langle\Psi(1)\rangle$. The perturbation-theory development is standard. In the end one can set the source equal to zero and obtain the set of equations:

$$\sum_3 G^{-1}(1,3)G(3,2)=\delta(1,2) \tag{5.5}$$

where

$$G^{-1}(1,2)=G_0^{-1}(1,2)-\Sigma(1,2) \tag{5.6}$$

defines the self-energy $\Sigma(1,2)$. The self-energy is given by

$$\begin{aligned}\Sigma(1,2)=&-3\sum_{3,4}V(1,2,3,4)G(3,4)+\sum_{3,4,5,6}V(1,6,3)G(3,4)G(6,5)P(4,5,2)\\&-2\sum_{\substack{3,4,5,\\6,7,8}}V(1,3,4,5)G(3,6)G(4,7)G(5,8)\Gamma(6,7,8,2)\\&-\sum_{\substack{3,4,5,\\6,7,8,\\9,1'}}V(1,1',3,4)G(1',9)G(3,6)G(4,5)P(5,6,7)G(7,8)P(8,9,2),\end{aligned} \tag{5.7}$$

where the three- and four-point vertex functions are given by

$$\Gamma(1,2,3)=2V(1,2,3)-3\sum_{4,5,6}\Xi(1,2,3,4)G(3,5)G(4,6)P(5,6,3)+\sum_{4,5,6,7}\Xi(1,2,4,5)G(4,6)G(5,7)\Gamma(6,7,3)\ , \tag{5.8}$$

$$P(1,2,3)=\Gamma(1,2,3)+\sum_{4,5,6,7}\Gamma(1,2,4,5)G(4,6)G(5,7)\Gamma(6,7,3)\ , \tag{5.9}$$

$$\Gamma(1,2,3,4)=\Xi(1,2,3,4)+\sum_{5,6,7,8}\Xi(1,2,5,6)G(5,7)G(6,8)\Gamma(7,8,3,4)\ , \tag{5.10}$$

and

$$\Xi(1,2,3,4)=\frac{\delta\Sigma(1,2)}{\delta G(3,4)}\ . \tag{5.11}$$

It is straightforward to then generate the graphical expansion for Σ as a power series in the V's and the fully interacting G's. Here we shall only need the lowest-order graphs in this expansion. These are given in Fig. 1. As we shall see below, despite the compact simplicity of the notation we have used, there are many contributions even to one-loop order.

VI. NONPERTURBATIVE RESULTS

The field theory we have constructed is rather complicated and involves a large number of objects: the correlation functions, $G_{\psi\psi}$, and the "response" functions $G_{\hat{\psi}\psi}$ and $G_{\psi\hat{\psi}}$. In this section we indicate the degree to which the theory can be simplified.

A. Symmetry

The first important result is that

$$G_{\alpha\hat{\beta}}(\mathbf{q},\omega)=-G^{*}_{\hat{\beta}\alpha}(\mathbf{q},\omega)\ . \tag{6.1}$$

The derivation of this result is given in Appendix A. As can be seen from Tables II and III, (6.1) is satisfied at zeroth order. It is then clear from the Dyson's equation (5.6), defining the self-energies, that

$$\Sigma_{\alpha\hat{\beta}}(\mathbf{q},\omega)=-\Sigma^{*}_{\hat{\beta}\alpha}(\mathbf{q},\omega)\ . \tag{6.2}$$

B. Continuity equation

If we use the general identity

$$\int D(\psi)\frac{\delta}{\delta\psi_{\hat{\alpha}}(1)}[\psi_{\beta}(2)e^{-A[\psi]}]=0\ , \tag{6.3}$$

we obtain

$$\left\langle\psi_{\beta}(2)\frac{\delta A[\psi]}{\delta\psi_{\hat{\alpha}}(1)}\right\rangle=\delta_{\beta,\hat{\alpha}}\delta(1,2)\ . \tag{6.4}$$

Setting $\hat{\alpha}=\hat{\rho}$, and using (3.13) to obtain

$$\frac{\delta A}{\delta\hat{\rho}(1)}=i\left[\frac{\partial\rho}{\partial t_1}(1)+\nabla\cdot\mathbf{g}(1)\right]\ , \tag{6.5}$$

we find

$$1\left[\frac{\partial}{\partial t_1}G_{\rho\beta}(1,2)+\nabla_1\cdot G_{g\beta}(1,2)\right]=\delta_{\beta,\hat{\rho}}\delta(1,2)\ , \tag{6.6}$$

which is a restatment of the continuity equation. Fourier transforming over space and time, we find

$$+\omega G_{\rho\beta}(\mathbf{q},\omega)-\mathbf{q}\cdot G_{g\beta}(\mathbf{q},\omega)=\delta_{\beta,\hat{\rho}}\ . \tag{6.7}$$

Thus we have a simple relation between $G_{\rho\beta}$ and the longitudinal part of $G_{g\beta}$.

C. Fluctuation-dissipation theorem

In a number of problems there is available a fluctuation-dissipation theorem (FDT) relating $G_{\psi\psi}$ and

TABLE II. The transverse part of the zeroth-order matrix $G^0_{\alpha\beta}$. $W_0=\rho_0\omega+iq^2\eta_0$.

	g_t	V_t	$\hat{g}_t$	$\hat{V}_t$
g_t	$\dfrac{2\beta^{-1}\eta_0 q^2\rho_0^2}{W_0W_0^*}$	$\dfrac{2\beta^{-1}\eta_0 q^2\rho_0}{W_0W_0^*}$	$\dfrac{\rho_0}{W_0}$	$\dfrac{\eta_0 q^2}{W_0}$
V_t	$\dfrac{2\beta^{-1}\eta_0 q^2\rho_0}{W_0W_0^*}$	$\dfrac{2\beta^{-1}\eta_0 q^2}{W_0W_0^*}$	$\dfrac{1}{W_0}$	$\dfrac{i\omega}{W_0}$
$\hat{g}_t$	$-\dfrac{\rho_0}{W_0^*}$	$-\dfrac{1}{W_0}$	0	0
$\hat{V}_t$	$-\dfrac{\eta_0 q^2}{W_0}$	$\dfrac{i\omega}{W_0}$	0	0

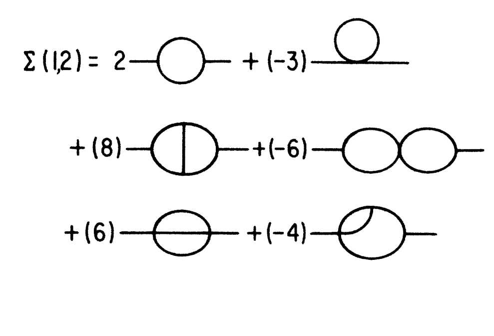

FIG. 1. The diagrammatic expansion for the self-energy given in (5.7) up to second order in (k_BT).

TABLE III. The longitudinal part of the zeroth-order matrix $G^0_{\alpha\beta}$. $D_0=\rho_0(\omega^2-q^2c_0^2)+i\omega q^2\Gamma_0$.

	ρ	g_l	V_l	$\hat{\rho}$	$\hat{g}_l$	$\hat{V}_l$
ρ	$\frac{2\beta^{-1}\Gamma_0 q^4\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^3\omega\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^3\omega\rho_0}{D_0D_0^*}$	$\frac{\rho_0\omega+\Gamma_0 q^2}{D_0}$	$\frac{q\rho_0}{D_0}$	$\frac{\Gamma_0 q^3}{D_0}$
g_l	$\frac{2\beta^{-1}\Gamma_0 q^3\omega\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^2\omega^2\rho_0^2}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^2\omega^2\rho_0}{D_0D_0^*}$	$\frac{qc_0^2\rho_0}{D_0}$	$\frac{\omega\rho_0}{D_0}$	$\frac{\omega\Gamma_1 q^2}{D_0}$
V_l	$\frac{2\beta^{-1}\Gamma_0 q^3\omega\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^2\omega^2\rho_0}{D_0D_0^*}$	$\frac{2\beta^{-1}\Gamma_0 q^2\omega^2}{D_0D_0^*}$	$\frac{qc_0^2}{D_0}$	$\frac{\omega}{D_0}$	$\frac{+i(\omega^2-q^2c_0^2)}{D_0}$
$\hat{\rho}$	$\frac{-(\rho_0\omega-i\Gamma_0 q^2)}{D_0^*}$	$\frac{-qc_0^2\rho_0}{D_0^*}$	$\frac{-qc_0^2}{D_0^*}$	0	0	0
$\hat{g}_l$	$\frac{-q\rho_0}{D_0^*}$	$\frac{-\omega\rho_0}{D_0^*}$	$\frac{-\omega}{D_0^*}$	0	0	0
$\hat{V}_l$	$\frac{-\Gamma_0 q^3}{D_0^*}$	$\frac{-\omega\Gamma_0 q^2}{D_0^*}$	$\frac{i(\omega^2-q^2c_0^2)}{D_0^*}$	0	0	0

$G_{\hat{\psi}\psi}$. In the present case, as shown in Appendix B, one has the fluctuation-dissipation theorem

$$G_{V_i\beta}(\mathbf{q},\omega)=-2\beta^{-1}\mathrm{Im}G_{\hat{g}_i\beta}(\mathbf{q},\omega) \tag{6.8}$$

where β is an unhatted variable. It is straightforward to verify that (6.8) is satisfied by all of the zeroth-order quantities in Table II. Using (6.8) we can relate a number of the response functions and correlation functions.

D. Transverse self-energies

The full matrix G can be divided into its longitudinal and transverse parts as in the linear case (Sec. II). In a similar fashion the self-energy $\Sigma_{\alpha_i\beta_j}(\mathbf{q},\omega)$, where α_i and β_j are in the set of vectors $(g_i,\hat{g}_i,V_i,\hat{V}_i)$ and i and j are vector labels, can be written in the form

$$\Sigma_{\alpha_i\beta_j}(\mathbf{q},\omega)=\hat{q}_i\hat{q}_j\Sigma^L_{\alpha\beta}(\mathbf{q},\omega)+(\delta_{ij}-\hat{q}_i\hat{q}_j)\Sigma^T_{\alpha\beta}(\mathbf{q},\omega) . \tag{6.9}$$

Since the transverse case contains only the components of the vector fields, it is less complicated and we will consider it first.

Using the simplification discussed above, we can invert (5.6) to obtain the response functions in the form

$$G^T_{\alpha\hat{\beta}}(\mathbf{q},\omega)=\frac{M_{\alpha\hat{\beta}}}{W} \tag{6.10}$$

where the $M_{\alpha\hat{\beta}}$ are given by

$$M_{g\hat{g}}=\rho_T , \tag{6.11}$$

$$M_{g\hat{V}}=q^2_\eta , \tag{6.12}$$

$$M_{V\hat{g}}=1 , \tag{6.13}$$

$$M_{V\hat{V}}=i\omega , \tag{6.14}$$

while

$$W=\rho_T\omega+iq^2\eta , \tag{6.15}$$

$$\rho_T=\rho_0-i\Sigma^T_{\hat{V}V}(\mathbf{q},\omega) , \tag{6.16}$$

and

$$q^2\eta=q^2\eta_0+i\Sigma^T_{\hat{g}V}(\mathbf{q},\omega) . \tag{6.17}$$

We also find that

$$G^T_{\hat{\beta}\alpha}=\frac{M_{\hat{\beta}\alpha}}{W^*} \tag{6.18}$$

where

$$M_{\alpha\hat{\beta}}=-(M_{\hat{\beta}\alpha})^* . \tag{6.19}$$

in keeping with the general symmetry relation $G_{\alpha\hat{\beta}}=-(G_{\hat{\beta}\alpha})^*$.

The correlation functions are given by

$$G^T_{\alpha\beta}=-\sum_{\gamma,\delta}G^T_{\alpha\hat{\gamma}}C^T_{\hat{\gamma}\hat{\delta}}G^T_{\hat{\delta}\beta} \tag{6.20}$$

where the matrix C^T is given by

$$C^T_{\hat{\alpha}\hat{\beta}}=-\Sigma^T_{\hat{\alpha}\hat{\beta}}+\delta_{\hat{\alpha}\hat{g}}\delta_{\hat{\beta}\hat{g}}2\beta^{-1}q^2\eta_0 . \tag{6.21}$$

We use the FDT, (6.8), in (6.20) to obtain, after multiplying from the right by $G^{T-1}_{\hat{\beta}\hat{\alpha}}$ and from the left by W,

$$\sum_\beta M_{V\hat{\beta}}C^T_{\hat{\beta}\hat{\alpha}}=\frac{2\beta^{-1}}{W^*}\sum_\beta \mathrm{Im}(M_{\hat{g}\beta}W)(G^{-1}_{\hat{\beta}\hat{\alpha}})^T . \tag{6.22}$$

This equation can be solved to give the matrix C^T, which depends on the self-energies of the form $\Sigma_{\hat{\alpha}\hat{\beta}}$, in terms of the self-energies between hatted and unhatted fields on the right-hand side of (6.22). Using the result

$$C^T_{\hat{\alpha}\hat{\beta}}=(C^T_{\hat{\beta}\hat{\alpha}})^* , \tag{6.23}$$

derived in Appendix A, we find that (6.22) reduces to the relations

$$\Sigma^{T'}_{\hat{g}\hat{V}}(\mathbf{q},\omega)=0 , \tag{6.24}$$

$$\Sigma^{T'}_{\hat{g}\hat{g}}(\mathbf{q},\omega)-\omega\Sigma^{T''}_{\hat{g}\hat{V}}(\mathbf{q},\omega)=2\beta^{-1}\Sigma^{T''}_{\hat{g}V}(\mathbf{q},\omega) , \tag{6.25}$$

$$\omega\Sigma^{T'}_{\hat{V}\hat{V}}(\mathbf{q},\omega)+\Sigma^{T''}_{\hat{g}\hat{V}}(\mathbf{q},\omega)=+2\beta^{-1}\Sigma^{T'}_{\hat{V}V}(\mathbf{q},\omega) , \tag{6.26}$$

where, as usual, the single and double prime indicate the real and imaginary parts. We can use these two relations to obtain the expression

$$G^T_{gg}(\mathbf{q},\omega)=\frac{2\beta^{-1}}{\omega}\mathrm{Re}(\rho_T G_{g\hat{g}})-\frac{\Sigma''_{\hat{g}\hat{V}}}{\omega}\ , \tag{6.27}$$

while

$$G^T_{VV}(\mathbf{q},\omega)=2\beta^{-1}\mathrm{Im}G_{V\hat{g}}\ , \tag{6.28a}$$

$$G^T_{Vg}(\mathbf{q},\omega)=2\beta^{-1}\mathrm{Im}G_{g\hat{g}}\ , \tag{6.28b}$$

which follows from (6.8) and (6.1). Thus, using the fluctuation-dissipation theorem, we can express all the correlation functions in terms of the "response" self-energies, with exception of the $\Sigma''_{\hat{g}\hat{V}}/\omega$ term in (6.27).

E. Longitudinal case

In the longitudinal case we follow a procedure similar to that in the transverse case. However, this case is more complicated since we have three fields (ρ,g,V) in both the hatted and unhatted sets. Inverting the G^{-1} matrix in (5.6) we obtain the response functions in the form

$$G^L_{\alpha\hat{\beta}}(\mathbf{q},\omega)=\frac{N_{\alpha\hat{\beta}}}{D} \tag{6.29}$$

with the matrix N given in Table IV, and

$$D=\rho_L(\omega^2-q^2c^2)+iL(\omega+iq\Sigma^L_{\hat{V}\rho})\ , \tag{6.30}$$

$$L(\mathbf{q},\omega)=q^2\Gamma_0+i\Sigma^L_{\hat{g}V}(\mathbf{q},\omega)\ , \tag{6.31}$$

$$qc^2(\mathbf{q},\omega)=qc_0^2+\Sigma^L_{\hat{g}\rho}(\mathbf{q},\omega)\ , \tag{6.32}$$

$$\rho_L(\mathbf{q},\omega)=\rho_0-i\Sigma^L_{\hat{V}V}(\mathbf{q},\omega)\ . \tag{6.33}$$

Similarly

$$G^L_{\hat{\beta}\alpha}=\frac{N_{\hat{\beta}\alpha}}{-D^*} \tag{6.34}$$

where

$$N_{\alpha\hat{\beta}}=(N_{\alpha\hat{\beta}})^*\ . \tag{6.35}$$

The correlation functions are given in this case by

$$G^L_{\alpha\beta}=-\sum_{\gamma,\delta}G^L_{\alpha\hat{\gamma}}C^L_{\hat{\gamma}\hat{\delta}}G^L_{\hat{\delta}\beta} \tag{6.36}$$

where the summations are over ρ, g, and V. C^L is given by

$$C^L_{\hat{\alpha}\hat{\beta}}=-\Sigma^L_{\hat{\alpha}\hat{\beta}}+\delta_{\hat{\alpha},\hat{g}}\delta_{\hat{\beta},\hat{g}}2\beta^{-1}q^2\Gamma_0 \tag{6.37}$$

and is zero if $\hat{\alpha}$ or $\hat{\beta}$ equals $\hat{\rho}$. So C^L has only four nonzero elements.

We can then, as in the transverse case, use the FDT to obtain expressions for the elements of the matrix C^L in terms of the self-energies, $\Sigma_{\alpha\hat{\beta}}$, which determine the response functions. After noting that $\Sigma^L_{\hat{g}\hat{g}}$ and $\Sigma^L_{\hat{V}\hat{V}}$ are real and $\Sigma^L_{\hat{g}\hat{V}}$ imaginary, and carrying out some algebra we obtain the relations

$$D_1\Sigma''_{\hat{V}\hat{g}}=2\beta^{-1}N''_{V\hat{V}}\mathrm{Re}(\rho_L N_{\hat{g}V})+\mathrm{Im}(N_{V\hat{V}}N^*_{V\hat{g}})C'_{\hat{V}\hat{g}}\ , \tag{6.38}$$

$$D_1(\Sigma_{\hat{g}\hat{g}}-2\beta^{-1}\Sigma''_{\hat{g}V})=-2\beta^{-1}N''_{V\hat{V}}\mathrm{Im}(\rho_L N^*_{V\hat{V}})+|N_{V\hat{V}}|^2C'_{\hat{V}\hat{g}}\ , \tag{6.39}$$

$$D_1\Sigma_{\hat{V}\hat{V}}=+2\beta^{-1}N'_{V\hat{g}}\mathrm{Re}(\rho_L N_{\hat{g}V})+|N_{V\hat{g}}|^2C_{\hat{V}\hat{g}}\ , \tag{6.40}$$

where

$$D_1=N''_{V\hat{V}}N''_{V\hat{g}}+N'_{V\hat{V}}N'_{V\hat{g}}\ , \tag{6.41}$$

and we have, for clarity, suppressed the index L on the Σ's. We have not succeeded in finding useful simple expressions, as given by (6.28) in the transverse case, giving $G_{\rho\rho}$, $G_{\rho g}$, and G_{gg} in terms of the response functions. This can, however, be achieved in the hydrodynamic limit. The FDT can be used to express $G_{V\rho}$, G_{Vg}, and G_{VV} simply in terms of $G_{\hat{g}\rho}$, $G_{\hat{g}g}$, and $G_{\hat{g}V}$, respectively.

F. The hydrodynamic limit

We now analyze these self-energy relations in the hydrodynamic limit. In the transverse case, where the hydrodynamic pole is diffusive, we are interested in the self-energies $\Sigma^T(\omega\sim q^2,q)$ as $q\to 0$. In the longitudinal case one has a traveling mode and we are interested in the self-energies $\Sigma^L(\omega\sim cq,q)$ as $q\to 0$.

1. Transverse case

The hydrodynamic limit is easily accessed if one identifies the explicit factors of $\mathbf{q}$ and ω necessitated by a symmetry and the conservation laws. We have, for example that

$$\Sigma^T_{\hat{g}V}=-iq^2\gamma_{\hat{g}V}(\mathbf{q},\omega)\ ,$$

$$\Sigma^T_{\hat{g}\hat{g}}(\mathbf{q},\omega)=-q^2\gamma_{\hat{g}\hat{g}}(\mathbf{q},\omega)\ , \tag{6.42}$$

$$\Sigma^T_{\hat{g}\hat{V}}(\mathbf{q},\omega)=q^2\gamma_{\hat{g}\hat{V}}(\mathbf{q},\omega)\ . \tag{6.43}$$

These results follow from conservation of momentum which says that every external $\hat{g}_i$ vertex contributing to $\Sigma^T_{\hat{g}V}(\mathbf{q},\omega)$ will supply a factor of q_i. Since the system is isotropic one finds that all $\Sigma^T_{\hat{g}\beta}$ must be of $O(q^2)$. We then use (6.24)–(6.26) to obtain conditions on the γ's as $\mathbf{q}$ and ω go to zero. From (6.25) we obtain

$$\gamma_{\hat{g}\hat{g}}(0,0)=2\beta^{-1}\gamma'_{\hat{g}V}(0,0)\ , \tag{6.44}$$

while (6.26) gives

$$\Sigma^{T'}_{\hat{V}\hat{V}}(0,0)=\lim_{\omega\to 0}\frac{1}{\omega}2\beta^{-1}\Sigma^{T'}_{\hat{V}V}(0,\omega)\ . \tag{6.45}$$

This tells us, since $\Sigma^{T'}_{\hat{V}\hat{V}}(0,0)$ is nonzero and finite, that $\Sigma^{T'}_{\hat{V}V}(0,0)=0$.

Using (6.44) and (6.45) we can write down the transverse correlation functions in the hydrodynamic limit in the same form as the zeroth-order result given in Table II. ρ_0 and η_0 are now replaced by their renormalized counterparts ρ_T and η, respectively. The renormalized η is given by

$$\eta=\eta_0+\gamma'_{\hat{g}\hat{V}}(0,0) \tag{6.46}$$

or

$$\eta=\eta_0+\tfrac{1}{2}\beta\gamma_{\hat{g}\hat{g}}(0,0)\ , \tag{6.47}$$

while

$$\rho_T(0,0)=\rho_0+\Sigma^{T''}_{\hat{V}V}(0,0) . \tag{6.48}$$

2. Longitudinal case

As in the transverse case we can extract certain explicit factors of the wave number using the conservation laws and symmetry. We have

$$\Sigma^L_{\hat{g}V}=-iq^2\gamma_{\hat{g}V} , \tag{6.49}$$

$$\Sigma^L_{\hat{g}\hat{g}}=-q^2\gamma_{\hat{g}\hat{g}} , \tag{6.50}$$

$$\Sigma_{\rho\hat{g}}=q\gamma_{\rho\hat{g}} , \tag{6.51}$$

$$\Sigma_{\rho\hat{V}}=q\gamma_{\rho\hat{V}} , \tag{6.52}$$

$$C'_{\hat{V}\hat{g}}=2\beta^{-1}q^2\gamma_L . \tag{6.53}$$

Starting with (6.38), we find, since $\Sigma''_{\hat{V}\hat{g}}$ must vanish as $O(q^2\omega)$ for $\mathbf{q}$ and ω going to zero, that

$$\gamma_L(0,\omega)=-\gamma''_{\rho\hat{g}}(0,\omega)\gamma''_{\hat{V}V}(0,\omega)/\omega^2 \tag{6.54}$$

and

$$\lim_{\omega\to 0}\left[\frac{\gamma''_{\hat{V}V}(0,\omega)}{\omega}\right]=\rho\gamma'_{\rho\hat{V}}(0,0)/c^2 , \tag{6.55}$$

where the renormalized density is

$$\rho(0,0)=\rho_0+\Sigma''_{\hat{V}V}(0,0) , \tag{6.56}$$

and the renormalized speed of sound,

$$c^2=c_0^2-\gamma'_{\rho\hat{g}}(0,0) , \tag{6.57}$$

(6.39) reduces to the result

$$\gamma_{\hat{g}\hat{g}}(0,0)-2\beta^{-1}\gamma'_{\hat{g}V}(0,0)=\lim_{\omega\to 0}\left[2\beta^{-1}\frac{\gamma''_{\rho\hat{g}}(0,\omega)}{\omega}\right] , \tag{6.58}$$

and (6.40) reduces to

$$C_{\hat{V}\hat{V}}(0,0)=-2\beta^{-1}\rho\gamma'_{\rho\hat{V}}(0,0)/c^2 . \tag{6.59}$$

The renormalization of all of the correlation functions in the hydrodynamic limit simply involves the replacement $\rho_0\to\rho$, $c_0^2\to c^2$, and $\Gamma_0\to\Gamma$ in the zeroth-order results, where Γ can be computed as

$$\Gamma=\Gamma_0+\gamma_{\hat{g}V}(0,0)+\lim_{\omega\to 0}\left[\frac{\rho\gamma''_{\rho\hat{g}}(0,\omega)}{\omega}\right] \tag{6.60}$$

or

$$\Gamma=\Gamma_0+\frac{\beta}{2}\gamma_{\hat{g}\hat{g}}(0,0) . \tag{6.61}$$

We can, because of the interrelations among the self-energies, compute the nonlinear correction to the viscosity by analyzing either $\Sigma_{\hat{g}\hat{g}}$ or the response self-energies $\Sigma_{\hat{g}V}$ and $\Sigma_{\hat{g}\rho}$. In the next section we will calculate the one-loop contribution to these renormalized quantities.

Note that in the hydrodynamic limit the correction function $G_{\rho\rho}$ can be computed as

$$G_{\rho\rho}(\mathbf{q},\omega)=+2\beta^{-1}\chi_{\rho\rho}(\mathbf{q})\mathrm{Im}G_{\hat{\rho}\rho}(\mathbf{q},\omega) , \tag{6.62}$$

where $\chi_{\rho\rho}(\mathbf{q})$ is the Fourier transform of $\langle\delta\rho(\mathbf{x})\delta\rho(\mathbf{x}')\rangle$.

VII. ONE-LOOP THEORY

In the last section we derived a number of relations among the different self-energies. In this section we evaluate these self-energies explicitly to $O(k_BT)$. We then identify those contributions which appear to be important as one approaches the glass transition and we also verify the self-energy relations found in the last section. We shall restrict ourselves to one-loop order $[O(k_BT)]$ in our analysis. Higher-order corrections can be generated following the development in Sec. III. We assume a "flat" static structure factor

$$\chi^0_{\rho\rho}(\mathbf{q})=\begin{cases}\rho_0/c^2, & q<\Lambda\\ 0, & q>\Lambda\end{cases} \tag{7.1}$$

where Λ serves as a large wave-number cutoff.

We use the diagrammatic expansion discussed in Sec. V to obtain the contributions to the self-energy matrix to one-loop order. In Fig. 2 all such diagrams that can be drawn for the four vertices, given by (3.22)–(3.25), are listed. They are either the bubble-type diagrams or a Hartree-type diagram with one correlation function closed on itself. As before, we divide the self-energies into longitudinal and transverse parts and consider the transverse case first.

A. Transverse case

As shown in the last section the effect of nonlinearities is to renormalize the shear viscosity and the ambient density. As shown in Sec. VI, the correction to the viscosity can be obtained from either $\Sigma^T_{\hat{g}V}$ or $\Sigma^T_{\hat{g}\hat{g}}$. In the present situation, where we are considering a flat static structure factor, the contribution to the transverse parts of these two self-energies came only from the diagrams involving the convective vertex (3.23). These contributions have been extensively investigated in the context of mode coupling theory and incompressible fluids. One finds the usual long-time tails[23] in three dimensions, while for two or less dimensions there are serious divergences in the small q and ω limit and conventional hydrodynamics breaks down. This means that such contributions must be treated carefully and will compete with the nonlinear density feedback mechanism in low-dimensional systems. We shall assume that in three dimensions the contributions generated by the convective nonlinearities simply renormalize η_0.

In the flat spectrum case there is no coupling between the shear viscosity and the density fluctuations. Such a coupling is generated by gradients of the density in the effective Hamiltonian $F_u[\rho]$. Such a model will be treated elsewhere.

Next we consider the renormalization of the density, which from (6.48), is given by $\Sigma^{T''}_{\hat{V}V}(0,0)$. Evaluating the four graphs contributing to this self-energy, shown in Fig. 2, in the small q and ω limit, we obtain, using the zeroth-order correlation functions,

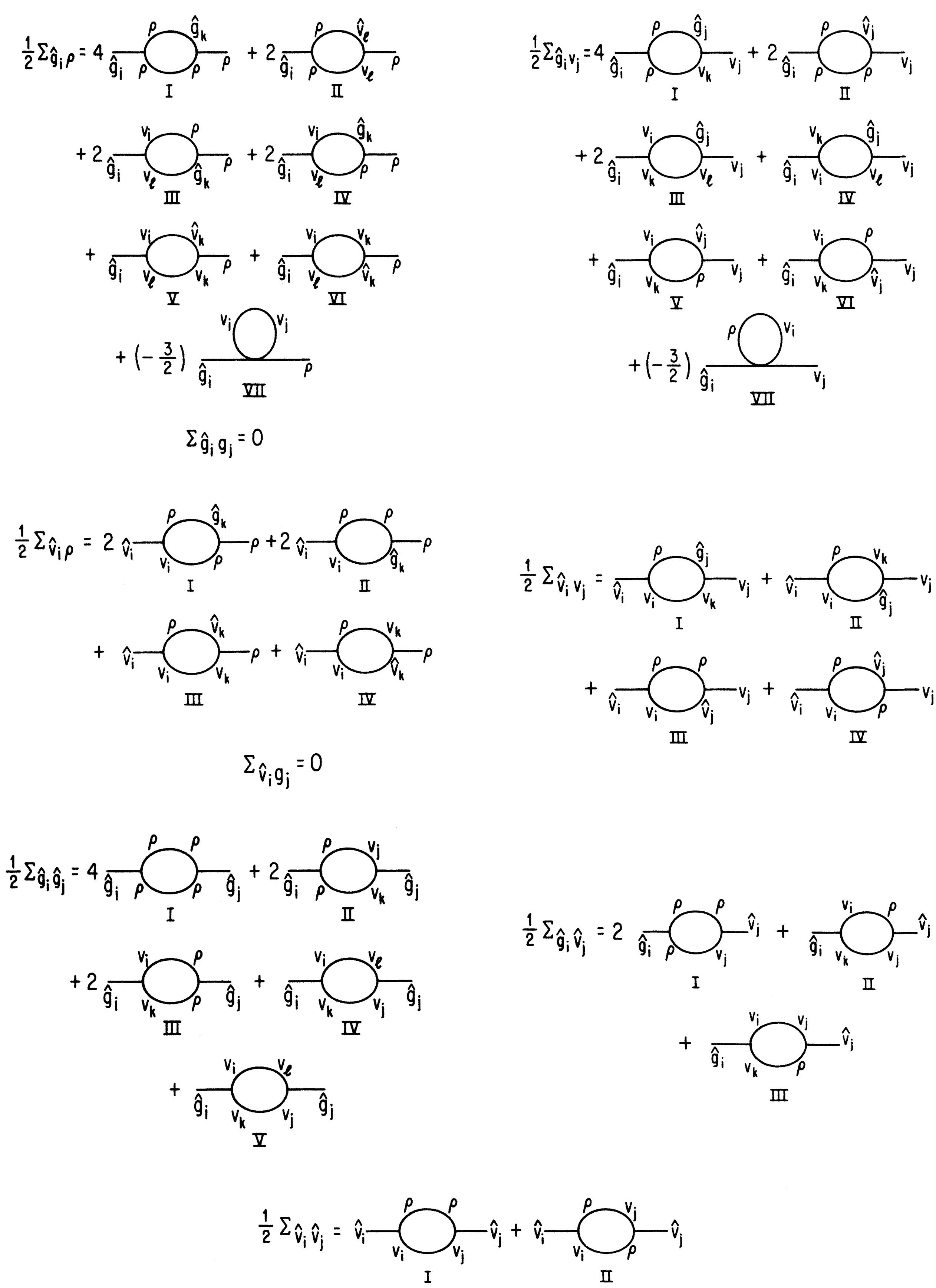

FIG. 2. The diagrams contributing to $O(k_B T)$ for all the different elements of the self-energy matrix.

$$\rho=\rho_0\left[1-\frac{\beta^{-1}}{3\pi^2\rho_0^2 l^3 c_c^3}[\Lambda l-\tan^{-1}(\Lambda l)]\right], \qquad (7.2)$$

where

$$l=[\eta_0(\eta_0+\Gamma_0)]^{1/2}/(\rho_0 c) \qquad (7.3)$$

is a length.

B. Longitudinal case

In this case the renormalization of the longitudinal viscosity comes from either $\Sigma_{\hat{g}V}$ and $\Sigma_{\hat{g}\rho}$ or $\Sigma_{\hat{g}\hat{g}}$. As in the transverse case, the diagrams involving convective vertices give constant contributions in three dimensions and they can be absorbed in the renormalization of the bare transport coefficient Γ_0. All the contributions to $\Sigma_{\hat{g}\rho}$ are of this type. The first diagram for $\Sigma^L_{\hat{g}\hat{g}}$ in Fig. 2 does not have a convective vertex in it. Instead it involves the density feedback mechanism identified by Leutheusser[6] and in Ref. 8 and is given by

$$\Sigma^L_{\hat{g}\hat{g}}(\mathbf{q},\omega) =q^2\beta\chi^{-2}\int\frac{d\omega_1}{2\pi}\int\frac{d^3k}{(2\pi)^3}G_{\rho\rho}(\mathbf{k},\omega_1) \times G_{\rho\rho}(\mathbf{q}-\mathbf{k},\omega-\omega_1)\,. \qquad (7.4)$$

Comparing (6.60) and (6.61), one finds that there must be a related contribution in $\Sigma^L_{\hat{g}V}$. It is given by the first graph contributing to $\Sigma^L_{\hat{g}V}$ in Fig. 2. Using these results we can define an effective longitudinal viscosity which can, using (6.60) or (6.61), be written as

$$\Gamma(\mathbf{q},\omega)=\Gamma_0+i\beta\chi^{-2}\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\int\frac{d^3k}{(2\pi)^3}\frac{G_{\rho\rho}(\mathbf{k},\omega_1)G_{\rho\rho}(\mathbf{q}-\mathbf{k},\omega_2)}{\omega-\omega_1-\omega_2} \qquad (7.5)$$

or

$$\Gamma(\mathbf{q},\omega)=\Gamma_0+\beta\chi^{-2}\int_0^\infty dt\, e^{i\omega t}\int\frac{d^3k}{(2\pi)^3}G_{\rho\rho}(\mathbf{k},t) \times G_{\rho\rho}(\mathbf{q}-\mathbf{k},t) \qquad (7.6)$$

where Γ_0 contains the constant contributions from the other diagrams. We shall look at the consequences of this nonlinear coupling between the longitudinal viscosity and the density correlations in the next section.

We consider next the renormalization of the sound speed as indicated by (6.67). This requires evaluation of $\Sigma'_{\hat{g}\rho}$. Explicit evaluation of the seven graphs contributing to $\Sigma_{\hat{g}\rho}$ in one-loop order (see Fig. 2), shows that $\lim_{q\to 0}\Sigma'_{\hat{g}\rho}/q=0$. Thus the hydrodynamic sound speed remains unchanged at one-loop order. The renormalization of the density is the same as in the transverse case. This completes the one-loop determination of the different quantities contributing at leading order in the small ω and q limit. We want, however, to consider a particular nonhydrodynamic contribution that will play a key role in our analysis in the next section.

The $O(q)$ contribution to $\Sigma_{\rho\hat{V}}$, for fixed ω, from the one-loop diagrams can be conveniently computed from $\Sigma_{\hat{V}\hat{V}}$ using (6.59). Evaluating the graphs for $\Sigma_{\hat{V}\hat{V}}$ given in Fig. 2, we obtain the explicit expression for $\gamma_{\rho\hat{V}}$:

$$\gamma_{\hat{V}\rho}=\lim_{q\to 0}\left[\frac{1}{q}\Sigma'_{\hat{V}\rho}(\mathbf{q},0)\right]=\frac{c^2}{3\beta^{-1}\rho}(I_T+I_L)\,, \qquad (7.7)$$

with

$$I_T=\int\frac{d^3k}{(2\pi)^3}\frac{d\Omega}{2\pi}G_{\rho\rho}(\mathbf{k},\Omega)G^T_{VV}(\mathbf{k},\Omega)\,, \qquad (7.8)$$

$$I_L=\int\frac{d^3k}{2\pi}\frac{d\Omega}{2\pi}G_{\rho\rho}(\mathbf{k},\Omega)G^L_{VV}(\mathbf{k},\Omega)\,. \qquad (7.9)$$

Using the zeroth-order expressions for the different correlation functions one finds that

$$I_T=\frac{\beta^{-2}\rho_0}{3c^2\pi^2\eta_0 l}[\Lambda l-\tan^{-1}(\Lambda l)] \qquad (7.10)$$

$$I_L=\frac{\rho_0\beta^{-2}}{3c^2\pi^2}\frac{\Lambda}{\Gamma_0} \qquad (7.11)$$

with l given by (7.3).

VIII. IMPLICATIONS FOR THE GLASS TRANSITION

Having identified the contributions to the various self-energies in the one-loop approximation, we can now look at the implications of these results in the case of a very dense system. There are two main points we want to make. The first point is that we can make direct contact with the model proposed by Leutheusser[6] and others[7–9] to describe the liquid-glass transition. The second and very important point is that we find an additional contribution which acts to cut off the dynamical transition predicted by the Leutheusser model. Its physical interpretation seems to be associated with the development of a nonhydrodynamic diffusive mode. The most important conclusion of this paper is that there is not a sharp dynamical transition in our model as one lowers the temperature or increases the density. However, at least within the context of the oversimplified model we analyze in this section, there are strong remnants of this transition and the overall behavior looks very similar to that found in recent experiments.[17]

Any model which includes the density fluctuations which drive Leutheusser's mechanism must include nonhydrodynamic corrections. Thus we must allow for the frequency dependence of the viscosities. We will, however, be somewhat discriminating and only include the frequency dependence associated with the density fluctuations in the contributions to $\Gamma(\mathbf{q},\omega)$. We assume that the other contributions to $\Gamma(\mathbf{q},\omega)$ can be absorbed into an additive bare contribution and start with (7.6) as our basic approximation for the longitudinal viscosity. We assume that the density and speed of sound are simply constants ρ and c.

It will be crucial that we include in our analysis the contribution to the self-energy $\Sigma_{\rho\hat{V}}$. This term is negligible in the hydrodynamic limit, but, as we shall see, plays a crucial role near the "glass transition." Using the results at one-loop order we can write, as finite q and ω generalizations of (7.7)–(7.9):

$$\Sigma_{\rho\hat{V}}(\mathbf{q},\omega)=q[\gamma(\mathbf{q},\omega)+\gamma_T(\mathbf{q},\omega)] , \tag{8.1}$$

where

$$\gamma(\mathbf{q},\omega)=\frac{c^2\beta}{3\rho^3}\int_0^\infty dt\, e^{i\omega t}\int\frac{d^3k}{(2\pi)^3}\frac{1}{k^2}\frac{d}{dt}G_{\rho\rho}(\mathbf{k},t)\times\frac{d}{dt}G_{\rho\rho}(\mathbf{q}-\mathbf{k},t) , \tag{8.2}$$

and we could write a similar expression for $\gamma_T(\mathbf{q},\omega)$ which reduces to (7.8) in the hydrodynamic limit. It is clear in the simplest model, while there is a flat structure factor [see (7.1)] and no coupling between the density feedback mechanism and the transverse viscosity, that γ_T will be insensitive to any glass transition and $\Sigma_{\rho\hat{V}}(\mathbf{q},\omega)$ will, in the sense discussed in detail below, cut off the transition. Since we expect, in a more realistic model, that the shear viscosity will be coupled back to the density, the lack of sensitivity of γ_T to the feedback may be misleading. In other words, if one couples the density to the transverse fluctuations, γ_T may be driven to zero as one approaches the glass transition and $\Sigma_{\rho\hat{V}}$ may not modify the nature of the transition. To test this possibility we will set $\gamma_T=0$ (which requires η_0 very large) and ask whether γ is driven to zero as one approaches the transition. We assume for the rest of this section that

$$\Sigma_{\rho\hat{V}}(\mathbf{q},\omega)=q\gamma(\mathbf{q},\omega) \tag{8.3}$$

with γ given by (8.2). With this set of assumptions we find, using (6.33) and Table IV, that

$$G_{\rho\hat{\rho}}(\mathbf{q},\omega)=\frac{\rho\omega+iq^2\Gamma(\mathbf{q},\omega)}{\rho(\omega^2-c^2q^2)+iq^2\Gamma(\mathbf{q},\omega)[\omega+iq^2\gamma(\mathbf{q},\omega)]} \tag{8.4}$$

and we can use (6.62) to determine $G_{\rho\rho}(\mathbf{q},\omega)$. Thus we have a set of nonlinear equations for Γ, γ, and $G_{\rho\rho}$ which appear to give the dominant contribution in the high-density and low-temperature limits. In gaining a feeling for these equations it is useful to follow the original analysis of Leutheusser. First let us ignore the contribution from γ and assume that the viscosity is, as we make the system progressively denser, growing larger. Then, for fixed wave number, we eventually reach a point where ω is small compared to Γq^2 and (8.4) reduces to

TABLE IV. The matrix $N_{\alpha\hat{\beta}}$ determining the response function $G_{\alpha\hat{\beta}}$ in Eq. (6.29).

	$\hat{\rho}$	$\hat{g}$	$\hat{V}$
ρ	$\rho_L\omega+iL$	$\rho_L q$	Lq
g	$\rho_L qc^2+L\Sigma_{\hat{V}\rho}$	$\rho_L\omega$	ωL
V	$qc^2+i\omega\Sigma_{\hat{V}\rho}$	$\omega+iq\Sigma_{\hat{V}\rho}$	$i(\omega^2-q^2c^2)$

$$G_{\rho\hat{\rho}}(\mathbf{q},\omega)=[\omega-\rho c^2/i\Gamma(\mathbf{q},\omega)]^{-1} . \tag{8.5}$$

Notice that $G_{\rho\hat{\rho}}(\mathbf{q},\omega)$, as given by (8.5), has no explicit dependence on wave number if the static structure factor, which is inversely proportional to c^2, and $\Gamma(\mathbf{q},\omega)$ are wave-number independent. One obtains self-consistency if the correlation functions inside the integrals in (7.6) and (8.2) are taken to be independent of wave number and they can be rewritten in the forms

$$\Gamma(\omega)=\Gamma_0+\lambda c^2\int_0^\infty e^{i\omega t}\psi^2(t)dt , \tag{8.6}$$

$$\gamma(\omega)=\frac{\lambda}{3}\int dt\, e^{i\omega t}[\dot{\psi}(t)]^2\equiv\frac{\lambda}{3}I(\omega) , \tag{8.7}$$

where $\psi(t)=G_{\rho\hat{\rho}}(t)$, the coupling λ is defined by

$$\lambda=\frac{(k_BT)\Lambda^3}{6\pi^2\rho c^2} . \tag{8.8}$$

Returning to our analysis of (8.4), we easily see that this equation, with (8.6) and $\gamma=0$, leads to a sharp glass transition. This follows after making the assumption that $\psi(t)$ can be written in the form

$$\psi(t)=i\psi_0\Theta(t)+\psi_v(t) \tag{8.9a}$$

or

TABLE V. The longitudinal viscosity Γ, I, and σ [see Eqs. (8.7) and (8.13)] as a function of λ resulting from a numerical integration of (8.11). The values of q/Λ used was $\frac{1}{8}$.

g	σ	I	Γ/Γ_0
0		0.56	1.0
1	0.4054	0.292	2.0
2	0.1434	0.190	4.4
2.5	0.0837	0.158	7.4
3	0.0443	0.133	13.8
3.5	0.2247	0.114	29.6
3.75	0.0166	0.106	44.0
4	0.0132	0.099	62.9
4.25	0.0113	0.093	84.2
4.5	0.0106	0.088	103.8
5	0.0090	0.079	152.1
6	0.0076	0.067	248.4
7	0.0071	0.058	342.6
8	0.0067	0.052	435.8
9	0.0065	0.047	526.6
10	0.0064	0.043	616.7

$$\psi(\omega)=\frac{\psi_0}{\omega}+\psi_v(\omega)\ . \tag{8.9b}$$

Inserting this ansatz into (8.5) and (8.6) and expanding in powers of ψ_v, one is easily led to a determination of ψ_0 and the value of the coupling ($\lambda^*=4$) where the solution (8.9) is first valid. It is even more convincing if one rewrites (8.4) and (8.6) in the time domain where they can be expressed in terms of the single equation

$$\ddot{\psi}(t)+\Gamma_0\dot{\psi}(t)q^2+\psi(t)q^2+\lambda q^2\int_0^t \psi^2(t)\dot{\psi}(t-\tau)d\tau=0\ . \tag{8.10}$$

One can solve this equation numerically and verify that one does reach the instability postulated in the frequency domain. We show $\psi(t)$, resulting from (8.10), in Fig. 3 (in dotted lines) for various choices of λ near the transition. For $\lambda=4$ the viscosity diverges. For $\lambda>4$ the system is nonergodic and $\psi(t)$ does not decay to zero for long times.

These results lead to the picture developed in Refs. 6–9 for the glass transition. Let us now return to (8.4) where we retain the nonhydrodynamic quantity γ and again look at the situation where Γ becomes large. In this case, on letting Γ become arbitrarily large, we find that

$$G_{\rho\hat{\rho}}(\omega)=\frac{1}{\omega+i\gamma q^2}\ .$$

One might conclude at this point that all of the discussion of a dynamical glass transition is incorrect and one should move on to look for other mechanisms to associate with glass formation. We believe that this is too pessimistic a point of view. It is true that there is no sharp transition, but, as we now indicate, there is evidence that the basic mechanism does lead to a slowing down similar to that seen in the fragile glass systems. The basic idea, which we test numerically below, is that as one increases λ there will be a range where $\Gamma(0)$ will increase sharply and one will see a decrease in $I(0)\sim\Gamma(0)^{-1}$. As one increases λ further, however, one finds a crossover, γ is no longer inversely proportional to Γ, and the transition is cut off.

We can analyze these statements quantitatively by looking at the set of equations (8.4), (8.5), and (8.7) in the time domain where they can be written in the form

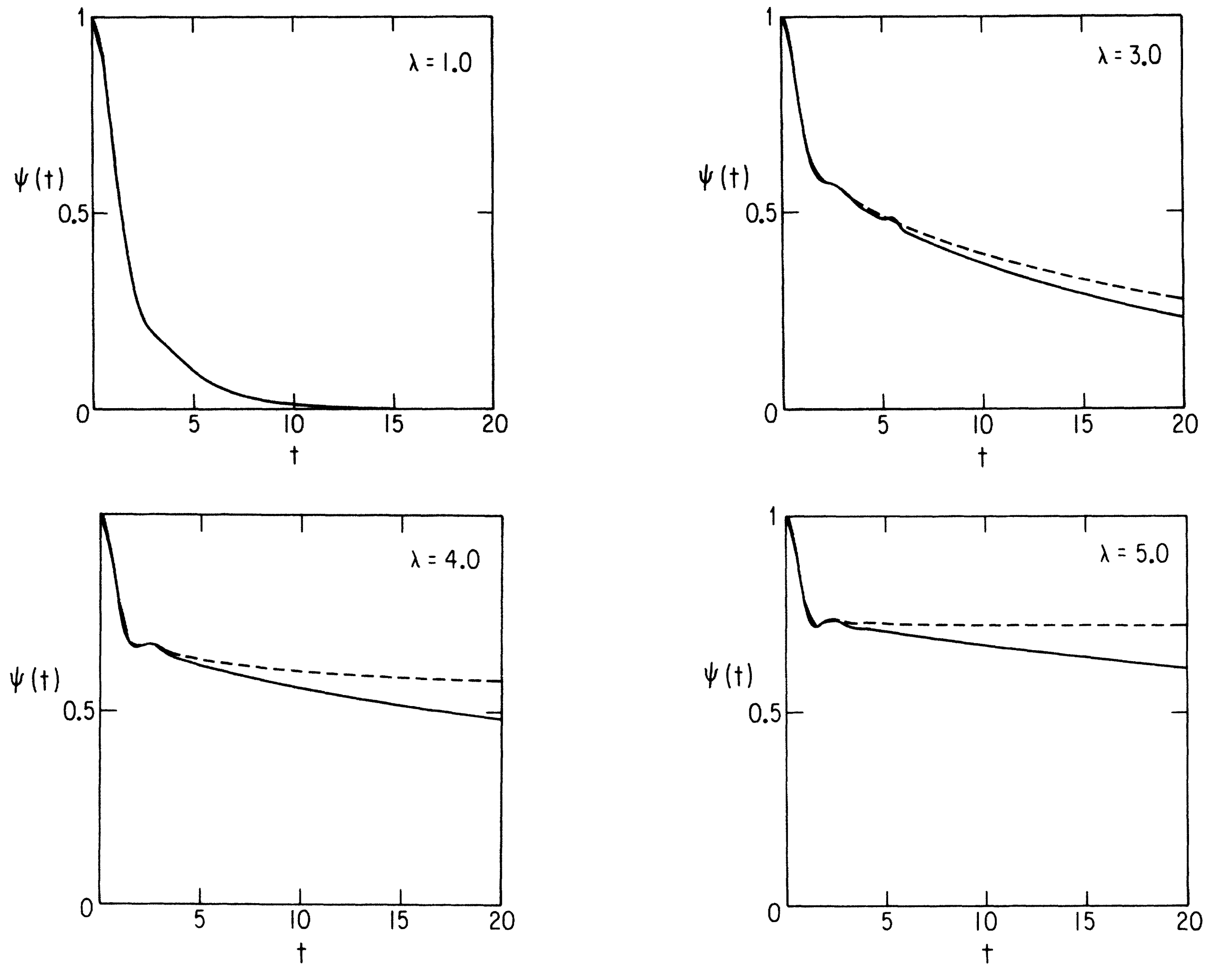

FIG. 3. The decay of the structure factor $\psi(t)$ with time t, for $\lambda=1.0$, 3.0, 4.0, and 5.0. The solid lines indicate the results of numerical integration of (8.11). The dotted line corresponds to the case where the γ correction is ignored. The value of q/Λ used is $\frac{1}{8}$.

$$\ddot{\psi}(t)+q^2\lambda_1\left[\dot{\psi}(t)+\lambda\int_0^t d\tau\,\dot{\psi}^2(t-\tau)\psi(\tau)+\frac{q^2\lambda}{3}\int_0^t d\tau\,\dot{\psi}^2(t-\tau)\left[\psi(\tau)+\lambda\int_0^\tau d\tau'\,\psi^2(\tau-\tau')\psi(\tau')\right]\right]=0\,, \tag{8.11}$$

where

$$\lambda_1=\frac{\Lambda^2\Gamma_0^2}{c^2}\,. \tag{8.12}$$

We have solved (8.11) numerically using the same choice for λ_1 $(=1)$ used by Leutheusser, and for which we know there is a transition for $\lambda=4$. In Fig. 3 we show $\psi(t)$ versus t for several choices of λ. This should be compared with the case where the γ correction is ignored (shown in dotted lines). In the figure one sees that the system continues to decay even for $\lambda>4$. A detailed analysis of the data shows that there is exponential decay at sufficiently long time and the decay rate, σ, defined by

$$\sigma=-\dot{\psi}/\psi \tag{8.13}$$

is given in Table V as a function of λ. We also give there $\Gamma(0)$ and $\gamma(0)$ as functions of λ. One finds the type of behavior described above. Looking in particular at the behavior of Γ, we see that there are two types of behavior. For larger λ one finds that Γ is essentially linear with λ:

$$\Gamma(0)=-308.69+92.76\lambda \tag{8.14}$$

and the fit is excellent for $\lambda>3.75$. Notice that if we extrapolate this result to small λ one finds that Γ vanishes at $\lambda=3.33$. For small λ, motivated by the previous theoretical work and the experiments of Toborak *et al.*,[17] we have plotted $\Gamma^{-1/2}(0)$ versus λ and find a reasonable fit for $1<\lambda<4$:

$$\Gamma^{-1/2}(0)=0.874-0.195\lambda\,.$$

Notice that this result predicts a transition for $\lambda=4.48$. Thus there is a remnant of the Leutheusser transition for this choice of parameters. Analysis of the $I(0)$ shows that it behaves like $0.364\lambda^{-0.93}$ for $2<\lambda<10$. Since γ is λ times $I(0)$, we see that γ increases slowly as λ increases.

The conclusion we draw is that density fluctuations are effective in increasing the viscosity, but they do not, by themselves, appear sufficient to give the strong temperature dependences seen in the more strongly coupled glassy systems [or very near T_G in fragile systems (see Ref. 2)]. These conclusions must be subjected to the following restrictions. (i) We have treated the wave-number dependences of this system in a very crude and quantitatively unsatisfactory way. As emphasized by Kirkpatrick,[9] the slowing down for dense fluids should be correlated with the peak in the structure factor which should play an important role in the analysis. (ii) Our identification of the parameter λ with increasing density and lower temperatures is not credible unless we appeal to the correlations mentioned in (i). Therefore our numerical analysis above can only be thought of as suggestive and comparison with experiments must await calculations which treat finite wave-number effects more carefully.

IX. CONCLUSIONS

We have looked at the effects of nonlinearities on the fluctuating hydrodynamics of compressible fluids. One of the main motivations was to investigate the range of validity of the density-driven dynamic instability proposed by Leutheusser. In previous work[8] it had been shown that this mechanism is present in a hydrodynamic approach. The main question was whether there exists a mechanism which cuts off this instability. We have uncovered such a cutoff in this paper. It arises from nonlinear density fluctuations just as with the original mechanism driving the instability. We feel that it is important to understand that such a cutoff enters naturally into the analysis within the perturbation theory presented here. It has been emphasized elsewhere[30] that it is important to deal with a model for compressible fluids with a realistic Poisson bracket structure. The structure and origins of the density nonlinearities studied by Siggia[31] differ significantly from those studied in Ref. 8. In particular, the cutoff mechanism discovered here is not present in the model studied by Siggia. Similarly, the origins of the density fluctuations leading to a "fake" instability in Ref. 30 are driven by nonlinearities in the effective Hamiltonian and not by the dynamical nonlinearities as in Ref. 8.

From one point of view our findings must be viewed as discouraging. Since this is no sharp transition, there is nothing particularly robust about the glass transition region. This means there are no clear-cut predictions such as exponents and transition temperatures. On the other hand, there is some remnant of the sharp transition and the cutoff mechanism does seem to be related to a nonhydrodynamic diffusion process. It may well be that a more ambitious calculation which takes into account correlations developing for wave numbers near the first structure factor maximum will lead to a more pronounced effect. We are planning to undertake such an analysis.

ACKNOWLEDGMENTS

We thank Dr. Sriram Ramaswamy and Dr. John Toner for many useful discussions and for their encouragement. We thank Dr. Norman Birge, Dr. Ted Kirkpatrick, Dr. Sidney Nagel, Dr. David Oxtoby, and Dr. Sidney Yip for useful discussions. This work was supported by the National Science Foundation Grant No. DMR-85-12901 and by the National Science Foundation Materials Research Laboratory at the University of Chicago.

APPENDIX A

To prove the relation (6.1), we note first, from (3.18), since ψ and $\hat{\psi}$ are real, that $A_U^*[\psi,-\hat{\psi}]=A_U[\psi,\hat{\psi}]$. Thus the quantity Z_U, defined by (6.10), is real. Similarly, if α is an unhatted variable, then

$$G_{\alpha\hat\beta}(1,2)=-G^*_{\alpha\hat\beta}(1,2)\ . \tag{A1}$$

Consider then the Fourier transform

$$G_{\alpha\hat\beta}(\mathbf{q},\omega)=\int d^3x_1\, e^{+i\mathbf{q}\cdot(\mathbf{x}_1-\mathbf{x}_2)} \times \int dt_1\, e^{-i\omega(t_1-t_2)}G_{\alpha\hat\beta}(1,2) \tag{A2}$$

and its complex conjugate

$$G^*_{\alpha\hat\beta}(\mathbf{q},\omega)=\int d^3x_1 \int dt_1\, e^{-i\mathbf{q}\cdot(\mathbf{x}_1-\mathbf{x}_2)} \times e^{+i\omega(t_1-t_2)}[-G_{\alpha\hat\beta}(1,2)]\ , \tag{A3}$$

where we have used (A1). Letting $\mathbf{x}_1-\mathbf{x}_2\to\mathbf{x}_2-\mathbf{x}_1$, $t_1-t_2\to t_2-t_1$, we have, using space and time translational invariance, $G_{\alpha\hat\beta}(\mathbf{x}_2-\mathbf{x}_1,\ t_2-t_1)=G_{\alpha\hat\beta}(2,1)$. However, since

$$G_{\alpha\hat\beta}(1,2)=G_{\hat\beta\alpha}(1,2) \tag{A4}$$

we obtain

$$G^*_{\alpha\hat\beta}(\mathbf{q},\omega)=-G_{\hat\beta\alpha}(\mathbf{q},\omega)\ . \tag{A5}$$

Consider next the expressions (6.24) and (6.40) for both the longitudinal and the transverse cases:

$$G_{\alpha\beta}=-\sum_{\gamma,\delta} G_{\alpha\hat\gamma}C_{\hat\gamma\hat\delta}G_{\hat\delta\beta}\ . \tag{A6}$$

Taking the matrix inverses from the right and left we obtain

$$C_{\hat\alpha\hat\beta}=-\sum_{\gamma,\delta} G^{-1}{}_{\hat\alpha\gamma}G_{\gamma\delta}G_{\delta\hat\beta}{}^{-1}\ . \tag{A7}$$

Focus on the complex conjugate

$$C^*_{\hat\alpha\hat\beta}=-\sum_{\gamma,\delta}(G^{-1})^*_{\hat\alpha\gamma}G^*_{\gamma\delta}(G^{-1})^*_{\delta\hat\beta}\ . \tag{A8}$$

It follows from (A6) that

$$(G^{-1})^{-1}_{\hat\alpha\gamma}=-G^{-1}{}_{\gamma\hat\alpha} \tag{A9}$$

and, since G is real and symmetric,

$$C_{\hat\alpha\hat\beta}=(C_{\hat\beta\hat\alpha})^*\ . \tag{A10}$$

APPENDIX B

To deduce the relation (6.8) we start with the general identity

$$\int D(\psi)\frac{\delta}{\delta\psi_{\hat\alpha}(1)}[\psi_\beta(2)e^{-A[\psi]}]=0\ . \tag{B1}$$

Using the MSR action (3.16) and choosing $\hat\alpha=\hat g$ in (B1), we obtain

$$2\beta^{-1}L_{ij}G_{\hat g_i\beta}(1,2)+i\frac{\partial}{\partial t_1}G_{g_i\beta}(1,2)+i\left\langle\left[\rho(1)\nabla_i\frac{\delta F_u}{\delta\rho(1)}+\sum_j\nabla_j(\rho(1)V_i(1)V_j(1))\right]\psi_\beta(2)\right\rangle+i\langle L_{ij}V_j(1)\psi_\beta(2)\rangle=0\ . \tag{B2}$$

Changing t_1 and t_2 to $-t_1,-t_2$, respectively,

$$2\beta^{-1}L_{ij}G_{\hat g_i\beta}(\mathbf{x}_1-t_1,\mathbf{x}_2-t_2)+i\frac{\partial}{\partial t_1}\epsilon_\beta G_{g_i\beta}(1,2) +i\epsilon_\beta\left\langle\left[\rho(1)(\nabla_i\delta F_u/\delta\rho(1))+\sum_j\nabla_j(\rho(1)V_i(1)V_j(1))\right]\psi_\beta(2)\right\rangle+i\epsilon_\beta\langle L_{ij}V_j(1)\psi_\beta(2)\rangle=0 \tag{B3}$$

where ϵ_β is the signature of ψ_β under time reversal and β is an unhatted variable. Multiplying (B3) by ϵ_β and subtracting from (B2) we obtain

$$2\beta^{-1}L_{ij}[G_{\hat g_j\beta}(t_1,t_2)-\epsilon_\beta G_{\hat g_j\beta}(\mathbf{x}_1,-t_1\mathbf{x}_2,-t_2)] =-2iL_{ij}G_{V_j\beta}(1,2)\ . \tag{B4}$$

Since the response function $G_{\hat g_i\beta}(1,2)$ is advanced, (B4) is equivalent to

$$\theta(t)G_{V_i\beta}(1,2)=i\beta^{-1}G_{\hat g_i\beta}(1,2)\ . \tag{B5}$$

Fourier transforming (B5) over space and time, we obtain

$$C_{V_i\beta}(\mathbf{q},\omega)=\beta^{-1}G_{\hat g_i\beta}(\mathbf{q},\omega) \tag{B6}$$

or

$$G_{V_i\beta}(\mathbf{q},\omega)=-2\beta^{-1}\mathrm{Im}G_{\hat g_i\beta}(\mathbf{q},\omega)\ , \tag{B7}$$

where the fluctuation function $C_{\alpha\beta}(\mathbf{q},\omega)$ is just the Laplace transform of the correlation function

$$C_{\alpha\beta}(\mathbf{q},\omega)=-i\int_0^{+\infty}dt\, e^{+i\omega t}G_{\alpha\beta}(\mathbf{q},t) \tag{B8}$$

(ω is assumed to have a small positive imaginary part).

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