PHYSICAL REVIEW E VOLUME 47, NUMBER 3 MARCH 1993

# Supercooling of a nematic liquid crystal

P. De*
*Department of Physics, University of Rhode Island, Kingston, Rhode Island 02881*

Robert A. Pelcovits
*Department of Physics, Brown University, Providence, Rhode Island 02912*

E. Vogel and J. Vogel
*Institut für Theoretische Physik, Rheinisch-Westfälische Technische Hochschule Aachen, Templergraben 55, W-5100 Aachen, Germany*
(Received 12 October 1992)

We investigate the supercooling of a nematic liquid crystal using fluctuating nonlinear hydrodynamic equations. The Martin-Siggia-Rose formalism [Phys. Rev. A **8**, 423 (1973)] is used to calculate renormalized transport coefficients to one-loop order. Similar theories for isotropic liquids have shown substantial increases of the viscosities as the liquid is supercooled or compressed due to feedback from the density fluctuations which are freezing. We find similar results here for the longitudinal and various shear viscosities of the nematic phase. However, the two viscosities associated with the nematic-director motion do not grow in any dramatic way; i.e., there is no apparent freezing of the director modes within this hydrodynamic formalism. Instead a glassy state of the nematic phase may arise from a "random-anisotropy" coupling of the director to the frozen density.

PACS number(s): 64.70.Md

## I. INTRODUCTION

The study of a supercooled nematic liquid crystal and the possible formation of a nematic glass [1] is potentially richer than corresponding studies of supercooled simple fluids. The presence of anisotropy in the nematic liquid due to the overall alignment of the molecular long axes introduces orientational degrees of freedom into the description of the system and yields a model similar to a spin glass with translational degrees of freedom coupled to the spin fluctuations. We use the term "nematic glass" to describe a liquid crystal where both the translational (density) and orientational (director) fluctuations are frozen. Assuming that we supercool the liquid starting from its nematic phase (rather than the isotropic phase), we expect a nematic glass to have long-range orientational order and thus be similar to a mixed magnetic phase where both ferromagnetic and spin-glass order coexist [2]. The presence of both translational (density) and orientational (director) fluctuations and their coupling leads to a novel glass-forming system [3]. Glass formation in this system could potentially occur in a two-stage process where the density or director modes freeze first, followed by the other, or in a process where both freeze simultaneously.

In recent years a theoretical approach to the study of glass formation has been developed using either mode-coupling calculations [4] or fluctuating nonlinear hydrodynamics [5]. These approaches were initiated by Leutheusser [4], who showed that a model of a dense fluid obtained from kinetic theory exhibits a sharp glass transition where the system becomes nonergodic. Das, Mazenko, Ramaswamy, and Toner [5] developed an equivalent model on the basis of fluctuating nonlinear dynamics. Subsequently, Das and Mazenko [6] discovered a nonhydrodynamic mechanism which cuts off the sharp transition, leading to a rounded transition. These authors claim that the cutoff is due to the proper mathematical treatment of the relationship $\mathbf{P}=\rho\mathbf{V}$, where $\mathbf{P}$ is the momentum density, $\rho$ is the mass density, and $\mathbf{V}$ is the velocity field. The physical origin of this cutoff is unknown. More recently, Schmitz, Dufty, and De [7] have argued that the calculation of Das and Mazenko is not correct. Furthermore, they claim that the perturbative calculations of Refs. [5] and [6] do not properly account for detailed balance. Restoring detailed balance to the perturbation theory apparently restores ergodicity and leads to a rounded transition. However, Das and Mazenko and Schmitz, Dufty and De all agree that the original Leutheusser theory as well as its hydrodynamic version with no cutoff mechanism are a good approximation to the more complete theories of Refs. [6] and [7] in describing the growth of the viscosities in the preglass transition regime. Eventually, the growth of the viscosities is limited by one of these cutoff mechanisms. Comparison of these theories with experiments yields some encouraging agreement [8], though this agreement is by no means complete, especially at very low frequencies.

In this paper we study the formation of a nematic glass using fluctuating nonlinear hydrodynamics. We will limit our attention primarily to the preglass transition regime in light of the discussion above, and we will not concern ourselves with the question of detailed balance or the Das-Mazenko cutoff mechanism. The primary advantage of the fluctuating hydrodynamic theory is that new slow variables (such as the director modes or variables associated with broken translational symmetries [9]) are readily incorporated. As described in detail in the next section, we supplement the nonlinear hydrodynamic equations used by Das *et al.* to study simple fluids, with two equations describing the dynamics of the director in a compressible nematic. The full set of equations we employ also includes a nonlinear coupling $(\hat{\mathbf{n}}\cdot\nabla\rho)^2$ between the density and director $\hat{\mathbf{n}}$. Density fluctuations couple to some, but not all, of the nematic viscosities through this coupling. The Leutheusser feedback mechanism then leads to the enhancement of these viscosities with a universal prediction for their power-law behavior. However, there is apparently no director feedback mechanism within this formalism, and we find no evidence for the freezing of the director modes. Nevertheless, it is tempting to speculate that the term $(\hat{\mathbf{n}}\cdot\nabla\delta\rho)^2$ in the free energy leads to freezing of the director. Once the density has frozen, or almost frozen, this term will mimic a random anisotropy field in an amorphous magnet [10]. The frozen density gradients $\nabla\rho$ play the role of the quenched random axis. This model is believed to exhibit a spin-glass phase, which would correspond in our case to frozen director modes, with *no* nematic long-range order in an infinite system. This destruction of long-range orientational order in an anisotropic glass was first suggested by Golubovic and Lubensky [3], who found that random internal stresses will destroy long-range order in three dimensions or less. However, for a finite-sized system, there would be apparent long-range order, especially if the coefficient of our biquadratic term is small. Whether this proposed freezing of the director takes place immediately upon freezing of the density or requires further supercooling is unclear and beyond the scope of our present theoretical treatment.

This paper is organized as follows. In the next section we formulate the nonlinear hydrodynamic equations for a compressible nematic including the above-mentioned nonlinear coupling of density and director modes. In Sec. III we use the Martin-Siggia-Rose formalism to study the effects of the nonlinearities on the bare transport coefficients. Finally, in Sec. IV we discuss the implications of our calculations for the growth of the viscosities and the mode structure as the nematic is supercooled. Various technical details appear in the Appendix.

## II. NONLINEAR HYDRODYNAMIC EQUATIONS

In contrast to the spherical molecules of simple liquids, the molecules of liquid crystals are elongated in shape [11]. Intermolecular interactions cause the anisotropic molecules to align along a preferred direction denoted by the director vector $\mathbf{n}(\mathbf{x},t)$. Fluctuations in the director can extend over macroscopic distances and decay over finite times, leading to new hydrodynamic modes in addition to the shear and sound wave modes of a simple liquid. The broken rotational symmetry also permits more viscosities: in a compressible nematic liquid crystal, there are six independent viscosities [12]. The linear equations for the hydrodynamic modes of a liquid crystal have been known for almost 20 years [13]. A systematic method for deriving the nonlinear contributions is to write them in the form of generalized Langevin equations [14]:

$$\frac{\partial\psi_i}{\partial t}=\bar{V}_i[\psi]-\sum_j\int d^3x'\,\Gamma_{ij}(\mathbf{x}')\frac{\delta H}{\delta\psi_j(\mathbf{x}')}+\Theta_i\ , \qquad (2.1)$$

where $\psi_i(\mathbf{x},t)$ represents one of the seven possible hydrodynamic fields: the mass density $\rho(\mathbf{x},t)$, three components of the momentum density $\mathbf{P}(\mathbf{x},t)$, the energy density $e(\mathbf{x},t)$, and two components of the fixed-length director $\mathbf{n}(\underline{\mathbf{x}},t)$. The label $i$ in this equation denotes the type of field, as well as the vector index on $\mathbf{P}$ and $\mathbf{n}$. $\bar{V}_i[\psi]$ represents the reversible part of the dynamic equations and is given by

$$\bar{V}_i[\psi(\mathbf{x})]=\sum_j\int d^3x'\{\psi_i(\mathbf{x}),\psi_j(\mathbf{x}')\}\frac{\delta H}{\delta\psi_j(\mathbf{x}')}\ , \qquad (2.2)$$

and $H$ is the energy obtained by integrating the free-energy density $F[\psi]$:

$$H=\int d^3x\,F[\psi(\mathbf{x})]\ . \qquad (2.3)$$

The Poisson bracket in Eq. (2.2) is defined in its usual manner as

$$\{\psi_i,\psi_j\}=\sum_{\alpha,\beta,k}\left[\frac{\delta\psi_i}{\delta r_k^{\alpha\beta}}\frac{\delta\psi_j}{\delta P_k^{\alpha\beta}}-\frac{\delta\psi_i}{\delta P_k^{\alpha\beta}}\frac{\delta\psi_j}{\delta r_k^{\alpha\beta}}\right]\ , \qquad (2.4)$$

where $r_k^{\alpha\beta}$ is the $k$th component of the vector $\mathbf{r}^{\alpha\beta}$, which points to the $\beta$th atom of the $\alpha$th molecule of the liquid crystal. The second term on the right-hand side of the equation of motion (2.1) represents the dissipative contributions in form of the dissipative matrix $\Gamma_{ij}(\mathbf{x},\mathbf{x}')$. Finally, $\Theta_i(\mathbf{x},t)$ denotes a Gaussian noise source which satisfies

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

It has been argued that the dominant transport anomalies at the glass transition in a simple liquid are due to the slow decay of density fluctuations, and the effects of energy fluctuations can be ignored [4,5]. We shall denote the global preferred direction of orientation by $\mathbf{n}_0=\mathbf{e}_z$. Local fluctuations in orientation are specified by $\delta\mathbf{n}=\mathbf{n}-\mathbf{n}_0$, where we restrict $\delta\mathbf{n}=(\delta n_x,\delta n_y,0)$ to linear order in $\delta n_i$ [15]. Then $\mathbf{n}$ satisfies $\mathbf{n}\cdot\mathbf{n}=1$ up to order $(\delta\mathbf{n})^2$. Furthermore, one can assume that the wave vector $\mathbf{k}$ of a disturbance lies in the $x-z$ plane. This allows us to treat $\delta n_x$ and $\delta n_y$ as longitudinal and transverse fluctuations with respect to $\mathbf{k}$, respectively [16]. Our set of dynamical variables $\psi_i$ then includes the density, three components of the momentum, and the director fluctuations $n_x$ and $n_y$, which are, respectively, equal to $\delta n_x$ and $\delta n_y$ in the present approximation.

In order to evaluate the Poisson bracket between the six dynamical variables $\rho$, $P_x$, $P_y$, $P_z$, $n_x$, and $n_y$, we need a microscopic description for each variable. We define

$$\rho(\mathbf{x},t)=\sum_{\alpha,\beta}m^{\beta}\delta(\mathbf{x}-\mathbf{r}^{\alpha\beta}(t))\ , \qquad (2.6)$$

$$P_i(\mathbf{x},t)=\sum_{\alpha,\beta}P_i^{\alpha\beta}(t)\delta(\mathbf{x}-\mathbf{r}^{\alpha\beta}(t))\ , \qquad (2.7)$$

$$n_i(\mathbf{x},t)=\frac{1}{\sqrt{N}}\sum_{\alpha} n_i^{\alpha}(t)\delta_{\mathbf{x},\mathbf{R}^{\alpha}} . \tag{2.8}$$

We model an elongated molecule with two atoms only: $\mathbf{R}^{\alpha}$ is the center of mass of the $\alpha$th nematic molecule and $n_i^{\alpha}=r_i^{\alpha}/|\mathbf{r}^{\alpha}|$, and $\mathbf{r}^{\alpha}$ is the relative vector between the atoms, which, we shall assume, model an elongated molecule of mass $m^{\alpha}$. The variable $\beta$ in $\mathbf{r}^{\alpha\beta}$ takes on values 1 or 2, and $\mathbf{r}^{\alpha}=\mathbf{r}^{\alpha 1}-\mathbf{r}^{\alpha 2}$, while $\mathbf{R}^{\alpha}=(\mathbf{r}^{\alpha 1}+\mathbf{r}^{\alpha 2})/2$.

With the definitions (2.6)–(2.8), the Poisson brackets required in (2.2) and defined in (2.4) can be computed in a straightforward manner with the following results:

$$\{\rho(\mathbf{x}),P_j(\mathbf{x}')\}=\nabla_j(\rho(x)\delta(\mathbf{x}-\mathbf{x}')) , \tag{2.9}$$

$$\{P_i(\mathbf{x}),P_j(\mathbf{x}')\}=-\nabla_j((\delta(\mathbf{x}-\mathbf{x}')P_i(\mathbf{x}))+\nabla_i'(\delta(\mathbf{x}-\mathbf{x}')P_j(\mathbf{x}')) , \tag{2.10}$$

$$\{P_i(\mathbf{x}),n_j(\mathbf{x}')\}=[(\lambda+1)\delta_{ij}n_k(\mathbf{x}')/2+(\lambda-1)\delta_{kj}n_i(\mathbf{x}')/2-\lambda(n_in_jn_k)(\mathbf{x}')]\nabla_k[\delta(\mathbf{x}-\mathbf{x}')]+\delta(\mathbf{x}-\mathbf{x}')(\nabla_i'n_j(\mathbf{x}')) , \tag{2.11}$$

where $\nabla_i\equiv\partial/\partial x_i$ and $\nabla_i'\equiv\partial/\partial x_i'$, etc. In writing Eq. (2.11) we have introduced the "form factor" $\lambda$, which is related to the shape of the molecules and equals unity only in the limit of infinitesimally thin molecules [17]. All other Poisson brackets are zero.

The final step in evaluating $\bar{V}_i[\psi]$ is to calculate $\delta H/\delta\psi_j$. Expressing the free-energy density $F(\rho,\mathbf{P},\mathbf{n})$ as a sum of kinetic and potential energies, we rewrite (2.3) as

$$H=\int d^3x\,(\varepsilon_k+\varepsilon_u^{\rho}+\varepsilon_u^{n}+\varepsilon_u^{c}) , \tag{2.12}$$

where $\varepsilon_k$ is the kinetic-energy density of the molecules, $\varepsilon_u^{\rho}$ is the potential-energy density due to density fluctuations, and $\varepsilon_u^{n}$ is that due to the director fluctuations. Finally, $\varepsilon_u^{c}$ is an energy density due to the coupling of density and director fluctuations. For the kinetic-energy density we have

$$\varepsilon_k(\mathbf{x},t)=\frac{\mathbf{P}^2(\mathbf{x},t)}{2\rho(\mathbf{x},t)} . \tag{2.13}$$

For the potential-energy density of the density fluctuations we choose the simplest form incorporating $\nabla\rho$:

$$\varepsilon_u^{\rho}(\mathbf{x},t)=\frac{A}{2}[\delta\rho(\mathbf{x},t)]^2+\frac{B}{2}[\nabla\rho(\mathbf{x},t)]^2 , \tag{2.14}$$

where $A$ and $B$ are phenomenological constants, and $\delta\rho=\rho(\mathbf{x},t)-\rho_0$, with $\rho_0$ being the uniform density. The gradient term in (2.14) is rotationally isotropic, which is unrealistic in an anisotropic system like the nematic phase. We will incorporate the effects of anisotropy below in $\varepsilon_u^{c}$, the coupling of $\nabla\rho$ to $\hat{\mathbf{n}}$.

The simplest choice for $\varepsilon_u^{n}$ is

$$\varepsilon_u^{n}(\mathbf{x},t)=\tfrac{1}{2}K[\nabla_in_j(\mathbf{x},t)][\nabla_in_j(\mathbf{x},t)] , \tag{2.15}$$

where $K$ is a Frank elastic constant and repeated indices are summed over. In general, the symmetry of a nematic phase allows for three independent elastic constants [11] corresponding to the distortions of splay, twist, and bend. We have made the simplification of setting the three elastic constants equal to $K$. While this equality is broken upon renormalization (see Sec. III), the difference between the elastic constants is not large and we will ignore it.

Finally, in $\varepsilon_u^{c}$ we incorporate the expected anisotropy in density fluctuations and choose

$$\varepsilon_u^{c}(\mathbf{x},t)=\tfrac{1}{2}I[\mathbf{n}(\mathbf{x},t)\cdot\nabla\rho(\mathbf{x},t)]^2 , \tag{2.16}$$

where $I$ is a phenomenological coupling constant. In general, one might introduce a coupling of the form $(I/2)(\mathbf{n}\cdot\nabla\rho)^2+(I'/2)(\mathbf{n}\times\nabla\rho)^2$ to account for the expected anisotropy in the density fluctuations; i.e., the energy of such fluctuations should depend on the relative orientation of the director and the wave vector of the fluctuation. However, because of the vector identity

$$(\nabla\rho)^2=(\mathbf{n}\cdot\nabla\rho)^2+(\mathbf{n}\times\nabla\rho)^2 , \tag{2.17}$$

we can eliminate the coupling proportional to $I'$ in favor of a redefinition of $I$ and the inclusion of the last term in (2.14). This choice simplifies our perturbation theory in Sec. III, and allows $I$ to be negative. However, rodlike molecules will probably be characterized by positive values of $I$, since the director will prefer to align perpendicular to the wave vector of the density field. We can think of (2.14) and (2.16) together as providing a simple model for the static structure factor of the nematic phase, where density correlations are not isotropic in space but depend on the local director orientation. The effect of local structure on a simple fluid as it is supercooled was considered by Das [18].

Using (2.9)–(2.17) we can evaluate the reversible terms in (2.1). For the $\mathbf{P}$ equation of motion we can express the result conveniently in terms of the divergence of a reactive stress tensor, i.e.,

$$\bar{V}_{\mathbf{P}_i}=-\sum_j\nabla_j\sigma_{ij}^{P,R},\quad i,j=x,y,z , \tag{2.18}$$

where

$$\sigma_{ij}^{P,R}=\frac{P_iP_j}{\rho}+\delta_{ij}\left[\tfrac{1}{2}\chi^{-1}(\delta\rho)^2-\tfrac{1}{2}K(\nabla_kn_l)^2-\rho I\nabla_k(n_k(\mathbf{n}\cdot\nabla\rho))+\frac{I}{2}(\mathbf{n}\cdot\nabla\rho)^2\right]+I(\mathbf{n}\cdot\nabla\rho)n_j\nabla_i\rho+K(\nabla_in_l)(\nabla_jn_l)-\sum_k\mu_{ijk}[-K\nabla^2n_k+J(\mathbf{n}\cdot\nabla\rho)\nabla_k\rho] \tag{2.19}$$

and

$$\mu_{ijk}=\tfrac{1}{2}(\lambda+1)n_j\delta_{ik}+\tfrac{1}{2}(\lambda-1)n_i\delta_{jk}-\lambda n_i n_j n_k \ . \tag{2.20}$$

The stress tensor appearing in (2.18) is not symmetric. However, the equation of motion for $\mathbf{P}$ is not sensitive to this asymmetry. A symmetric choice for $\sigma_{ij}^R$ would yield the same forces as well as guaranteeing conservation of angular momentum [12], which is not an issue in our analysis.

The reversible part of the director equation of motion is given by

$$\bar{V}_{n_i}=-\frac{1}{\rho}\mathbf{P}\cdot\nabla n_i+(\mathbf{\Omega}\times\mathbf{n})_i +\lambda\sum_{k=x,y,z}(n_k\delta_{ij}-n_j n_j n_k)A_{kj}, \quad i=x,y \ , \tag{2.21}$$

where

$$A_{kj}=\frac{1}{2\rho}(\nabla_k P_j+\nabla_j P_k) \tag{2.22}$$

and

$$\mathbf{\Omega}=\frac{1}{2\rho}(\nabla\times\mathbf{P}) \ . \tag{2.23}$$

A dissipative stress tensor can be introduced to write the dissipative contribution to (2.1). The dissipative momentum stress tensor $\sigma_{ij}^{PD}$ is defined via the relation

$$\sum_{j=x,y,z}\nabla_j\sigma_{ij}^{P,D}=\sum_{j=x,y,z}\Gamma_{P_iP_j}\frac{\delta H}{\delta P_j}, \quad i=x,y,z \ . \tag{2.24}$$

The uniaxial symmetry of the nematic dictates that $\sigma_{ij}^{P,D}$ has the following form [13]:

$$\begin{aligned}-\sigma_{ij}^{P,D}=\ &2(\nu_1^0+\nu_2^0-2\nu_3^0)n_k n_m A_{mk} n_i n_j\\ &+2(\nu_3^0-\nu_2^0)(n_i n_k A_{kj}+n_l n_k A_{ik})\\ &+(\nu_5^0-\nu_4^0+\nu_2^0)(n_i n_j A_{kk}+\delta_{ij}n_k n_m A_{km})\\ &+2\nu_2^0 A_{ij}+(\nu_4^0-\nu_2^0)A_{kk}\delta_{ij} \ .\end{aligned} \tag{2.25}$$

The five bare viscosity coefficients $\nu_1^0$, $\nu_2^0$, $\nu_3^0$, $\nu_4^0$, and $\nu_5^0$ are in general not equal to each other. If $\nu_1^0=\nu_2^0=\nu_3^0=\nu_4^0-\nu_5^0$, we recover the usual form for the dissipative stress tensor of a simple fluid. If the nematic phase is incompressible, then all terms in $\sigma_{ij}^{P,D}$ which are proportional to $A_{kk}$ must vanish, implying that $\nu_2^0=\nu_4^0$ and $\nu_5^0=0$, and three independent viscosities remain. As we are interested in density fluctuations, we will work with the full tensor displayed in (2.25). Comparing (2.24) and (2.25), we can identify the nonzero elements of the viscosity matrix $\Gamma_{P_iP_j}$:

$$\Gamma_{P_xP_x}=-(\nu_4^0+\nu_2^0)\nabla_x^2-\nu_3\nabla_z^2 \ , \tag{2.26}$$

$$\Gamma_{P_yP_y}=-\nu_2^0\nabla_x^2-\nu_3^0\nabla_z^2 \ , \tag{2.27}$$

$$\Gamma_{P_zP_z}=-\nu_3^0\nabla_x^2-(2\nu_1^0+\nu_2^0-\nu_4^0+2\nu_5^0)\nabla_z^2 \ , \tag{2.28}$$

$$\Gamma_{P_xP_z}=-(\nu_5^0+\nu_3^0)\nabla_x\nabla_z \ . \tag{2.29}$$

All other elements of the viscosity matrix $\Gamma_{P_iP_j}=0$. If we go to the limit of a simple fluid ($\nu_1^0=\nu_2^0=\nu_3^0=\nu_4^0-\nu_5^0$), then $\Gamma_{P_iP_j}$ reduces to the tensor $L_{ij}$, used in Refs. [5] and [18].

The dissipative contribution to the director equation is proportional to the "molecular field," $\delta H/\delta n_i$, with the proportionality constant conventionally written as $1/\gamma_1^0$, where $\gamma_1^0$ has the units of viscosity. Thus we identify

$$\Gamma_{n_in_j}=\frac{1}{\gamma_1^0}\delta_{ij}, \quad i,j=x,y \ . \tag{2.30}$$

All other elements of the viscosity matrix $\Gamma_{ij}$ are zero. In particular, all off-diagonal elements $\Gamma_{n_iP_j}$ can be shown to be zero on the basis of time-reversal symmetry [12].

The compressible nematic is thus characterized by six hydrodynamic fields and six independent viscosities: $\nu_1^0$, $\nu_2^0$, $\nu_3^0$, $\nu_4^0$, $\nu_5^0$, and $\gamma_1^0$. Another viscosity $\gamma_2$ commonly discussed in the literature which describes the torque exerted on the director by a shear flow is related to $\gamma_1$ via $\gamma_2=-\lambda\gamma_1^0$, where $\lambda$ is the reactive coefficient introduced in Eq. (2.11).

Our final nonlinear hydrodynamic equations for the compressible nematic are as follows:

$$\frac{\partial\rho}{\partial t}+\nabla\cdot\mathbf{P}=0 \ , \tag{2.31}$$

$$\frac{\partial P_i}{\partial t}=-\sum_{j=x,y,z}\nabla_j\sigma_{ij}^P, \quad i=x,y,z \ , \tag{2.32}$$

$$\frac{\partial n_i}{\partial t}=(\mathbf{\Omega}\times\mathbf{n})_i-\frac{1}{\rho}(\mathbf{P}\cdot\nabla)n_i +\lambda\sum_{k,l=x,y,z}A_{kl}n_k(\delta_{il}-n_i n_l) +\frac{K}{\gamma_1^0}\nabla^2 n_i-\frac{I}{\gamma_1^0}(\mathbf{n}\cdot\nabla\rho)\nabla_i\rho, \quad i=x,y \ , \tag{2.33}$$

where

$$\sigma_{ij}^P=\sigma_{ij}^{P,R}+\sigma_{ij}^{P,D} \ , \tag{2.34}$$

with $\sigma_{ij}^{P,R}$ and $\sigma_{ij}^{P,D}$ given by (2.19) and (2.25), respectively.

## III. TRANSPORT COEFFICIENTS

To investigate the effects of the nonlinearities in the equations of motion (2.31)–(2.34) on the transport properties of the nematic phase, we use the Martin-Siggia-Rose (MSR) formalism [19]. This formalism allows us to calculate the correlation functions $G_{ij}(\mathbf{x},t;\mathbf{x}',t')\equiv\langle\delta\psi_i(\mathbf{x},t)\delta\psi_j(\mathbf{x}',t')\rangle$, where $\psi_i$ represents any of our six hydrodynamic fields and the brackets refer to the average over the noise source $\Theta_i$ defined in (2.5). It will also enable us the calculate response functions, and obtain corrections to the bare viscosities introduced in the previous section. We refer the reader to Refs. [6] and [19] for full details on the MSR method; we summarize the essential ideas here.

We define a generating function $Z_U[\psi,\hat{\psi}]$ as follows:

$$Z_U[\psi,\hat{\psi}]=C\int D(\psi)D(\hat{\psi})\exp(-A_U[\psi,\hat{\psi}])\times\exp\int d\mathbf{x}\,dt\; U_i(\mathbf{x},t)\psi_i(\mathbf{x},t)\;, \quad (3.1)$$

where $C$ is a constant and $\psi$ collectively represents the six hydrodynamic fields $\psi_i$. Functional differentiation of $Z_U$ with respect to $U$ generates the correlation functions $G_{ij}$:

$$G_{ij}(\mathbf{x},t;\mathbf{x}',t')=\frac{\delta^2\ln Z_U}{\delta U_i(\mathbf{x},t)\delta U_j(\mathbf{x}',t')}\;. \quad (3.2)$$

The six auxiliary fields $\hat{\psi}$ were introduced to exponentiate each of the six hydrodynamic equations of motion (2.1). The integration over the noise source $\Theta_i$ has been replaced by an integration over the fields $\psi_i$. The action $A_U$ is given by

$$A_U[\psi,\hat{\psi}]=\int d\mathbf{x}\,dt\,d\mathbf{x}'dt'\,\hat{\psi}_i(\mathbf{x},t)\beta^{-1}\times\Gamma_{ij}(\mathbf{x},t;\mathbf{x}',t')\hat{\psi}_j(\mathbf{x}';t')+i\int d\mathbf{x}\,dt\,\hat{\psi}_j(\mathbf{x},t)\left[\frac{\partial\psi_j(\mathbf{x})}{\partial t}-\overline{H}_j[\psi]\right], \quad (3.3)$$

where $\overline{H}_j$ is defined by

$$\overline{H}_j[\psi]=\overline{V}_j[\psi]-\sum_j\int d\mathbf{x}'dt'\Gamma_{ij}(\mathbf{x}'t')\frac{\delta H}{\delta\psi_j(\mathbf{x}',t')}\;. \quad (3.4)$$

Introducing the "vector" $\phi=(\rho,\mathbf{P},n_x,n_y,\hat{\rho},\hat{\mathbf{P}},\hat{n}_x,\hat{n}_y)$, we can then rewrite $A_U[\psi,\hat{\psi}]\equiv A_U[\phi]$ as follows:

$$A_U[\phi]=\tfrac{1}{2}\int d1d2\sum_{\alpha,\beta}\phi_\alpha(1)G_0^{-1}(1,2)\phi_\beta(2)+\sum_{N=3}\frac{1}{N}\int d1d2\cdots dN\sum_{\alpha,\beta,\gamma,\ldots}V^N_{\alpha\beta\gamma\cdots}\phi_\alpha(1)\phi_\beta(2)\phi_\gamma(3)\cdots\;. \quad (3.5)$$

Here the integration variables $1,2,3,\ldots$ stand for $(\mathbf{x},t)$ and the sequence $\alpha\beta\gamma\cdots$ contains $N$ members. The explicit expressions $G_0^{-1}$ for the vertices $V_{\alpha\beta\gamma\cdots}$ will be presented later. The action $A_U$ generates the full nonlinear hydrodynamic equations for a nematic liquid. When we omit the vertices, what remains then is a pure quadratic Gaussian field theory. The corresponding $A_U$ contains the inverse of the linearized correlation matrix $G_0^{-1}$ and thus generates the linearized hydrodynamic equations of a nematic liquid. We first discuss this limit before doing perturbation theory in the vertices.

The linearized theory, as well as the perturbation theory, are most easily discussed in Fourier space. We Fourier transform (3.5) in space and time, recalling our discussion following (2.5), where we assumed that the wave vector $\mathbf{k}$ lies in the $x$-$y$ plane. The matrix $G_0^{-1}(\mathbf{k},\omega)$ is displayed in Table I, where we have introduced the following tensors for notational convenience:

$$\alpha_{ij}=\delta_{ij}\frac{K}{2}(\lambda+1)k_z+\delta_{ix}\delta_{jz}\frac{K}{2}(\lambda-1)k_x\;, \quad (3.6)$$

$$L^0_{ij}=\delta_{ij}\delta_{jx}\Gamma^0_{xx}+\delta_{iy}\delta_{jy}\Gamma^0_{yy}+\delta_{iz}\delta_{jz}\Gamma^0_{zz}+(\delta_{ix}\delta_{jz}+\delta_{ij}\delta_{jz})\Gamma^0_{xz}\;, \quad (3.7)$$

where $\Gamma^0_{xx}$, $\Gamma^0_{yy}$, $\Gamma^0_{zz}$, and $\Gamma^0_{xz}$ are the Fourier transforms of the viscosity matrix elements (2.26)–(2.29), respectively.

The inversion of $G_0^{-1}$ is most easily accomplished by decomposing $\mathbf{P}$ and $\mathbf{n}$ into longitudinal and transverse components. The calculation is straightforward though tedious, especially for the longitudinal portion. The elements of the matrix $G_0$ provide the physical correlation and response functions for the linearized theory. Correlation functions of any two hatted variables are identically zero. Correlation functions of unhatted variables can be found to leading order in $k^2$ from the corresponding

TABLE I. The zeroth-order matrix $G_0^{-1}$. The tensors $\alpha_{ij}$ and $L_{ij}$ are defined in Eqs. (3.6) and (3.7), and $\chi^{-1}=A+Bk^2$.

| | $\rho$ | $P_i$ | $n_j$ | $\hat{\rho}$ | $\hat{P}_j$ | $\hat{n}_j$ |
|---|---|---|---|---|---|---|
| $\rho$ | 0 | 0 | 0 | $-\omega$ | $\rho_0\chi^{-1}k_j$ | 0 |
| $P_i$ | 0 | 0 | 0 | $k_i$ | $-\omega\delta_{ij}+iL_{ij}/\rho_0$ | $-\frac{1}{\rho_0 K}\alpha_{ij}$ |
| $n_i$ | 0 | 0 | 0 | 0 | $-\alpha_{ij}k^2$ | $\delta_{ij}\left[\omega+\frac{iKk^2}{\gamma_1}\right]$ |
| $\hat{\rho}$ | $\omega$ | $-k_i$ | 0 | 0 | 0 | 0 |
| $\hat{P}_i$ | $-\rho_0\chi^{-1}k_j$ | $\omega\delta_{ij}+iL_{ij}/\rho_0$ | $\alpha_{ji}k^2$ | 0 | $2\beta^{-1}L_{ij}$ | 0 |
| $\hat{n}_i$ | 0 | $\frac{1}{\rho_0 K}\alpha_{ij}$ | $\delta_{ij}\left[\omega+\frac{iKk^2}{\gamma_1}\right]$ | 0 | 0 | $2\beta^{-1}\gamma_1^{-1}$ |

response functions via the fluctuation-dissipation theorems:

$$\langle \rho\rho \rangle = -2\beta^{-1}\chi \mathrm{Im}\langle \rho\hat{\rho} \rangle \ , \quad (3.8)$$

$$\langle n_x n_x \rangle = -2\beta^{-1}\chi_n \mathrm{Im}\langle n_x \hat{n}_x \rangle \ , \quad (3.9)$$

$$\langle n_y n_y \rangle = -2\beta^{-1}\chi_n \mathrm{Im}\langle n_y \hat{n}_y \rangle \ , \quad (3.10)$$

where $\chi$ is the static density structure factor given by $(A+Bq^2)^{-1}$ and $\chi_n$ is the static director structure factor given by $(Kk^2)^{-1}$. We could also write corresponding relations for the momentum correlation functions; however, they are not needed for elucidating the mode structure and calculating the corrections to the transport coefficients.

The density and director response functions in the linearized theory are given by the following expressions:

$$\langle \delta\rho(\mathbf{k},\omega)\delta\hat{\rho}(-\mathbf{k},-\omega) \rangle^0 = \frac{\omega\rho_0 + ik^2\Gamma_0}{\rho_0(\omega^2 - c_0^2k^2) + i\omega k^2\Gamma_0} \ , \quad (3.11)$$

$$\langle \delta n_x(\mathbf{k}\omega)\delta\hat{n}_x(-\mathbf{k},-\omega) \rangle^0 = \frac{\omega\rho_0 + ik^2\nu_L^0}{\rho_0(\omega + ik^2\tilde{\Gamma}_s^0)(\omega + ik^2\tilde{\Gamma}_f^0)} \ , \quad (3.12)$$

$$\langle \delta n_y(\mathbf{k},\omega)\delta\hat{n}_y(-\mathbf{k},-\omega) \rangle^0 = \frac{\omega\rho_0 + ik^2\nu_T^0}{\rho_0(\omega + ik^2\Gamma_s^0)(\omega + ik^2\Gamma_f^0)} \ . \quad (3.13)$$

The results are correct up to terms of relative order $k^2$. The viscosities appearing in (3.11)–(3.13) are given by

$$\Gamma_0 = -(\nu_1^0 + \nu_2^0 - 2\nu_3^0)\sin^2(2\theta)/2 + (\nu_2^0 + \nu_4^0)\sin^2\theta + (2\nu_1^0 + 2\nu_5^0 + \nu_2^0 - \nu_4^0)\cos^2\theta \ , \quad (3.14)$$

$$\nu_L^0 = \nu_3^0\cos^2(2\theta) + (\nu_1^0 + \nu_2^0)\sin^2(2\theta)/2 \ , \quad (3.15)$$

$$\nu_T^0 = \nu_2^0\sin^2\theta + \nu_3^0\cos^2\theta \ , \quad (3.16)$$

$$\tilde{\Gamma}_{s,f}^0 = \frac{1}{2}\left[\frac{\nu_L^0}{\rho_0} + \frac{K}{\gamma_1}\right] \pm \frac{1}{2}\left[\left[\frac{\nu_L^0}{\rho_0} + \frac{K}{\gamma_1}\right]^2 - \left[(1+\lambda\cos 2\theta)^2\frac{K}{\rho_0} + \frac{4K\nu_L^0}{\rho_0\gamma_1}\right]\right]^{1/2} \ , \quad (3.17)$$

$$\Gamma_{s,f}^0 = \frac{1}{2}\left[\frac{\nu_T^0}{\rho_0} + \frac{K}{\gamma_1}\right] \pm \frac{1}{2}\left[\left[\frac{\nu_T^0}{\rho_0} + \frac{K}{\gamma_1}\right]^2 - \left[(\lambda+1)^2\frac{K}{\rho_0}\cos^2\theta + \frac{4K\nu_T^0}{\rho_0\gamma_1}\right]\right]^{1/2} \ , \quad (3.18)$$

where $\theta$ is the angle between $\mathbf{k}$ and the $z$ axis.

In writing (3.17) and (3.18) we have assumed that the director modes appearing in (3.12) and (3.13) are diffusive rather than propagating. This is true for equilibrated nematics [13] where the orientational relaxation time of the director $(Kk^2/\gamma_1)^{-1}$ is small compared to the shear diffusion times $(\nu_L^0 k^2/\rho_0)^{-1}$ and $(\nu_T^0 k^2/\rho_0)^{-1}$. We shall see subsequently that this does not remain true when the nematic phase is supercooled or compressed rapidly, and propagating shear modes can appear. In the absence of supercooling, (3.17) and (3.18) can be approximated as follows:

$$\tilde{\Gamma}_f^0 \approx \nu_L^0/\rho_0 \ , \quad (3.19)$$

$$\tilde{\Gamma}_s^0 \approx \Gamma_s^0 \approx K/\gamma_1 \ , \quad (3.20)$$

$$\Gamma_f^0 \approx \nu_T^0/\rho_0 \ . \quad (3.21)$$

Thus there are two "slow" modes with viscosities $\Gamma_s^0$ and $\tilde{\Gamma}_s^0$ corresponding to the slow relaxation of director fluctuations, while the fast modes with viscosities $\Gamma_f$ and $\tilde{\Gamma}_f$ are like ordinary shear waves. In addition to these four modes we also have two sound modes appearing in (3.11), with speed $c_0$ and damping $\Gamma_0$.

We now calculate the corrections to the linearized theory due to the vertices $V^N$ appearing in (3.5). If we perform the rescalings $\psi \to \beta^{+1/2}\psi$ and $\hat{\psi} \to \beta^{-1/2}\hat{\psi}$, we see that the quadratic part of $A_U$ is $O((k_BT)^0)$, and the higher-order terms proportional to $V^N$ are of order $(k_BT)^{(N/2)-1}$. Thus we can systematically compute corrections to the linearized theory in powers of $k_BT$. In particular, one-loop diagrams will be $O(k_BT)$.

The inverse of the correlation matrix for the complete nonlinear theory satisfies the formal equation

$$G^{-1}(1,2) = G_0^{-1}(1,2) - \Sigma(1,2) \ , \quad (3.22)$$

which defines the self-energy $\Sigma$. We can then write corresponding equations for the renormalized transport coefficients by referring to Table I. As shown in Ref. [6], the renormalized viscosities are most readily obtained by looking at the renormalization of the terms in the action which are quadratic in the hatted fields. Thus the elements of the viscosity matrix $\Gamma_{ij}$ renormalize as follows:

$$\Gamma_{xx}(\mathbf{k},\omega) = \Gamma_{xx}^0 + \frac{\beta}{2}\Sigma_{\hat{P}_x\hat{P}_x}(\mathbf{k},\omega) \ , \quad (3.23)$$

$$\Gamma_{yy}(\mathbf{k},\omega) = \Gamma_{yy}^0 + \frac{\beta}{2}\Sigma_{\hat{P}_y\hat{P}_y}(\mathbf{k},\omega) \ , \quad (3.24)$$

$$\Gamma_{zz}(\mathbf{k},\omega) = \Gamma_{zz}^0 + \frac{\beta}{2}\Sigma_{\hat{P}_z\hat{P}_z}(\mathbf{k},\omega) \ , \quad (3.25)$$

$$\Gamma_{xz}(\mathbf{k},\omega) = \Gamma_{xz}^0 + \frac{\beta}{2}\Sigma_{\hat{P}_x\hat{P}_z}(\mathbf{k},\omega) \ , \quad (3.26)$$

$$1/\gamma_1(\mathbf{k},\omega) = \frac{1}{\gamma_1^0} + \frac{\beta}{2}\Sigma_{\hat{n}_x\hat{n}_x}(\mathbf{k}\omega) = \frac{1}{\gamma_1^0} + \frac{\beta}{2}\Sigma_{\hat{n}_y\hat{n}_y}(\mathbf{k},\omega) \ . \quad (3.27)$$

Referring to Table I we find that $K/\gamma_1$, $\lambda$, and $C$ renormalize as follows:

$$\frac{K}{\gamma_1}=\left[\frac{K}{\gamma_{1_0}}\right]+\frac{i}{k^2}\Sigma_{n_x\hat{n}_x}(\mathbf{k},\omega)$$

$$=\left[\frac{K}{\gamma_1}\right]_0+\frac{i}{k^2}\Sigma_{n_y\hat{n}_y}(\mathbf{k},\omega)\ , \tag{3.28}$$

$$\lambda=\lambda_0-\frac{2}{k_2}\Sigma_{\hat{n}_yP_y}(\mathbf{k},\omega)\ , \tag{3.29}$$

$$c^2=c_0^2=+\frac{1}{k_z}\Sigma_{\hat{P}_x\rho}\ . \tag{3.30}$$

The uniaxial symmetry of the nematic phase as well as conservation of momentum allows us to the write momenta self-energies as follows [cf. (2.26)–(2.29)]:

$$\Sigma_{\hat{P}_x\hat{P}_x}(\mathbf{k},\omega)=-k_x^2[\gamma_2(\mathbf{k},\omega)+\gamma_4(\mathbf{k},\omega)]-k_z^2\gamma_3(\mathbf{k},\omega)\ , \tag{3.31}$$

$$\Sigma_{\hat{P}_y\hat{P}_y}(\mathbf{k},\omega)=-k_x^2\gamma_2(\mathbf{k},\omega)-k_z^2\gamma_3(\mathbf{k},\omega)\ , \tag{3.32}$$

$$\Sigma_{\hat{P}_z\hat{P}_z}(\mathbf{k},\omega)=-k_x^2\gamma_3(\mathbf{k},\omega)-k_z^2[2\gamma_1(\mathbf{k},\omega)+\gamma_2(\mathbf{k},\omega)-\gamma_4(\mathbf{k},\omega)+2\gamma_5(\mathbf{k},\omega)]\ , \tag{3.33}$$

$$\Sigma_{\hat{P}_x\hat{P}_z}(\mathbf{k},\omega)=-k_zk_z[\gamma_3(\mathbf{k},\omega)+\gamma_5(\mathbf{k},\omega)]\ , \tag{3.34}$$

where the functions $\gamma_i(\mathbf{k},\omega)$, $i=1,\ldots,5$ renormalize the viscosities $\nu_i$ as follows:

$$\nu_i(\mathbf{k},\omega)=\nu_i^0+\frac{\beta}{2}\gamma_i(\mathbf{k},\omega),\quad i=1,\ldots,5\ . \tag{3.35}$$

In the hydrodynamic limit the response functions will have the same form as in the linearized theory, with the bare transport coefficients replaced by their renormalized values, which depend on $\mathbf{k}$ and $\omega$. As discussed in the Introduction, we are ignoring any possible nonhydrodynamic terms which might cut off a sharp transition, and focus instead on the behavior of the renormalized viscosities. Equations (3.14)–(3.18) will then be valid for the renormalized quantities, and we have the following relations for the generalized transport coefficients appearing in the response functions:

$$\Gamma(\mathbf{k},\omega)=\Gamma^0-\beta\left[\frac{k_x^2}{k^4}\Sigma_{\hat{P}_x\hat{P}_x}(\mathbf{k},\omega)+\frac{k_z^2}{k^4}\Sigma_{\hat{P}_z\hat{P}_z}(\mathbf{k},\omega)+2\frac{k_xk_z}{k^4}\Sigma_{\hat{P}_x\hat{P}_z}(\mathbf{k},\omega)\right]\ , \tag{3.36}$$

$$\nu_L(\mathbf{k},\omega)=\nu_L^0-\beta\left[\frac{k_z^2}{k^4}\Sigma_{\hat{P}_x\hat{P}_x}(\mathbf{k},\omega)+\frac{k_x^2}{k^4}\Sigma_{\hat{P}_z\hat{P}_z}(\mathbf{k},\omega)-\frac{2k_xk_z}{k^4}\Sigma_{\hat{P}_x\hat{P}_z}(\mathbf{k},\omega)\right]\ , \tag{3.37}$$

$$\nu_T(\mathbf{k},\omega)=\nu_T^0-\beta\left[\frac{1}{k^2}\right]\Sigma_{\hat{P}_y\hat{P}_y}(\mathbf{k},\omega)\ . \tag{3.38}$$

Using (3.32)–(3.35) we see that these renormalized viscosities are well behaved in the limit $\mathbf{k}\to 0$.

The preceding discussion indicates that we need to calculate the following self-energies in order to obtain the renormalized transport coefficients: $\Sigma_{\hat{P}_x\hat{P}_x}$, $\Sigma_{\hat{P}_y\hat{P}_y}$, $\Sigma_{\hat{P}_z\hat{P}_z}$, $\Sigma_{\hat{P}_x\hat{P}_z}$, $\Sigma_{\hat{n}_x\hat{n}_x}$, $\Sigma_{n_x\hat{n}_x}$, $\Sigma_{\hat{n}_yP_y}$, and $\Sigma_{\hat{P}_x\rho}$. We have done so to one-loop order under the following condition: we keep only those diagrams where there is a possibility of feedback from either density or director fluctuations. Thus we consider diagrams where the propagators are either $G_{\rho\rho}(\mathbf{k},\omega)$ or $G_{n_in_j}(\mathbf{k},\omega)$, $i,j=x,y$. (In the Appendix we show that mixed propagators cannot yield a feedback.) The diagrams will be bubble type and contain two three-point vertices. The symmetrized three-point vertices in the action (3.5) that contribute to the diagrams within our approximation are given by

$$V_{\alpha\beta\gamma}=\tfrac{1}{2}[\tilde{V}_{\alpha\beta\gamma}(1,2,3)+\tilde{V}_{\beta\alpha\gamma}(2,1,3)+\tilde{V}_{\gamma\beta\alpha}(3,2,1)+\tilde{V}_{\alpha\gamma\beta}(1,3,2)+\tilde{V}_{\beta\gamma\alpha}(2,3,1)+\tilde{V}_{\gamma\alpha\beta}(3,1,2)]\ , \tag{3.39}$$

where

$$\tilde{V}_{\alpha\beta\gamma}(1,2,3)=\sum_{i=1}^{11}\tilde{V}_{\alpha\beta\gamma}^{(i)}(1,2,3)\ , \tag{3.40}$$

and the $\tilde{V}^{(i)}$ are given in Fourier space by

$$\tilde{V}_{\alpha,\beta,\gamma}^{(1)}(1,2,3)=\frac{i}{2}\sum_{i,j}\delta_{\alpha\hat{P}_i}\delta_{\beta\rho}\delta_{\gamma\rho}i\{A\delta_{ij}(k_2+k_3)_j+B[(k_2+k_3)_jk_{3i}k_{3j}-(k_2+k_3)_ik_{2j}k_{3j}]\}\delta(1+2+3)(2\pi)^4\ , \tag{3.41}$$

$$\tilde{V}_{\alpha,\beta,\gamma}^{(2)}(1,2,3)=iK\sum_{j,l}\delta_{\alpha\hat{P}_j}\delta_{\beta n_l}\delta_{\gamma n_l}(i)^3k_{2j}k_3^2\delta(1+2+3)(2\pi)^4\ , \tag{3.42}$$

$$\tilde{V}_{\alpha,\beta,\gamma}^{(3)}(1,2,3)=\frac{i}{2}(\lambda+1)K\sum_{j,l}\delta_{\alpha\hat{P}_j}\delta_{\beta n_l}\delta_{\gamma n_j}(i)^3(k_{2l}+k_{3l})k_3^2\delta(1+2+3)(2\pi)^4\ , \tag{3.43}$$

$$\tilde{V}_{\alpha,\beta,\gamma}^{(4)}(1,2,3)=\frac{i}{2}(\lambda-1)K\sum_{j,l}\delta_{\alpha\hat{P}_j}\delta_{\beta n_j}\delta_{\gamma n_l}(i)^3(k_{2l}+k_{3l})k_3^2\delta(1+2+3)(2\pi)^4\ , \tag{3.44}$$

$$\tilde{V}_{\alpha,\beta,\gamma}^{(5)}(1,2,3)=-i\lambda K\sum_{j}\delta_{\alpha\hat{P}_z}\delta_{\beta n_j}\delta_{\gamma n_j}(i)^3(k_{2z}+k_{3z})k_3^2\delta(1+2+3)(2\pi)^4\ , \tag{3.45}$$

$$\tilde{V}^{(6)}_{\alpha,\beta,\gamma}(1,2,3)=-iI\sum_j \delta_{\alpha\hat{p}_j}\delta_{\beta\rho}\delta_{\gamma\rho}(i)^3(k_{3j}k_{3z}^2)\delta(1+2+3)(2\pi)^4 , \quad (3.46)$$

$$\tilde{V}^{(7)}_{\alpha,\beta,\gamma}(1,2,3)=-i\frac{I}{2}(\lambda+1)\sum_j \delta_{\alpha\hat{p}_j}\delta_{\beta\rho}\delta_{\gamma\rho}(i)^3(k_{2j}k_{3z})(k_{2z}+k_{3z})\delta(1+2+3)(2\pi)^4 , \quad (3.47)$$

$$\tilde{V}^{(8)}_{\alpha,\beta,\gamma}(1,2,3)=-i\frac{I}{2}(\lambda-1)\sum_j \delta_{\alpha\hat{p}_z}\delta_{\beta\rho}\delta_{\gamma\rho}(i)^3(k_{2j}k_{3z})(k_{2j}+k_{3j})\delta(1+2+3)(2\pi)^4 , \quad (3.48)$$

$$\tilde{V}^{(9)}_{\alpha,\beta,\gamma}(1,2,3)=-iI\rho^0\sum_j \delta_{\alpha\hat{p}_j}\delta_{\beta n_x}\delta_{\gamma\rho}(i)^3(k_{2j}+k_{3j})[(k_{2x}+k_{3x})k_{3z}+(k_{2z}+k_{3z})k_{3x}]\delta(1+2+3)(2\pi)^4 , \quad (3.49)$$

$$\tilde{V}^{(10)}_{\alpha,\beta,\gamma}(1,2,3)=i\lambda I\delta_{\alpha\hat{p}_z}\delta_{\beta\rho}\delta_{\gamma\rho}(i)^3(k_{2z}+k_{3z})k_{2z}k_{3z}\delta(1+2+3)(2\pi)^4 . \quad (3.50)$$

$$\tilde{V}^{(11)}_{\alpha\beta\gamma}(1,2,3)=-i\frac{I}{\gamma_1}\sum_l \delta_{\alpha\hat{n}_l}\delta_{\beta\rho}\delta_{\gamma\rho}(i)^2k_{2l}k_{3z}\delta(1+2+3)(2\pi)^4 , \quad (3.51)$$

where $\delta(1+2+3)\equiv\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3)$. The three-point vertices appearing in (3.42)–(3.50) arise from the reactive momentum stress tensor (2.19) excluding the convective term $(P_iP_j/\rho)$. The vertex (3.51) arises from the dissipative part of the director equation; as we shall see, it ultimately plays no role in the growth of the viscosities.

Using these vertices we have evaluated the relevant self-energies. The results are quite complex and are tabulated in the Appendix. The renormalized transport coefficients have the following form:

$$\Gamma(\mathbf{k},\omega)=\Gamma_0+\frac{\beta}{2}\int_0^\infty dt\, e^{i\omega t}\int\frac{d\mathbf{k}'}{(2\pi)^3}\{G_{\rho\rho}(\mathbf{k}',t)G_{\rho\rho}(\mathbf{k}-\mathbf{k}',t)[\chi^{-2}+f_0(\mathbf{k},\mathbf{k}')]+G_{n_xn_x}(\mathbf{k}',t)G_{n_xn_x}(\mathbf{k}-\mathbf{k}',t)K^2f_1(\mathbf{k},\mathbf{k}')$$
$$+G_{n_yn_y}(\mathbf{k}',t)G_{n_yn_y}(\mathbf{k}-\mathbf{k}'t)K^2f_2(\mathbf{k},\mathbf{k}')+G_{\rho\rho}(\mathbf{k}',t)G_{n_xn_x}(\mathbf{k}-\mathbf{k}',t)I^2f_3(\mathbf{k},\mathbf{k}')\} , \quad (3.52)$$

$$\nu_L(\mathbf{k},\omega)=\nu_L^0+\frac{\beta}{2}\int_0^\infty dt\, e^{i\omega t}\int\frac{d\mathbf{k}'}{(2\pi)^3}\times[G_{\rho\rho}(\mathbf{k}',t)G_{\rho\rho}(\mathbf{k}-\mathbf{k}',t)K^2g_1(\mathbf{k},\mathbf{k}')+G_{n_yn_y}(\mathbf{k}',t)G_{n_yn_y}(\mathbf{k}-\mathbf{k}',t)K^2g_2(\mathbf{k},\mathbf{k}')] , \quad (3.53)$$

$$\nu_T(\mathbf{k},\omega)=\nu_T^0+\frac{\beta}{2}K^2k_x^2\int_0^\infty dt\, e^{i\omega t}\times\int\frac{d\mathbf{k}'}{(2\pi)^3}[h_1(\mathbf{k},\mathbf{k}')G_{n_xn_x}(\mathbf{k}',t)\times G_{n_yn_y}(\mathbf{k}-\mathbf{k}',t)+h_2G_{\rho\rho}(\mathbf{k}',t)G_{\rho\rho}(\mathbf{k}-\mathbf{k}',t)] , \quad (3.54)$$

where $G_{\rho\rho}(\mathbf{k},t)$, $G_{n_xn_x}(\mathbf{k},t)$, and $G_{n_yn_y}(\mathbf{k},t)$ are the correlation functions of the density and director modes, respectively, as functions of $\mathbf{k}$ and $t$. They can be found by inverse Laplace transforming the response functions. The momentum-dependent functions $f_0, f_1, f_2, f_3, g_0, g, g_2, h_1$, and $h_2$ are discussed in the Appendix; their precise form is not necessary here. However, we do note that their angular dependence implies that all of the viscosities $\nu_i$, $i=1,\ldots,5$, are subject to density and director feedback. On the other hand, there is *no* feedback for $c$, $\lambda$, and most importantly $K/\gamma_1$. As we shall see in the next section, this latter result is important in eliminating the possibility of director freezing.

## IV. IMPLICATIONS FOR SUPERCOOLING

We now consider the implications of our results from the previous section for the supercooling of a nematic liquid crystal. Our analysis follows Leutheusser's original approach to the density feedback mechanism, except we are also interested in a potential director feedback mechanism, and the behavior of the six viscosity coefficients.

We begin by recalling Leutheusser's argument for the freezing of the density fluctuations in a simple fluid. The density response function (3.11) can be rewritten as

$$\Phi_1(\mathbf{k},\omega)\equiv\langle\delta\rho(\mathbf{k},\omega)\delta\hat{\rho}(-\mathbf{k},-\omega)\rangle$$
$$=\frac{1}{\omega-\dfrac{\rho k^2c^2}{\omega\rho+ik^2\Gamma}} . \quad (4.1)$$

If the viscosity $\Gamma$ grows as the fluid is supercooled (or compressed), then (4.1) indicates that

$$\Phi_1(k,\omega)\sim\frac{1}{\omega+i\rho c^2/\Gamma}, \quad \Gamma\to\infty . \quad (4.2)$$

This form indicates that the liquid is freezing, in particular as $\Gamma \to \infty$, $\Phi_1$ develops a pole at $\omega = 0$. In deriving (4.2) we assumed that $\Gamma$ was growing very large. Leutheusser showed that this will in fact occur via a feedback mechanism that couples density fluctuations to $\Gamma$, specifically the result (3.52) (dropping the terms proportional to $G_{nn}$ for the moment). The correlation function $G_{\rho\rho}(\mathbf{k},t)$ is given by the inverse Laplace transform of $\Phi_1$ multiplied by $\chi$. Thus, ignoring the $\mathbf{k}$ dependence, (3.52) yields an equation of the form

$$\Gamma(\omega) = \Gamma^0 + \lambda \int_0^\infty dt\, e^{i\omega t} \Phi_1^2(t) , \tag{4.3}$$

where $\Phi_1(t)$ is the inverse Laplace transform of $\Phi_1(\omega)$. Equations (4.2) and (4.4) can be solved to yield a glass transition where $\Phi_1(\omega) \sim 1/\omega$, $G_{\rho\rho}(t) \to$ nonzero constant, as $t \to \infty$ and $\Gamma(\omega=0)$ diverges. In particular $\Gamma(\omega=0)$ diverges as

$$\Gamma(\omega=0) \sim (T - T_G)^{\mu} , \tag{4.4}$$

where $\mu \approx 1.8$ and $T_G$ is the glass-transition temperature.

As discussed in the Introduction, it is now believed that (4.2) is not correct and should be replaced by

$$\Phi_1(\mathbf{k},\omega) \sim \frac{1}{\omega + i\rho c^2/\Gamma + i\gamma} , \tag{4.5}$$

where $\gamma$ does not go to zero as the fluid is supercooled. The origin of $\gamma$ is still a subject of debate [6,7]. Its presence will cut off the Leutheusser transition and (4.4) will not be true asymptotically. Nevertheless, it is believed that there will be a substantial growth in the viscosity as the fluid is supercooled, which is eventually rounded off.

We now proceed with a Leutheusser-style analysis for the nematic phase, bearing in mind the preceding discussion about the limits of such an analysis. We first examine the longitudinal director response function given in (3.12):

$$\Phi_2(\mathbf{k},\omega) \equiv \langle \delta n_x(\mathbf{k},\omega) \delta \hat{n}_x(-\mathbf{k},-\omega) \rangle = \cfrac{1}{\omega + \cfrac{ik^2\omega \dfrac{K\rho_0}{\gamma_1} - k^4 \left[ \nu_L \dfrac{K}{\gamma_1} + \frac{1}{4}(1+\lambda\cos 2\theta)^2 K \right]}{\omega\rho_0 + ik^2\nu_L}} , \tag{4.6}$$

where we have used (3.17). Equation (3.53) indicates that $\nu_L$ does incorporate density feedback [through the product $G_{\rho\rho}(\mathbf{k}',t)G_{\rho\rho}(\mathbf{k}-\mathbf{k}',t)$] and hence it will grow as the nematic is supercooled. Equation (4.6) reduces then in the large $\nu_L$ limit to

$$\Phi_2(\mathbf{k},\omega) \sim \frac{1}{\omega + ik^2 K/\gamma_1}, \quad \nu_L \to \infty . \tag{4.7}$$

In the Appendix we show that $K/\gamma_1$ does *not* renormalize in any dramatic way and is not affected by the density or possible director feedback mechanism. Experimentally, $K/\gamma_1$ has been measured in supercooled nematic phases [20] and found to be consistent with $K/\gamma_1 \sim e^{-1/T}$. Thus, as the temperature is reduced, the director mode will slow down but there will be no sharp Leutheusser transition or even a rounded one. The viscosity $\nu_L$ will grow and appear to diverge due to the density fluctuations, as will $\nu_T$. The absence of a freezing transition for the director modes is true even if the director modes are propagating rather than diffusive. In the limit of large $\nu_L$, (4.7) is still obtained even if $\tilde{\Gamma}_s$ and $\tilde{\Gamma}_f$ in (3.17) become complex (which implies that there are damped, propagating modes). Thus to study the growth of the viscosities in (3.52)–(3.54), all terms proportional to either $G_{n_x n_x}$ or $G_{n_y n_y}$ can be dropped, and feedback from density fluctuations alone occurs.

We now discuss the experimental implications of our results. First we summarize our results in terms of the viscosity coefficients $\nu_1$, $\nu_2$, $\nu_3$, $\nu_4$, $\nu_5$, $\gamma_1$, and $\gamma_2$. The latter two viscosities, as we have already indicated, show no pretransitional growth, at least within the context of our theory. As noted above, the transport coefficient $K/\gamma_1$ has been measured in supercooled nematics and only activated behavior is observed. However, we do expect that the five viscosities $\nu_i$ will show significant growth as the nematic is supercooled. As they are all driven by the same density feedback mechanism, we expect them to show the *same* power-law behavior indicated in (4.4), with eventual rounding off. At this time we cannot predict how wide the temperature regime will be where (4.4) is valid. We also expect on the basis of the results of Refs. [18] and [21], where the effects of local structure on a simple fluid were considered that the shear modes of the nematic phase will become propagating modes at sufficiently high frequency. If the Leutheusser scenario were not ultimately invalidated by a cutoff mechanism, then at the glass transition and below the shear modes would propagate at any frequency and a nonzero shear modulus would be present. Again, at this time we cannot predict what the lower-frequency cutoff will be; calculation of this cutoff will be sensible once the controversy regarding the rounding off of the glass transition is resolved. At that time it will also be sensible to study in detail (3.53)–(3.55) and calculate the $\mathbf{k}$ and $\omega$ dependence of the viscosities. The modes associated with the director relaxation remain diffusive. In that sense there will be an interchange of the "fast" and "slow" modes of Eqs. (3.19)–(3.21), with director relaxation becoming fast and shear relaxation becoming slow.

## ACKNOWLEDGMENTS

E.V. and J.V. were supported in part by the Alexander von Humboldt Foundation (Feodor Lynen Program).

## APPENDIX

In this appendix we provide the technical details of our one-loop calculation of the self-energies appearing in (3.24)–(3.31). We consider only those diagrams where the propagators are either $G_{\rho\rho}(\mathbf{k},\omega)$ or $G_{nn}(\mathbf{k},\omega)$ in order to search for a growth in the viscosities due to density and/or director feedback. We will ignore mixed propagators $G_{\rho n_x}$. As discussed by Forster [16], this correlation function contains both sound poles as well as $\tilde{\Gamma}_s$ and $\tilde{\Gamma}_f$. When expanded in terms of these poles, the strengths of the sound poles can be determined and for large viscosities yield a constant contribution, rather than the $1/\omega$ needed for feedback. The strengths of the director poles cannot be determined to the order in $k$ that we have worked, but is is unlikely that a $1/\omega$ pole would emerge.

The diagrams potentially yielding feedback are all of the bubble variety formed from two three-point vertices. In Eqs. (3.42)–(3.52) we have listed those vertices which contribute to the self-energies we need. While we have chosen the external momentum $\mathbf{k}$ to lie in the $x$-$y$ plane, we note that internal momenta can point in any direction.

We find the following results for the self-energies with two hatted external lines:

$$\Sigma_{\hat{P}_x\hat{P}_x}=\int\frac{d^3k'^3}{(2\pi)^3}\frac{d\omega'}{2\pi}\left[G_{\rho\rho}(\mathbf{k}',\omega')G_{\rho\rho}(\mathbf{k}-\mathbf{k}',\omega-\omega')\left[\tfrac{1}{4}\{Ak_x+B[k_x'(\mathbf{k}\cdot\mathbf{k}')-k_x\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')]\}\right.\right.$$
$$\times\{2Ak_xB[k_x'k'^2+k_x\mathbf{k}'\cdot(\mathbf{k}'-\mathbf{k})]\}$$
$$-I^2(k_x-k_x')(k_z-k_z')^2[(k_x-k_x')(k_z-k_z')^2+k_x'k_z'^2]$$
$$-I^2(\lambda+1)[\tfrac{1}{4}(\lambda+1)k_z^2k_x'(k_z-k_z')+k_z(k_x-k_x')(k_z-k_z')^2]$$
$$\times[k_x'(k_z-k_z')+k_z'(k_x-k_x')]$$
$$-\left\{I[(k_x-k_x')(k_z-k_z')^2+k_x'k_z'^2]\right.$$
$$\left.+\frac{I}{2}(\lambda+1)k_z[k_x'(k_z-k_z')+k_z'(k_x-k_x')]\right\}$$
$$\left.\times\{Ak_x+B[k_x'(\mathbf{k}\cdot\mathbf{k}')-k_x\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')]\}\right]$$
$$+G_{n_xn_x}(\mathbf{k}',\omega')G_{n_xn_x}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$\times\{-K^2k_x'(\mathbf{k}-\mathbf{k}')^2[k_x'(\mathbf{k}-\mathbf{k}')^2+k'^2(k_x-k_x')]-(2\lambda K^2k_x^2+\lambda^2K^2k_x^2)(\mathbf{k}-\mathbf{k}')^2(k'^2+(\mathbf{k}-\mathbf{k}')^2)\}$$
$$\left.+G_{n_yn_y}(\mathbf{k}',\omega)G_{n_yn_y}(\mathbf{k}-\mathbf{k}',\omega-\omega')(-K^2k_x'(\mathbf{k}-\mathbf{k}')^2[k_x'(\mathbf{k}-\mathbf{k}')^2+k'^2(k_x-k_x')])\right], \tag{A1}$$

$$\Sigma_{\hat{P}_y\hat{P}_y}=\int\frac{d^3\mathbf{k}'}{(2\pi)^3}\frac{d\omega'}{2\pi}\left[G_{\rho\rho}(\mathbf{k}',\omega')G_{\rho\rho}(\mathbf{k}-\mathbf{k}',\omega-\omega')\right.$$
$$\times\left[-\frac{B^2}{4}k'^2(k_y')^2(\mathbf{k}\cdot\mathbf{k}')+I^2k_y'(k_z-k_z')^2[k_y'(k_z')^2-k_y'(k_z-k_z')^2]\right.$$
$$-I^2(\lambda+1)[\tfrac{1}{4}(\lambda+1)k_z^2k_y'(k_z-k_z')-k_zk_y'(k_z-k_z')^2][k_y'(k_z-k_z')-k_y'k_z']$$
$$\left.-\left\{I[k_y'k_z'^2-k_y'(k_z-k_z')^2]+\frac{I}{2}(\lambda+1)k_z[k_y'(k_z-k_z')-k_y'k_z']\right\}Bk_y'(\mathbf{k}\cdot\mathbf{k}')\right]$$
$$+G_{n_xn_x}(\mathbf{k}',\omega')G_{n_xn_x}(\mathbf{k}-\mathbf{k}',\omega-\omega)\{-K^2k_y'(\mathbf{k}-\mathbf{k}')^2[k_y'(\mathbf{k}-\mathbf{k}')^2-k_y'k'^2]\}$$
$$\left.+[G_{n_yn_y}(\mathbf{k}',\omega)G_{n_yn_y}(\mathbf{k}-\mathbf{k}',\omega-\omega')]\{K^2k_y'(\mathbf{k}-\mathbf{k}')^2[k_y'(\mathbf{k}-\mathbf{k}')^2-k_y'k'^2]\}\right], \tag{A2}$$

$$\Sigma_{\hat{P}_z\hat{P}_z}=\int\frac{d^3\mathbf{k}'}{(2\pi)^3}\frac{d\omega'}{2\pi}[\,G_{\rho\rho}(\mathbf{k}',\omega')G_{\rho\rho}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$\times(\tfrac{1}{4}\{Ak_z+B[k_z'(\mathbf{k}\cdot\mathbf{k}')-k_z\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')]\}$$
$$\times\{2Ak_z+B[k_z'k'^2+k_zk\mathbf{k}'\cdot(\mathbf{k}'-\mathbf{k})]\}-I^2(k_z-k_z')^3[(k_z-k_z')^3+k_z'^3]$$
$$-I^2(\lambda-1)(k_z-k_z')^3k_x(k_z^2-k_z'^2)-I[(k_z-k_z')^3+k_z'^3]$$
$$\times\{Ak_z+B[k_z'(\mathbf{k}-\mathbf{k}')-k_z\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')]\})$$
$$+[G_{n_xn_x}(\mathbf{k}',\omega')G_{n_xn_x}(\mathbf{k}-\mathbf{k}',\omega-\omega')+G_{n_yn_y}(\mathbf{k}'\omega')G_{n_yn_y}(\mathbf{k}-\mathbf{k}',\omega-\omega')]$$
$$\times[(-K^2)\{k_z'(\mathbf{k}-\mathbf{k}')^2[k_x'(\mathbf{k}-\mathbf{k})^2+k'^2(k_z-k_z')]\}+(k'^2+(\mathbf{k}-\mathbf{k}')^2)$$
$$\times(\mathbf{k}-\mathbf{k}')^2(\lambda^2k_z^2-2\lambda k_zk_z')]\;, \tag{A3}$$

$$\Sigma_{\hat{P}_x\hat{P}_z}=\int\frac{d^3\mathbf{k}'}{(2\pi)^3}\frac{d\omega'}{2\pi}(\,G_{\rho\rho}(\mathbf{k}',\omega')G_{\rho\rho}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$\times\Bigg\{(Ak_x+B[k_x'(\mathbf{k}\cdot\mathbf{k}')-k_x\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')])\{2Ak_z+B[k_z'k'^2+k_z\mathbf{k}'\cdot(\mathbf{k}'-\mathbf{k})]\}$$
$$-\frac{I}{2}\{Ak_x+B[k_x'(\mathbf{k}\cdot\mathbf{k}')-k_x\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')\}[(k_z-k_z')^3+k_z'^3+(\lambda-1)k_xk_z'(k_z-k_z')]$$
$$-I^2(k_x-k_x')(k_z-k_z')^2[(k_z-k_z')^3+k_z'^3]-\frac{I^2}{2}(\lambda+1)k_z(k_z-k_z')^3[k_x'(k_z-k_z')+k_z'(k_x-k_x')]$$
$$+I^2(\lambda-1)k_zk_z'(k_x-k_x')(k_z-k_z')^3-\frac{I^2}{2}(\lambda^2-1)k_zk_zk_x'k_z'(k_z-k_z')-\frac{I}{2}(\lambda+1)k_zk_z'(k_x-k_x')$$
$$\times(Ak_z+B[k_z'(\mathbf{k}\cdot\mathbf{k}')-k_z\mathbf{k}'\cdot(\mathbf{k}-\mathbf{k}')])\Bigg\}$$
$$+G_{n_xn_x}(\mathbf{k}',\omega')G_{n_xn_x}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$\times[(\mathbf{k}-\mathbf{k}')^2K^2\{-\tfrac{1}{2}(\lambda+1)k_x[k_z'(\mathbf{k}-\mathbf{k}')^2+k'^2(k_z-k_z')]-\tfrac{1}{2}(\lambda-1)k_xk_z'[k'^2+(\mathbf{k}-\mathbf{k}')^2]\}$$
$$+\lambda^2K^2k_xk_z(\mathbf{k}-\mathbf{k}')^2[k'^2+(\mathbf{k}-\mathbf{k}')^2]]+[G_{n_xn_x}(\mathbf{k}',\omega')G_{n_xn_x}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$+G_{n_yn_y}(\mathbf{k}',\omega')G_{n_yn_y}(\mathbf{k}-\mathbf{k}',\omega-\omega')$$
$$\times[(-K^2)(\mathbf{k}-\mathbf{k}')^2k_x'[k_z'(\mathbf{k}-\mathbf{k}')^2+k'^2(k_z-k_z')+k_zk'^2+(\mathbf{k}-\mathbf{k}')^2k_z]])\;, \tag{A4}$$

$$\Sigma_{\hat{n}_x\hat{n}_x}=\int\frac{d^3\mathbf{k}'}{(2\pi)^3}\frac{d\omega'}{2\pi}\left[G_{\rho\rho}(\mathbf{k}',\omega')G_{\rho\rho}(\mathbf{k}-\mathbf{k}',\omega-\omega')\left[\frac{I}{\gamma_1}\right]^2k_x'(k_z-k_z')[k_x'(k_z'-k_z)+k_z'(k_x'-k_x)]\right]\;. \tag{A5}$$

The remaining self-energies needed, $\Sigma_{n_x\hat{n}_x}$, $\Sigma_{\hat{n}_yP_y}$, and $\Sigma_{\hat{P}_x\rho}$, do not exhibit feedback. The first of these renormalizes the Frank constants and has no graphs of the form we are considering. However, the finite graph contributing to $\Sigma_{n_x\hat{n}_x}$ does break the one-constant approximation. Similarly, the other two self-energies also do not have any graphical contributions of the form we are considering.

With the self-energies (A1)–(A5), the viscosities $\nu_i$, $i=1,\ldots,5$ can in principle be calculated to one-loop order using (3.32)–(3.36). In general, these expressions are quite complicated, and not much will be learned by displaying them. Even the hydrodynamic limit is difficult to evaluate (except for $\nu_2$ and $\nu_3$) due to the anisotropy of the propagators. Finally, the functions $f_0, f_1, f_2, f_3, g_0, g_1, g_2, h_1$ and $h_2$ appearing in (3.53)–(3.55) can in principle be calculated using (3.37)–(3.39) and (A1)–(A5).

*Present address: Spectral Sciences, Inc., 99 S. Bedford St., Burlington, MA 08103.

[1] For experimental studies of a nematic glass, see, e.g., J. I. Spielberg and E. Gelerinter, Phys. Rev. A **32**, 3647 (1985); H. R. Zeller, Phys. Rev. Lett. **48**, 334 (1982); W. J. Laprice and D. L. Uhrich, J. Chem. Phys. **71**, 1498 (1979).
[2] M. Gabay and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).
[3] For a theoretical study for an anisotropic glassy system see, e.g., L. Golubovic and T. C. Lubensky, Phys. Rev. Lett. **63**, 1082 (1989).
[4] U. Bengtzelius, W. Götze, and A. Sjölander, J. Phys. C **17**, 5915 (1984); W. Götze, Z. Phys. B **56**, 139 (1984), E. Leutheusser, Phys. Rev. A **29**, 2765 (1984). For a review see

W. Götze, in *Liquids, Freezing and Glass Transitions,* edited by J. P. Hansen, D. Levesque, and J. Zinn-Justin (North-Holland, New York, 1991).

[5] S. P. Das, G. F. Mazenko, S. Ramaswamy, and J. Toner, Phys. Rev. Lett. **54**, 118 (1985). For a review see B. Kim and G. F. Mazenko, Adv. Chem. Phys. **78**, 129 (1990).

[6] S. P. Das and G. F. Mazenko, Phys. Rev. A **34**, 2265 (1986).

[7] R. Schmitz, J. W. Dufty, and P. De (unpublished).

[8] See the review articles cited in Refs. [4] and [5].

[9] B. Kim, Phys. Rev. A **46**, 1992 (1992).

[10] See, e.g., Y. Y. Goldschmidt, in *Recent Progress in Random Magnets,* edited by D. H. Ryan (World Scientific, Singapore, 1992).

[11] For an introduction to liquid crystals, see, e.g., P. G. de Gennes, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1974).

[12] See Refs. [11] and [16], as well as M. J. Stephen and J. P. Straley, Rev. Mod. Phys. **46**, 617 (1974); D. Forster, T. C. Lubensky, P. C. Martin, J. Swift, and P. S. Pershan, Phys. Rev. Lett. **26**, 1016 (1971); F. Jähnig and H. Schmidt, Ann. Phys. **71**, 129 (1972).

[13] P. C. Martin, O. Parodi, and P. S. Pershan, Phys. Rev. A **6**, 2401 (1972).

[14] S. Ma and G. F. Mazenko, Phys. Rev. B **11**, 4077 (1975).

[15] We have found that the nonlinearities in $\delta n_i$ do not yield a feedback mechanism, and in the interest of simplicity we will ignore them from the start.

[16] D. Forster, Ann. Phys. **85**, 505 (1974); *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, New York, 1975).

[17] D. Forster, Phys. Rev. Lett. **32**, 1161 (1974).

[18] S. P. Das, Phys. Rev. A **36**, 211 (1987).

[19] P. C. Martin, E. D. Siggia, and H. A. Rose, Phys. Rev. A **8**, 423 (1973).

[20] S. D. Goren, C. Korn, and S. B. Marks, Phys. Rev. Lett. **34**, 1212 (1975); J. W. Doane, C. E. Tarr, and M. A. Nickerson, *ibid.* **33**, 620 (1974); S. Meiboom and R. C. Hewitt, *ibid.* **30**, 261 (1973).

[21] S. P. Das, Phys. Rev. A **42**, 6116 (1990).