Dynamics of nematic elastomers

Olaf Stenull and T. C. Lubensky
Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA
(Dated: March 22, 2022)

We study the low-frequency, long-wavelength dynamics of soft and semi-soft nematic elastomers using two different but related dynamic theories. Our first formulation describes the purely hydrodynamic behavior of nematic elastomers in which the nematic director has relaxed to its equilibrium value in the presence of strain. We find that the sound-mode structure for soft elastomers is identical to that of columnar liquid crystals. Our second formulation generalizes the derivation of the equations of nematic hydrodynamics by the Harvard group to nematic elastomers. It treats the director explicitly and describes slow modes beyond the hydrodynamic limit.

PACS numbers: 83.80.Va, 61.41.+e, 62.30.+d

I. INTRODUCTION

Nematic elastomers (NEs) \[1,2,3\] are rubbery materials with the macroscopic symmetry properties of nematic liquid crystals \[5,6\]. In addition to the elastic degrees of freedom of ordinary rubber, nematic elastomers possess the internal, orientational degree of freedom of liquid crystals. Since NEs are amorphous solids rather than fluids, their mechanical properties differ significantly from those of standard nematic liquid crystals. The interplay between elastic and orientational degrees of freedom is responsible for several fascinating properties of NEs. For example, temperature change or illumination can change the orientational order and cause the elastomer to extend or contract by several hundred percent \[7,8,9\]. Nematic elastomers display a soft elasticity \[10,11,12,13\] characterized by vanishing shear stresses for a range of longitudinal strains applied perpendicular to the nematic direction. They exhibit an anomalous elasticity in which certain bending and shear moduli are length-scale dependent and vanish or diverge at long length-scales \[14,15,16\].

In addition to unusual static properties, NEs have an intriguing dynamic mechanical behavior. Early rheology experiments on liquid crystalline elastomers \[17\] found no nematic effects in NEs. Later, however, several experiments \[18,19,20,21\] observed a genuinely unconventional response to oscillatory shear. It turns out that an internal relaxation of the nematic director leads to a dynamic mechanical softening of NEs. This behavior has been christened dynamic soft elasticity. It makes NEs interesting for device applications in areas such as mechanical vibration damping \[14\] (exploiting the fact that the mechanical loss is record-high over a wide range of temperatures and frequencies) or acoustics where NEs open the possibility of acoustic polarization \[22\] (using the fact that only particular soft shears are strongly attenuated) analogous to the optical polarization in birefringent media.

The theoretical investigation of the dynamic-mechanical properties of NEs was pioneered by Terentjev, Warner and coworkers (TW) \[1,18,22,23,24\]. References \[1\] and \[22\] present a detailed derivation of equations governing the long-wavelength low-frequency dynamics of NEs and derive their associated mode structure, which includes non-hydrodynamic, rapidly decaying modes. This derivation, which assumes a single relaxation time for the director, is based on the Lagrangian approach \[25\] to the dynamics of continuum systems in which non-dissipative forces are calculated as derivatives of an energy functional and dissipative forces are derivatives of a a Rayleigh dissipation function. The TW work focuses primarily on rheological response in both soft and semi-soft NEs at zero wavenumber, and it neglects contributions to dynamical equations arising from the Frank free energy for director distortions, which are higher order in wavenumber than those arising from network elasticity. As a result, as we shall see, the mode structure derived in Refs. \[1\] and \[22\] misses diffusive modes along symmetry directions in soft NEs. When contributions from the Frank free energy are included and those from the elastic network are excluded, the TW approach reproduces the original Leslie-Ericksen equations \[26,27\] of nematic hydrodynamics, excluding the rotational inertial term that is usually discarded \[28\] or is missing entirely in alternative derivations \[2\] of these equations.

In this paper, we will present alternative approaches to deriving the equations governing the dynamics of NEs. First we will derive the exclusively hydrodynamical equations for those variables whose characteristic frequencies vanish with wavenumber. These equations, like those for the hydrodynamics of smectic \[5,29\] and columnar \[5\] liquid crystals, exclude non-hydrodynamic director modes. Interestingly, the modes for soft NEs predicted by these equations are identical in form to those of columnar liquid crystals \[5\] with three pairs of sound modes and diffusive modes along symmetry directions where sound velocities vanish. One pair of sound modes is predominantly longitudinal with non-vanishing velocity at all angles. A second pair is predominantly transverse with a velocity that vanishes for wavevector either parallel or perpendicular to the uniaxial symmetry axis, and a third pair is completely transverse with a velocity that vanishes for wavevector parallel to the symmetry axis but is nonzero otherwise. We then derive the phenomenological equa-
ations for the slow dynamics of all displacement and director variables in NEs using the Poisson-Bracket formalism [30, 31] for obtaining the dynamics of coarse-grained variables, applied so successfully to the study of dynamical critical phenomena [32], for deriving phenomenological equations for any set of coarse-grained variables whose dynamics is slow on a time scale set by microscopic collision times. Like TW, we assume a single relaxation time for the director. We do, however, discuss how this constraint can be relaxed. When applied to fluid nematic liquid crystals, the Poisson-bracket formalism is equivalent to that used by the “Harvard group” [33, 34] in its derivation of hydrodynamics of nematics. Our dynamical equations for NEs reduce to the equations of nematohydrodynamics derived by the Harvard group when elastic rigidities vanish and to the purely hydrodynamical equations for NEs when fast modes are removed. When contributions from the Frank free energy are ignored, our equations are identical to those derived by TW. We will not discuss random stresses or inhomogeneities in this paper, though they contribute static components in light scattering experiments [35] that may obscure the observation of the NE modes we discuss here.

The outline of our paper is as follows. In Sec. III we briefly review the general Poisson-bracket formalism for obtaining coarse-grained dynamics. Section III focuses on the pure hydrodynamics of NEs. We set up equations of motion for the momentum density and the elastic displacement. We derive the appropriate elastic energy entering these equations by integrating out the director degrees of freedom. Then we compare the hydrodynamic equations to those of conventional uniaxial solids and columnar liquid crystals. After extracting the sound velocities of the modes we finally determine the full mode structure in the incompressible limit. Section IV features our formulation that explicitly accounts for the dynamics of the director. We set up equations of motion for the momentum density, the elastic displacement and the director. These are then compared to the equations of motion for uniaxial solids and to the equations of motion derived by TW. We determine the mode structure. Finally, we compare the so obtained modes to the modes obtained by TW. A brief summary is given in Sec. V.

II. COARSE-GRAINED DYNAMICS

Stochastic dynamical equations for coarse-grained fields [31, 32] can be obtained by combining the Poisson-Bracket formalisms of Classical mechanics [25], which guarantees the correct reactive couplings between fields with opposite signs under time reversal, and the Langevin [30] approach to stochastic dynamics, which provides a description of dissipative processes and noise forces. Let \( \Phi_{\mu}(x, t), \mu = 1, 2, \ldots \) be a set of coarse-grained fields whose statistical mechanics is described by a coarse-grained Hamiltonian \( \mathcal{H} \). The dynamical equations for \( \Phi_{\mu}(x, t) \) are first-order differential equations in time:

\[
\dot{\Phi}_{\mu}(x, t) = -\int d^d x' \int dt' \left\{ \Phi_{\mu}(x, t), \Phi_{\nu}(x', t') \right\} \frac{\delta \mathcal{H}}{\delta \Phi_{\nu}(x', t')} - \Gamma_{\mu,\nu} \frac{\delta \mathcal{H}}{\delta \Phi_{\nu}(x, t)} + \zeta_{\mu}(x, t). \tag{2.1}
\]

Here and in the following the Einstein summation convention is understood. The first term on the right hand side is a non-dissipative velocity, also known as the reactive term, that contains the Poisson bracket \( \left\{ \Phi_{\mu}(x, t), \Phi_{\nu}(x', t') \right\} \) of the coarse grained fields \( \Phi_{\mu} \). The reactive term couples \( \Phi_{\mu} \) to \( \delta \mathcal{H}/\delta \Phi_{\nu} \) only if \( \Phi_{\mu} \) and \( \Phi_{\nu} \) have opposite signs under time reversal (when external magnetic fields are zero). The second term on the right hand side is a dissipative term. \( \Gamma_{\mu,\nu} \) are the components of the so-called dissipative tensor. This tensor couples \( \Phi_{\mu} \) to \( \delta \mathcal{H}/\delta \Phi_{\nu} \) only if \( \Phi_{\mu} \) and \( \Phi_{\nu} \) have the same sign under time reversal. If the noise term \( \zeta_{\mu} \) is present, Eq. (2.1) represents a stochastic or Langevin equation. As such Eq. (2.1) may be used to set up a dynamic functional [38, 39, 40] to study the effects of nonlinearities and fluctuations via dynamical field theory. In this paper we are not interested in these effects. Hence, we focus on linearized hydrodynamic equations and pay no further attention to noise.

III. PURE HYDRODYNAMICS

Hydrodynamics describes the dynamics of those degrees of freedom whose characteristic frequencies \( \omega \) vanish as wave number tends to zero. In other words hydrodynamics focuses exclusively on the leading low-frequency, long-wavelength behavior. All other degrees of freedom, even though they might be slow, are strictly speaking not hydrodynamic ones. In this section we will derive the hydrodynamic equations of NEs. These equations apply for frequencies \( \omega \) such that \( \omega \tau \ll 1 \), where \( \tau \) is the longest non-hydrodynamic decay time in the system. As we will show in the next section, the characteristic time for director decay is in fact very slow with \( \tau \sim 10^{-2} \) sec. [41, 42] so that the regime of applicability of hydrodynamics is quite small for current NEs. It is imaginable, however, that other systems will be found with shorter decay times.

There are two general classes of hydrodynamic variables: conserved variables and broken symmetry variables. A single-component NE has the same set of conserved variables as a fluid: energy density \( \epsilon \), mass density \( \rho \), and momentum density \( g \). It also has the same broken symmetry variables as a crystalline solid, namely three displacement variables, though strictly speaking, these variables in an elastomer are not associated with a macroscopic broken symmetry because in the absence of orientational order elastomers have the same macroscopic rotational and translational symmetry as a fluid.
Since it is rotational symmetry that distinguishes a nematic elastomer from an isotropic one, it could be argued that the nematic director should be a hydrodynamic variable, but as in smectic and columnar liquid crystals, the director degrees of freedom decay in microscopic times to their preferred configuration in the presence of strain and are thus not hydrodynamic variables. Elastomers differ from equilibrium crystals in at least two important ways: The first, alluded to above, is that they are not periodic and thus do not have mass-density wave order parameters whose phases act as broken symmetry hydrodynamic variables; rather Lagrangian displacement variables u take their place. The second is that an elastomer is permanently crosslinked: it is a classical rubber in volume such that \( \delta \rho/\rho = -\nabla \cdot \mathbf{u} \) and in which permutation in which there is translation of a mass-density-wave without mass motion is prohibited. Thus, mass density is not an independent hydrodynamic variable, and we are left with a total of 5 + 3 − 3 = 5 independent variables and 7 associated hydrodynamic modes. These modes are heat diffusion and, depending on direction, either six propagating sound modes, four propagating sound modes and two diffusive displacement/velocity modes, or two longitudinal sound modes and diffusive displacement/velocity modes. In what follows, we will consider only isothermal processes so that heat heat diffusion can be ignored.

A. Elastic energy

To derive the hydrodynamical equations for NEs, we first need the appropriate elastic free energy. The elastic constant measuring the energy of strains in planes containing the anisotropy axis (the shear modulus \( C_\perp \)) to be defined in Eq. (3.2) vanishes as a result of the broken symmetry brought about by the establishment of nematic order. Thus a good starting point for this free energy is that of a uniaxial solid with this elastic constant simply set to zero. This leaves certain soft directions in which distortions cost zero energy, and as in smectic and columnar liquid crystals, curvature-like terms that are quadratic in second-order spatial derivatives have to be added to ensure stability. To make contact with dynamical equations involving the director to be presented in Sec. IV, rather than simply adding these terms, we find it useful to derive them from the free energy of a nematic elastomer expressed in terms of both the strain and the directors. We will restrict our attention to harmonic distortions.

Elastomers are permanently crosslinked pieces of rubber whose static elasticity is most easily described in Lagrangian coordinates in which \( x \) labels a mass point in the unstretched (reference) material and \( \mathbf{R}(x) = x + \mathbf{u}(x) \), where \( \mathbf{u}(x) \) is the displacement variable, labels the position of the mass point \( x \) in the stretched (target) material. We will use Lagrangian coordinates throughout this paper. We will, however on occasion make reference to Eulerian coordinates in which \( \mathbf{r} \equiv \mathbf{R}(x) \) specifies a position in space and \( \mathbf{u}(\mathbf{r}) \) the displacement variable at that position.

The elastic energy of a nematic elastomer can be divided into three parts:

\[
\mathcal{H} = \mathcal{H}_u + \mathcal{H}_n + \mathcal{H}_{u.n},
\]

(3.1)

where \( \mathcal{H}_u \) is the usual elastic energy of a uniaxial solid, \( \mathcal{H}_n \) is the Frank free energy of a nematic, and \( \mathcal{H}_{u.n} \) is the energy of coupling between strain and director distortions.

Choosing the coordinate system so that the z-direction coincides with the uniaxial direction, we have to harmonic order 4:

\[
\mathcal{H}_u = \int d^3x \left\{ \frac{C_1}{2} u_{zz}^2 + C_2 u_{zz} u_{ii} + \frac{C_3}{2} u_{ii}^2 + C_4 u_{ab}^2 + C_5 u_{zz}^2 \right\}. \tag{3.2}
\]

(3.2)

Here, \( u_{ij} \) are linearized components of the Lagrange strain tensor \( \mathbf{u} \):

\[
u_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i). \tag{3.3}
\]

(3.3)

We will use the convention that indices from the beginning of the alphabet, \( \{a,b\} \), assume the values 1 and 2 whereas indices from the middle of the alphabet, \( \{i,j,k,l\} \), run from 1 to 3. Note that, compared to our work on the anomalous elasticity of NEs \( \mathcal{H}_u \), the elastic constants in Eq. (3.2) have a somewhat different definition that is geared towards taking the incompressible limit. Here we have arranged things so that the terms featuring \( C_2 \) and \( C_3 \) involve the trace of the strain tensor. In the incompressible limit that we will eventually take one has \( u_{ii} = 0 \) so that \( C_2 \) and \( C_3 \) drop out.

Expanded to harmonic order in the deviation \( \delta \mathbf{n} = \mathbf{n} - \mathbf{n}_0 \) from the uniform equilibrium state \( \mathbf{n}_0 = \hat{e}_z \), the Frank energy reads

\[
\mathcal{H}_{\delta n} = \int d^3x \left\{ \frac{K_1}{2} (\partial_a \delta n_a)^2 + \frac{K_2}{2} (\varepsilon_{ab} \partial_a \delta n_b)^2 + \frac{K_3}{2} (\partial_\alpha \delta n_\alpha)^2 \right\}, \tag{3.4}
\]

(3.4)

where \( \varepsilon_{ab} = -\varepsilon_{ba} \) is the two-dimensional Levi-Civita symbol. For notational simplicity in the remainder of this paper we will replace \( \delta \mathbf{n} \) with \( \mathbf{n} \) with the understanding that it has only two components \( n_a \). With this notation, \( \mathcal{H}_{\delta n} \) can be expressed as

\[
\mathcal{H}_{\delta n} = -\frac{1}{2} \int d^3x n_a M_{ab}(\nabla) n_b, \tag{3.5}
\]

(3.5)

where

\[
M_{ab} = (K_1 - K_2) \partial_a \partial_b + (K_2 \partial_a^2 + K_3 \partial_b^2) \delta_{ab}. \tag{3.6}
\]

(3.6)
and where $\partial^2_{\perp \perp} = \partial_a \partial_a$ with the Einstein convention understood.

The coupling energy, finally, can be written in the form

$$H_{u,n} = \int d^3x \left\{ \frac{D_1}{2} Q^2_a + D_2 u_{az} Q_a \right\},$$

(3.7)

where once more terms beyond harmonic order have been neglected and where

$$Q_a = \delta n_a - \frac{1}{2} (\partial_z u_a - \partial_a u_z) \equiv \delta n_a - \hat{Q}_a.$$  (3.8)

As explained above the director is not a genuine hydrodynamic variable and hence it should be integrated out of the elastic energy as long as we focus on the hydrodynamic limit. We do so by minimizing $H$ over $n_a$ to find

$$n_a = \hat{Q}_a - \frac{D_2}{D_1} u_{az} + \frac{M_{ab}(\nabla)}{D_1} \left( \hat{Q}_b - \frac{D_2}{D_1} u_{bz} \right).$$  (3.9)

Inserting this equation into $H$ and retaining only the dominant terms in gradients, we obtain

$$H_{u-cl} = \int d^3x \left\{ C_1 u_{zz}^2 + C_2 u_{zz} u_{ii} + C_3 u_{ii}^2 \right\} + C_4 u_{ab}^2 + C_5 R u_{az}^2 + \frac{K_1}{2} (\partial^2_{\perp \perp} u_z)^2 + \frac{K_3}{2} (\partial^2_{\perp \perp} u_a)^2 \right\}.$$  (3.10)

where

$$C_5^R = C_5 - \frac{D_2^2}{2D_1},$$  (3.11a)

$$K_1^R = \frac{1}{4} \left( 1 + \frac{D_2}{D_1} \right)^2 K_1,$$  (3.11b)

$$K_3^R = \frac{1}{4} \left( 1 - \frac{D_2}{D_1} \right)^2 K_3.$$  (3.11c)

The superscript $R$ indicates that $C_5, K_1$ and $K_3$ have been renormalized by director fluctuations. Note that the elastic constants $K_1^R$ and $K_3^R$ are not the same as the Frank splay and bend constants $K_1$ and $K_3$. This is in contrast to smectic-A and columnar liquid crystals in which the coefficients of quartic gradient terms in the effective elastic energy arising from the relaxation of director modes are identical to the Frank elastic constants.

The modulus $D_2$ can have either sign, and there are no thermodynamic constraints preventing either $1 + D_2/D_1$ or $1 - D_2/D_1$ from being zero. Thus, either $K_1^R$ or $K_3^R$ in Eq. (3.11) could be zero. In this case, contributions to $(\partial^2_{\perp \perp} u_z)^2$ or $(\partial^2_{\perp \perp} u_a)^2$ in $H_{u-cl}$ arising from network elasticity would have to be added to ensure stability. Except for the exceptional cases when $D_2 = \pm D_1$, those contributions are generally smaller than the ones represented in Eq. (3.11), and we will ignore them. If $C_5^R$ vanishes, then the dependence of the elastic energy on $u_{az}$ drops out. This is the famous soft elasticity. If the condition for softness (3.11) is not strictly fulfilled and there is a small but finite remnant $C_5^R$ then small shears in the planes containing the anisotropy axis do cost a small energy. This non-ideal behavior is known as semi-softness.

The energy (3.10) combines attributes of both smectic and columnar liquid crystals. The vertical displacement $u_z$ is analogous to the displacement variable $u$ of a smectic liquid crystal. It is soft for distortions in the $\perp$-direction, and a bending term $K^R_3 (\partial^2_{\perp \perp} u_a)^2/2$ is needed to stabilize it. The in-plane displacements $u_{az}$ are analogous to those of column liquid crystal, which are soft for distortions in the $z$-direction, and a bending term $K^R_3 (\partial^2_{\perp \perp} u_a)^2/2$ is need to stabilize them.

### B. Hydrodynamic equations

We are now in a position to write down the full hydrodynamic equations for nematic elastomers. For simplicity, we will restrict our attention to isothermal processes so that we can ignore temperature diffusion. This leaves us with six hydrodynamical variables, the momentum density $g_i(x)$ and the displacement $u_i(x)$. These are independent variables at the reference point $x$ that satisfy the continuum generalizations of the usual relations for the momentum and displacement of a particle: $\partial g_i(x)/\partial x^j = \delta_{ij} (\delta_{\perp \perp} \delta_{\perp \perp} - 1/2)$, $\partial u_i(x)/\partial x^j = \delta_{ij}$, and $\partial g_j(x)/\partial u_i(x') = 0$. These relations yield a non-vanishing Poisson bracket between $u_i(x)$ and $g_j(x')$

$$\{u_i(x), g_j(x')\} = \delta_{ij} (\delta_{\perp \perp} \delta_{\perp \perp} - 1/2).$$

(3.12)

The $g - g$ and $u - u$ Poisson brackets are zero. Using these results, we obtain the equations of motion

$$v_i \equiv \dot{u}_i = \frac{\delta H_{kin}}{\delta g_i} = \frac{1}{\rho} g_i,$$  (3.13a)

$$\dot{g}_i = - \frac{\delta H_{u-cl}}{\delta u_i} + \eta_{ijkl} \partial_{ij} \partial_{kl} v_k,$$  (3.13b)

where $\mathbf{v}$ is the velocity field, and $H_{kin} = \int d^3x g_i g_i/(2\rho)$ is the coarse-grained kinetic energy. $\eta_{ijkl}$ is the viscosity tensor, which has five independent components. We can parametrize the stress tensor so that the entropy production from viscous stresses takes on the same form as the elastic energy $H_u$,

$$T \dot{S} = \int d^3x \left\{ \frac{\eta_{1}}{2} \dot{u}_{zz}^2 + \eta_2 \dot{u}_{zz} \dot{u}_{ii} + \frac{\eta_3}{2} \dot{u}_{ii}^2 \right\} + \eta_4 \dot{u}_{ab}^2 + \eta_5 \dot{u}_{az}^2 \right\},$$

(3.14)

where the $\eta$'s are low frequency viscosities. $\eta^R$ is an effective viscosity that is, as its counterpart $C_5^R$, renormalized by director fluctuations. Equation (3.14) contains all contributions to the entropy production equation in the truly hydrodynamic limit we are considering. Nonhydrodynamic variables like the director have already relaxed.
to their local equilibrium values. Thermodynamic stability requires $\eta_i \geq 0$ for $i = 1, 3, 4, \eta_5^R \geq 0$ and $\eta_1 \eta_3 \geq \eta_2^2$.

A few observations about Eqs. (3.13) are in order. First they are identical in form to the equations for a conventional elastic material (without vacancy diffusion). The distinction between such a material and a nematic elastomer appears only in the form of $\mathcal{H}_{\text{ne}}$. The absence of any dissipative term proportional to $-\delta \mathcal{H}/\delta u_i$ in Eq. (3.13a) reflects the tethered or crosslinked character of the elastomer. In non-crosslinked systems, this equation would contain an additional dissipative term proportional to $-\delta \mathcal{H}_{\text{ne}}/\delta u_i$ describing permeation. Second, these equations are expressed in Lagrangian coordinates, and the derivatives $\mathbf{u}$ and $\mathbf{g}$ are time derivatives at constant value of the reference position $\mathbf{x}$. In Eulerian coordinates, $\mathbf{u}$ and $\mathbf{g}$ become functions of points $\mathbf{r} = \mathbf{R}(\mathbf{x})$ in space: $\mathbf{u}_E(r, t) = \mathbf{u}(x(r), t)$ and similarly for $\mathbf{g}_E$. The time derivative $\dot{\mathbf{u}} = d\mathbf{u}/dt = d\mathbf{u}_E/dt + \mathbf{v} \cdot \nabla \mathbf{u}_E$ is the time derivative in the reference frame moving with the local fluid velocity.

To obtain a more explicit form for the equations of motion, we note that Eq. (3.13b) allows us to replace $\dot{g}$ in Eq. (3.15), with $\rho \ddot{u}_i$ to produce the standard mechanical equation for a solid with dissipation,

$$\rho \ddot{u}_a = (C_2 + \eta_2 \partial_t) \partial_a u_{zz} + (C_3 + \eta_3 \partial_t) \partial_a u_{zi} + 2(C_4 + \eta_4 \partial_t) \partial_a u_{ab} + (C_5^R + \eta_5^R \partial_t) \partial_a u_{az} - K^R_3 \partial^2 a_{uu} ,$$

$$\rho \ddot{u}_z = (C_1 + \eta_1 \partial_t + C_2 + \eta_2 \partial_t) \partial_z u_{zz} + (C_2 + \eta_2 \partial_t + C_3 + \eta_3 \partial_t) \partial_z u_{zi} + (C_5^R + \eta_5^R \partial_t) \partial_z u_{bz} - K^R_1 \partial^2 a_{zz} .$$

Apart from the appearance of $C_5^R$, which is zero in a soft NE, rather than $C_5$ and bending terms with quartic derivatives, these equations are identical to those of a uniaxial solid.

Upon switching to frequency space one can see that the static renormalization of the shear modulus $C_5$, Eq. (3.11), has a dynamical generalization with a frequency dependent $C_5^R(\omega) = C_5^R - i \omega \eta_5^R$, whose real part vanishes when $C_5^R = 0$. This form for $C_5^R(\omega)$ is only valid in the hydrodynamic limit with $\omega \tau \ll 1$. As we will discuss more fully in the next section, there are other terms in $C_5^R(\omega)$ when this limit is not obeyed.

C. Sound velocities

To assess the mode structure of the equations of motion (3.15), we begin with an analysis of propagating sound modes in the absence of dissipation. To keep arguments simple, we will restrict ourselves here to ideally soft NEs with $C_5^R = 0$. When $C_5^R \neq 0$, the bending terms can be neglected, and the dynamical equations and associated equations are identical to those of uniaxial solids. We will return to the case when $C_5^R$ is nonzero in Sec. IV.

The sound modes have frequencies $\omega(q) = c(\theta) q$ where $\theta$ is the angle that $\mathbf{q}$ makes with the $z$-axis. The non-dissipative bending terms give rise to modes with $\omega \sim q^2$ along the symmetry direction. These modes, however, mix with dissipative ones and become overdamped diffusive modes with $\omega \sim -i q^2$. Thus, to obtain the true non-dissipative sound-mode structure, we can ignore the bending terms. Consequentially, the non-dissipative sound-mode structure is that of a uniaxial solid with $C_5 = 0$, which it turns out is identical to that of columnar liquid crystals, though the input variables are slightly different (there is no $u_z$ in a columnar liquid crystal, but there is an independent density). The modes break up into displacements $w_i = \varepsilon_{ab} q_a u_a / q_z$ perpendicular to both $\mathbf{q}$ and the $z$-axis and coupled displacements $u_z = (\varepsilon_{ab} q_a u_a) / q_z$ and $u_z$ in the plane that contains $\mathbf{q}$ (see Fig. 1). In Fourier space, the hydrodynamic equations are

$$\rho \omega^2 u_{zt} = C_4 q_z^2 u_{zt} ,$$

$$\rho \omega^2 u_{zz} = (C_1 + 2C_2 + C_3) q_z^2 u_{zz} + (C_3 + 2C_4) q_z^2 u_{zz} .$$

Thus, there is a transverse sound mode with velocity

$$c_t(\theta) = \sqrt{\frac{C_4}{\rho} \sin \theta}$$

that vanishes when $\theta = 0$ and reaches a maximum at $\theta = \pi/2$. The sound velocities $c_1(\theta)$ and $c_2(\theta)$ of the
other modes are coupled and satisfy

\[
c_1^2 c_2^2 = \frac{1}{\rho^2} [(2C_4 + C_3)(C_1 + 2C_2 + C_3) \\
- (C_2 + C_3)^2 \cos^2 \theta \sin^2 \theta]
\] (3.17b)

\[
c_1^2 + c_2^2 = \frac{1}{\rho} [(2C_4 + C_3) \sin^2 \theta + (C_1 + 2C_2 + C_3) \cos^2 \theta]
\] (3.17c)

These equations are identical to the equations satisfied by the sound velocities in a columnar system [5]. One of the sound modes is purely longitudinal in the limit \(C_3 \to \infty\); its velocity is non-vanishing for all \(\theta\). The second mode is like that of a smectic-\(A\) [29] with a sound velocity that vanishes at \(\theta = 0\) and \(\theta = \pi/2\). The third mode is a purely transverse sound mode whose velocity vanishes only at \(\theta = 0\). Figure 2 plots the three sound velocities.

**D. Full incompressible mode structure**

Having found the general sound mode structure in the non-dissipative limit, we turn to a full analysis of modes in the incompressible limit. To discuss this limit, it is useful to decompose \(u(q)\) into a longitudinal part \(u_l\) along \(q\) and components \(u_t\), perpendicular to both \(q\) and \(z\), and \(u_T\) perpendicular to \(q\) in the plane containing \(q\) and \(z\) as shown in Fig. 1

As was the case in the dissipationless limit, \(u_t\) decouples from \(u_T\) and \(u_l\). In the incompressible limit \(u_l\) vanishes and we are left with

\[
\rho \omega^2 u_t = (C_4 q_1^2 + K_3^R q_z^2) u_t - i \omega (\eta_4 q_1^2 + \eta_5^R q_z^2) u_t,
\] (3.18a)

\[
\rho \omega^2 u_T = \left[ (C_1 + 2C_4) \frac{q_1^2 q_2^2}{q_2^2} + K_1 q_z^2, \frac{q_1^6}{q_2^2} + K_3^R q_z^2 \right] u_T \\
- i \omega \left[ (\eta_1 + 2\eta_4) q_1^2 q_2^2 + \eta_5^R (q_1^2 - q_z^2)^2 \right] u_T.
\] (3.18b)

These equations produce propagating modes with respective frequencies

\[
\omega_{t,\pm} = \pm \sqrt{\frac{C_4}{\rho} |q_1| - i \frac{2\eta_4 q_1^2 + \eta_5^R q_z^2}{4\rho}},
\] (3.19a)

\[
\omega_{T,\pm} = \pm \sqrt{\frac{C_1 + 2C_4}{\rho} |q_1 q_z|} q \\
- i \frac{2(\eta_1 + 2\eta_4) q_1^2 q_2^2 + \eta_5^R (q_1^2 - q_z^2)^2}{4\rho q_2^2},
\] (3.19b)

as long as \(q\) is not along a symmetry direction in which the sound velocity is zero. When \(q_\perp = 0\), the \(t\)- and \(T\)-modes become diffusive with identical frequencies

\[
\omega_{t,\pm} = \omega_{T,\pm} = \frac{1}{4\rho} \left[ -i \eta_5^R \pm \sqrt{-(\eta_5^R)^2 + 16\eta_1 K_3^R} \right] q_1^2.
\] (3.20)

As in conventional nematics \(K_1 \sim K_3 \sim 10^{-6}\) dynes and \(\rho \sim 1\) gram/cm\(^3\). The viscosity should be larger than the 0.01 poise characteristic of fluids. Thus, we can expect that \(16\rho K_3^R \ll (\eta_5^R)^2\). In this limit the above modes become slow and a fast diffusive mode with frequencies

\[
\omega_{t,s} = \omega_{T,s} = -i \frac{2K_3^R}{\eta_5^R} q_z^2,
\] (3.21a)

\[
\omega_{t,f} = \omega_{T,f} = -i \frac{\eta_5^R}{2\rho} q_z^2.
\] (3.21b)

When \(q_z = 0\), the \(t\)-modes remain propagating sound modes, but the \(T\)-modes become diffusive with frequencies

\[
\omega_{T,\pm} = \frac{1}{4\rho} \left[ -i \eta_5^R \pm \sqrt{-(\eta_5^R)^2 + 16\rho K_1^R} \right] q_1^2,
\] (3.22)

which in the limit \(16K_1^R \ll (\eta_5^R)^2\) reduce to

\[
\omega_{T,s} = -i \frac{2K_1^R}{\eta_5^R} q_z^2,
\] (3.23a)

\[
\omega_{T,f} = -i \frac{\eta_5^R}{2\rho} q_z^2.
\] (3.23b)

Note that there is one diffusive and one propagating sound mode in the incompressible limit when \(q_z = 0\). The attenuation (the imaginary part of \(q\)) of the diffusive modes is proportional to \(\sqrt{\omega}\) and that of the sound
modes is proportional to $\omega$. This is the explanation of the large difference in attenuation of the two polarizations of transverse wave found by TW that makes NEs candidates for acoustic polarizers. We will return to this issue in Sec. IV C.

Equations (3.21) and (3.22) show that one misses the slow diffusive modes if one neglects the Frank energy. It is a legitimate question, however, under what conditions these modes can be observed experimentally. Manifestly, they should be observable directly in the symmetry directions in which the respective sound velocities vanish whereas they should not be seen in directions which differ significantly from these symmetry directions. Between the two extremes there will be a crossover from slow diffusive to propagating behavior at certain crossover angles. These angles can be estimated by comparing the magnitude of the terms in Eqs. (3.19) and (3.21) or respectively (3.25). For $\theta \approx 0$ we get for example from Eqs. (3.19) and (3.21) that the crossover is expected at an angle $\theta_0$ such that

$$|\sin \theta_0| \approx \frac{\sqrt{\frac{\rho}{C_4}} K_\text{F}^R}{\eta_0} q.$$  

(3.24)

The wave vectors in light scattering experiments on NE dynamics typically have a magnitude $q \sim 10^5$ cm. The elastic moduli $C_1$ and $C_4$ should be comparable to the shear modulus of rubber, $C_1 \sim C_4 \sim 10^7$ dynes/cm$^2$. Hence, we estimate

$$|\sin \theta_0| \approx 10^{-4},$$  

(3.25)

i.e., the range around the nematic direction in which the slow diffusive behavior is observable is extremely narrow. Applying the same reasoning to the slow diffusive $T$-mode for $\theta \approx \pi/2$ we obtain a further crossover angle $\theta_{\pi/2}$ with

$$|\cos \theta_{\pi/2}| \approx 10^{-4},$$  

(3.26)

signaling the same extremely narrow angle range for slow diffusive behavior as above.

IV. DYNAMICS WITH DISPLACEMENTS AND DIRECTOR

Nematic elastomers, like their conventional nematic-liquid-crystal counterparts, are characterized by a Frank director $\mathbf{n}$ that responds dynamically to external forces. In this section, we will use the Poisson-bracket approach to derive phenomenological equations for the dynamics of both the director and displacements in nematic elastomers. Though our derivation is different from that of TW, our dynamical equations are in fact identical to theirs if they include contributions from the Frank free energy for the director. Our equations also reduce to the standard equations of nematohydrodynamics when elasticity due to network crosslinking is turned off.

It must be emphasized, however, that our equations predict non-hydrodynamic modes characterized by a decay time $\tau$ that does not approach infinity with vanishing wavenumber $q$. In any real system, there are many non-hydrodynamic modes with characteristic decay times $\tau_0$. For a dynamical theory to provide a correct description of a system over a frequency range from zero to some maximum frequency $\omega_M$, it must include contributions from all modes, both hydrodynamic and non-hydrodynamic, with characteristic frequencies up to a few times $\omega_M$. An isotropic rubber is characterized by rather long decay times $\tau_R$ of Rouse-like modes of chain segments. At frequencies $\omega$ such that $\omega \tau_R \ll 1$, its viscosities become frequency independent described by the hydrodynamical equations of an isotropic solid. However, when $\omega \tau_R \geq 1$, viscosities develop nontrivial frequency dependence, and hydrodynamics breaks down. In our theory for nematic elastomers, we assume that there is a single director relaxation time $\tau$ and that it is much larger than $\tau_R$ so that we do not need to worry about the frequency dependence of viscosities arising from Rouse modes. We will, however, point out where these assumptions, also made by TW, can be modified. If $\tau \leq \tau_R$, dynamics at frequencies $\omega \tau_R \geq 1$ will be dominated by Rouse modes, and it may be difficult to distinguish director relaxation modes from Rouse modes in experiments in which displacements are probed. The hydrodynamical description of the preceding section, however, remains valid when $\omega \tau_R \ll 1$.

A. Equations of motion

When we keep the director as a dynamical variable, our formulation is closely related to nematodynamics. The equation of motion for the director has a reactive coupling to the velocity $\mathbf{v} = \dot{\mathbf{u}}$ arising from the Poisson bracket of $n_i(x)$ with $g_j(x')$. This Poisson bracket is generally derived in Eulerian coordinates in which there is a contribution $\mathbf{v} \cdot \nabla \mathbf{n}$ to the equation for $\partial_t \mathbf{n}$. This term can be combined with $\partial_t \mathbf{n}$ to yield the Lagrangian time derivative $\dot{\mathbf{n}}$. The remaining part of the Poisson bracket is

$$\{n_i(x), g_j(x')\} = -\lambda_{ijk} \partial_k \delta(x - x').$$  

(4.1)

The properties of the tensor $\lambda_{ijk}$ are dictated by three constraints: First, the magnitude of the director has to be conserved, i.e., $\mathbf{n} \cdot \dot{\mathbf{n}} = 0$ implying $n_i \lambda_{ijk} = 0$; second, the equations of motion must be invariant under $\mathbf{n} \rightarrow -\mathbf{n}$ implying $\lambda_{ijk}$ must change sign with $\mathbf{n}$; and third, under rigid uniform rotations, the director has to obey $\dot{\mathbf{n}} = \frac{1}{2} (\nabla \times \mathbf{u}) \times \mathbf{n}$. The only tensors and vectors available to use for the construction of $\lambda_{ijk}$ are $\delta_{ij}$, $n_i$, and the Levi-Civita anti-symmetric tensor $\epsilon_{ijk}$, and the only tensors that satisfy the first two conditions above are $\delta_{ij} n_k$ and $\delta^T_{ik} n_j$ where $\delta^T_{ij} = \delta_{ij} - n_i n_j$. Thus, the first two conditions imply that $\lambda_{ijk}$ has two independent components, which can be expressed as parts symmetric
and antisymmetric under interchange of $j$ and $k$:

$$\lambda_{ijk} = \frac{\lambda}{2} (\delta^T_{ij} n_k + \delta^T_{ik} n_j) + \frac{\lambda_2}{2} (\delta^T_{ij} n_k - \delta^T_{ik} n_j).$$  \(4.2\)

The third condition implies that the coefficient $\lambda_2$ of the antisymmetric part must be equal to $-1$. There are no constraints on the value of $\lambda$, which is equal to the ratio of two dissipative coefficients of the Leslie-Eriksen theory \[43\]. When its absolute magnitude is positive, it determines the equilibrium tilt angle of the director under uniform shear \[23\].

Using the Poisson brackets for the director and those for the momentum density and displacement discussed in the preceding section, we obtain the equations of motion for a nematic elastomer

\[\dot{n}_i = \lambda_{ijk} \partial_j \dot{u}_k - \Gamma \frac{\partial H}{\partial n_i}, \quad (4.3a)\]

\[\dot{u}_i = \frac{1}{\rho} \dot{q}_i, \quad (4.3b)\]

\[\dot{g}_i = \lambda_{kji} \partial_j \partial_k \dot{u}_k - \frac{\partial H}{\partial g_i} + \nu_{ijkl} \partial_j \partial_k \dot{u}_k. \quad (4.3c)\]

The first terms on the respective right hand sides of Eqs. \(4.3a\) and \(4.3b\) as well as the first and second term on the right hand side of Eq. \(4.3b\) stem from the Poisson brackets. $H$ is the full elastic energy of NEs as stated in Eq. \(4.1\). The second term on the right hand side of Eq. \(4.3b\) is a dissipative term that describes diffusive relaxation. There is no dissipative contribution to Eq. \(4.3b\) because NEs are tethered. $\nu_{ijkl}$ is a viscosity tensor that has the same structure as $\eta_{ijkl}$. It has five independent components $v_1$ to $v_5$. We use different symbols here for the viscosities than we used in Sec. \(3\) so that we can cleanly keep track of differences between the two theories.

We have assumed in Eqs. \(4.3a\) that both $\Gamma$ and $\nu_{ijkl}$ are local in time or, equivalently, that their temporal Fourier transforms are independent of frequency as they are at frequencies $\omega < \tau^{-1}$ than their respective inverse characteristic times $\tau^{-1}$ and $\tau^{-1}$. At larger frequencies, however, both $\Gamma$ and $\nu_{ijkl}$ do depend on frequency. In polymer gels, viscosities are proportional to $\sqrt{\omega}$ at frequencies $\omega \tau^{-1} \gg 1$ because of the large number of closely spaced modes contributing at these frequencies. Though there are to our knowledge no microscopic calculations of $\Gamma(\omega)$, there is no reason why many closely spaced modes should not lead to Rouse-like behavior at frequencies $\omega \tau^{-1} \gg 1$. There is also no reason why $\tau_{\Gamma}$ and $\tau_R$ should not be equal or nearly so. If the decay time $\tau$ for the nonhydrodynamic director modes predicted by the the theory with the low frequency approximations to $\Gamma$ and $\nu_{ijkl}$ is greater than $\tau_{\Gamma}$ and $\tau_R$, then this theory provides a correct description of the dynamics for frequencies $0 < \omega < \min(\tau_{\Gamma}^{-1}, \tau_R^{-1})$ including $\omega \sim \tau^{-1}$. If on the other hand $\tau < \min(\tau_R, \tau_{\Gamma})$, the full frequency dependence of $\Gamma$ must be used for frequencies $\omega \gg \min(\tau_{\Gamma}^{-1}, \tau_R^{-1})$. In all cases, however, the hydrodynamic theory of the preceding section is regained in the limit $\omega \ll \min(\tau_{\Gamma}^{-1}, \tau_R^{-1})$. To keep our discussion simple, we will continue to assume that both $\Gamma$ and $\nu_{ijkl}$ are frequency independent. Our equations can, however, incorporate the frequency dependence of these quantities merely by replacing them by their frequency-dependent forms, $\Gamma(\omega)$ and $\nu_{ijkl}(\omega)$.

At this point we would like to comment on the precise relation of Eqs. \(4.3\) to the equations of nematodynamics. One retrieves the latter from the former by setting all $\Gamma$ and $\nu_{ijkl}$ to $\nu_{ijkl}$ or $\rho$ with help of Eq. \(4.3b\). To facilitate in contact with the work of TW we choose to keep $\nu$. For the same reason we opt to work with $\mathbf{Q}$ rather than $\mathbf{n}$. This represents no difficulty since $\mathbf{Q}$ and $\mathbf{n}$ are simply related via Eq. \(4.3\). Collecting we obtain after some algebra the following effective equations of motion for $\mathbf{Q}$ and $\mathbf{u}$:

\[
\{[\partial_t + \Gamma D_1] \delta_{ab} - \Gamma M_{ab} (\mathbf{v})\} \dot{Q}_b = [\lambda \partial_t - \Gamma D_2] u_{az} + \Gamma M_{ab} (\mathbf{v}) \dot{\Omega}_b, \quad (4.4a)\]

and

\[
\rho \partial_t^2 u_a = -\frac{\lambda + 1}{2 \Gamma} \partial_t \partial_b [Q_a - \lambda u_{az}] + [C_2 + \nu_2 \partial_t] \partial_b u_{zz} + 2[C_4 + \nu_4 \partial_t] \partial_b u_{ab} + C_5 + \nu_5 \partial_t \partial_b \frac{D_2}{2} \partial_b u_{az} + \frac{D_2 - D_1}{2} \partial_b Q_a, \quad (4.4b)\]

\[
\rho \partial_t^2 u_z = -\frac{\lambda - 1}{2 \Gamma} \partial_t \partial_b [Q_b - \lambda u_{bz}] + C_1 + \nu_1 \partial_t \partial_b + C_2 + \nu_2 \partial_t \partial_b u_{zz} + 2[C_3 + \nu_3 \partial_t] \partial_b u_{ii} + C_5 + \nu_5 \partial_t \partial_b \frac{D_2}{2} \partial_b u_{bz} + \frac{D_1 + D_2}{2} \partial_b Q_b. \quad (4.4c)\]

To make contact to uniaxial solids we now take a brief detour and consider the simplified case $M_{ab} (\mathbf{v}) = 0$. In this case Eq. \(4.4a\) is diagonal in frequency space and readily solved with the result

\[
Q_a = -\frac{D_2}{D_1} \frac{1 - i \omega \tau_{\Gamma}^{-1}}{1 - i \omega \tau_{\Gamma}^{-1}} u_{az}. \quad (4.5)\]

In writing Eq. \(4.5\) we have used the relaxation times $\tau_1 = 1/(\Gamma D_1)$ and $\tau_2 = -\lambda/(\Gamma D_2)$. $\tau_1 \approx 10^{-2}$ s \[11, 12\] is essentially the relaxation time of the director. Pure hydrodynamic behavior is obtained when both $\omega \tau_1$ and
\( \omega \tau_2 \) are much less than 1 as our calculations will verify. In terms of the relaxation times, \( \lambda \) is given by
\[
\lambda = -\tau_2 D_2 / (\tau_1 D_1) .
\]
Note that \( \lambda \) is usually negative, but if it is positive, there will be no inconsistencies arising from a negative \( \tau_2 \).

Inserting expression (4.5) into the time-wise Fourier transformed equations of motion for \( u_a \) and \( u_z \), we obtain
\[
-\rho \omega^2 u_a = C_2(\omega) \partial_a u_{zz} + C_3(\omega) \partial_a u_{ii} + 2C_4(\omega) \partial_b u_{ab} + C^R(\omega) \partial_t u_{az} ,
\]
(4.6a)
\[
-\rho \omega^2 u_z = [C_1(\omega) + C_2(\omega)] \partial_z u_{zz} + [C_2(\omega) + C_3(\omega)] \partial_z u_{ii} + C^R(\omega) \partial_t u_{bz} ,
\]
(4.6b)
with \( C_1(\omega) = C_1 - i\omega \tau_1, C_2(\omega) = C_2 - i\omega \tau_2 \) etc. and
\[
C^R(\omega) = C_5 - i\omega \left( \nu_5 + \frac{\lambda^2}{2\tau_2} \right) - \frac{D_2}{2D_1} \left( 1 - i\omega \tau_2 \right)^2
\]
where
\[
\nu_5^R = \nu_5 + \frac{\lambda^2}{2\tau_2} \left( 1 - \frac{\tau_1}{\tau_2} \right)^2 .
\]
Note that Eqs. (4.6) are identical in form to Eq. (4.2) for uniaxial solids with \( K^R_1 = K^R_3 = 0 \). Note also that we can identify \( \eta_1 = \nu_1 \) and so on. Indicating the consistency of our two approaches, we can identify the renormalized viscosities \( \eta_5^R \) and \( \nu^R_5 \). As pointed out earlier, Rouse modes will be important for \( \omega \tau_R \geq 1 \) and \( \omega \tau_1 \geq 1 \) leading to a frequency dependence of the viscosities. In this regime we have to let \( \nu_i \to \nu_i(\omega) \) and \( \Gamma \to \Gamma(\omega) \). For \( \omega \tau_R \gg 1 \) and \( \omega \tau_1 \gg 1 \), in particular, the viscosities are proportional to \( \sqrt{\omega} \).

Before we move on we point out that Eq. (4.6) become identical to the equations of motion by TW if we take the incompressible limit and if we neglect the Frank energy, provided of course, that we take into account that somewhat different conventions are used and provided that corresponding quantities are identified properly. A detailed comparison to TW at the level of final results for the modes will be given in Sec. IV.C.

B. Mode structure

1. The \( t \)-direction

By applying \( \tilde{\epsilon}_{ba} q_b \) (including the summation over \( a \)) to both sides of the Fourier transformed version of Eq. (4.6) we obtain a diagonalized equation of motion for \( Q_t \) that is readily solved with the result
\[
Q_t = \frac{D_2}{D_1} \frac{1 - i\omega \tau_2 + q^2 K_1}{1 - i\omega \tau_1 + q^2 K_1} \frac{1}{2} iq_z u_t ,
\]
(4.9)
where we have used the abbreviated notation
\[
q^2 K_1 = K_2 q_1^2 + K_3 q_2^2 .
\]
(4.10)

Application of the same procedure to Eq. (4.6b) yields
\[
-\omega^2 \rho u_t = \frac{\lambda + 1}{2\Gamma} i\omega q_z \left[ iQ_t + \frac{\lambda}{2} q_z u_t \right] - C_4(\omega) q_1^2 u_t
\]
\[
+ \frac{D_2 - D_1}{2} iq_z Q_t - \frac{1}{2} \left[ C_5(\omega) - \frac{D_2}{D_1} \right] u^2 .
\]
(4.11)

\( u_t \) is a purely transverse displacement that does not couple to any displacement in the plane containing \( q \). As a result, the elastic constants \( C_2 \) and \( C_3 \), which couple to \( u_t \), do not appear in this equation of motion.

Next we insert Eq. (4.9) into (4.11) which gives an effective equation of motion for \( u_t \) alone. This equation of motion has solutions with \( u_t \neq 0 \) for frequencies satisfying the secular equation
\[
\omega^2 \rho - C_4(\omega) q_1^2 + \frac{\lambda(\lambda + 1) i\omega - \lambda \tau_2^{-1} - 2\Gamma C_5(\omega) q^2}{4\Gamma} q_z^2
\]
\[
\times D_1 \left[ 1 - i\omega \tau_1 + q^2 K_1/D_1 \right]
\]
\[
+ \frac{\lambda + 1 - \tau_1^{-1} - \tau_2^{-1}}{4\Gamma} q^2 D_2 \left[ 1 - i\omega \tau_2 + q^2 K_1/D_2 \right] = 0 .
\]
(4.12)

Evidently this secular equation is too complicated to find useful closed solutions for \( \omega \). However, since we are interested in the long-wavelength behavior, we can perturbatively determine solutions in the form of a power series in the wavevector.

Let us first consider the simplified case of vanishing \( C \)’s and \( D \)’s. In this case we obtain as expected the well known slow and fast \( t \)-direction modes of nematic liquid crystals
\[
\omega_{1,s} = -i \left[ K_2 q_1^2 + K_3 q_2^2 \right] \left\{ \Gamma + \frac{(1 + \lambda)^2 q^2}{2(\nu_5 q_1^2 + 2\nu q_1^2)} \right\} ,
\]
(4.13a)
\[
\omega_{1,f} = -i \frac{2\nu q_1^2}{2\rho} v_5 q_2^2 ,
\]
(4.13b)
as well as the previously announced spurious zero-frequency mode. In writing Eqs. (4.13) we have considered as usual the limit \( K \rho \Gamma/\nu \ll 1 \), where \( K \) stands symbolically for all the Frank constants and \( \nu \) stands ambiguously for all the viscosities. In comparing Eqs. (4.13) and (4.14) to the original results on nematic liquid crystals as given in Ref. 33 one should be aware of slight differences in the notations. If we wish to use the notation of Ref. 33 we have to replace in Eqs. (4.13) \( \Gamma \to \gamma^{-1}, \nu_1 \to 2\nu_1, \nu_4 \to \nu_2, \nu_5 \to 2\nu_3, q_1 \to q_1 \) and \( q_2 \to q_3 \).

Now to the full secular equation (4.12). Solving this equation perturbatively leads also to three modes, namely one massive mode and two propagating modes.
The massive mode has the frequency

$$\omega_{t,m} = -i \tau_1^{-1} - i \Gamma \left[ K_2 q_\perp^2 + K_3 q_z^2 \right] + i \frac{\nu_5^R - \nu_5}{2 \rho} q_z \omega_z. \quad (4.14)$$

Note that Eq. (4.14) does not depend on $C_5^R$. Hence, this mode is shared by soft and semi-soft NEs. As long as the sound velocities of the soft modes are finite, their frequencies are

$$\omega_{t,\pm} = \pm \sqrt{2C_4 q_\perp^4 + C_5^R q_z^4 - i \frac{2\nu_4 q_\perp^4 + \nu_5^R q_z^2}{4\rho}}, \quad (4.15a)$$

in full agreement with Eq. (3.19a) when $C_5^R = 0$ and the identifications $\nu_4 = \eta_4$ and $\nu_5^R = \eta_5^R$ are made. For soft NEs the soft $t$-modes become diffusive if $q_\perp = 0$. The frequencies of these diffusive modes are easily identified with those given in Eqs. (3.20) and (3.21).

2. The $T$-direction

Except for the $t$-direction analysis of the modes is prohibitively complicated unless one resorts to the incompressible limit. In this limit $C_3 = \infty$ and $u_t = 0$, and as a result, $C_2, C_3, \nu_2$ and $\nu_3$ do not appear in the equations of motion for $u_T$.

We need the equations of motion for the $\perp$ direction as intermediate results for studying the $T$-direction. Applying $q_\perp$ (including the summation over $a$) to the Fourier transformed counterparts of Eqs. (4.16a) and (4.16b) we obtain

$$Q_\perp = -D_2 \frac{1 - i \omega \tau_2 + q_\perp^2 K_1 / D_2}{D_1 1 - i \omega \tau_1 + q_\perp^2 K_1 / D_1} 2 i q_\perp u_z$$

$$\omega^2 p_{\perp} = \frac{\lambda + 1}{2 \Gamma} i \omega q_\perp \left[ i Q_\perp + \frac{\lambda}{2} (q_\perp u_z + q_\perp u_{\perp}) \right]$$

$$-2C_4(q_\perp^2 u_\perp + \frac{D_2 - D_1}{2} i q_\perp Q_\perp)$$

$$-\frac{1}{2} \left[ C_5(q_\perp u_z + q_\perp u_{\perp}) \right], \quad (4.16a)$$

where we have used the shorthand

$$q^2 K_\perp = K_1 q_\perp^2 + K_3 q_z^2. \quad (4.17)$$

In addition to Eqs. (4.16a) we also need the Fourier transformed version of Eq. (4.16c) in the incompressible limit,

$$-\omega^2 p_{u_z} = \frac{\lambda - 1}{2 \Gamma} i \omega q_\perp \left[ i Q_\perp + \frac{\lambda}{2} (q_\perp u_z + q_\perp u_{\perp}) \right]$$

$$-C_1(q_\perp^2 u_z + \frac{D_1 + D_2}{2} i q_\perp Q_\perp)$$

$$-\frac{1}{2} \left[ C_5(q_\perp u_z + q_\perp u_{\perp}) \right]. \quad (4.18)$$

Next we set up an equation of motion for $u_T$ that depends on $u_\perp, u_z$ and $Q_\perp$. Then we eliminate $Q_\perp$ with help of Eq. (4.16a). Finally, we exploit that $u_\perp = -q_\perp u_T / q$ and $u_z = q_\perp u_T / q$ in the incompressible limit. These steps provide us with an effective equation of motion for $u_T$ alone. In order to allow for solutions $u_T \neq 0$ the frequencies have to satisfy a condition analogous to Eq. (4.12). We opt not to write down this secular equation here because it is rather lengthy and because it can be obtained in a straightforward manner from the ingredients given above.

We proceed as above and first consider the simplified case of vanishing $C'$s and $D'$s. As anticipated we obtain a spurious zero-frequency mode as well as the slow and fast $T$-direction modes of nematodynamics,

$$\omega_{T,s} = \frac{-i [K_1 q_\perp^2 + K_3 q_z^2] }{2\rho q^2}$$

$$\times \left\{ \Gamma + \frac{[q_\perp^2 - \lambda(q_\perp^2 - q_z^2)]^2}{2\nu_4(q_\perp^2 - q_z^2)^2 + 4(\nu_1 + 2\nu_4)q_\perp q_z^2} \right\}, \quad (4.19a)$$

$$\omega_{T,f} = -i \frac{2(\nu_1 + 2\nu_4) q_\perp^2 q_z^2 + \nu_5(q_\perp^2 - q_z^2)^2}{2\rho q^2}, \quad (4.19b)$$

where $K \rho \Gamma / \nu \ll 1$ is implied.

By perturbatively solving the full secular equation we extract the three $T$-direction modes for NEs. We find one massive mode

$$\omega_{T,m} = \frac{-\tau_1^{-1} - i \Gamma [K_1 q_\perp^2 + K_3] q_z^2}{2\rho}$$

$$+ \frac{\nu_5^R - \nu_5}{2 \rho} \left( q_\perp^2 q_z^2 \right) \quad (4.20)$$

and two soft modes. For non-vanishing sound velocities the soft modes have frequencies

$$\omega_{T,\pm} = \pm \sqrt{2(C_1 + 2C_4) q_\perp^2 q_z^2 + C_5^R(q_\perp^2 - q_z^2)^2}$$

$$\times \frac{-2\rho q^2}{4\rho q^2}$$

$$- \frac{2(\nu_1 + 2\nu_4) q_\perp^2 q_z^2 + \nu_5(q_\perp^2 - q_z^2)^2}{4\rho q^2}. \quad (4.21)$$

When $C_5^R = 0$ this result reduces, provided that the viscosities are properly identified, to Eq. (4.19b). In the case of ideal soft elasticity the sound velocities vanish if $q_\perp = 0$ or $q_z = 0$. For $q_\perp = 0$ the frequencies of the then diffusive $T$-modes are identical to those for the diffusive $t$-modes, see Eqs. (3.20) and (3.21). For $q_z = 0$ we retrieve Eqs. (3.22) and (3.23).

C. Comparison to TW

Now we compare our findings to those by TW. First, we will demonstrate that our equations of motion (4.4) are identical to the equations of motion by TW if we restrict ourselves to the incompressible limit and if we neglect the Frank energy. Second, we will compare our
TABLE I: Correspondence between quantities used by TW and quantities used in our work.

| TW this paper | this paper |
|---------------|------------|
| 2C1          | C1        |
| 2C2          | C2        |
| 2C3          | C3        |
| 2C4          | C4        |
| 2A1          | ν1        |
| 2A2          | ν2        |
| 2A3          | ν3        |
| 4A5          | ν5 + \sqrt{A_4}/(2Γ) |
| 4A5          | ν5        |
| 1/Γ          | τ1        |
| 1/Γ          | τ2        |
| 1/Γ          | τ3        |

where the

Equation (4.24) reveals that the stress tensor 

and (4.4c) as 

in identifying corresponding quantities, see Table I.

In order to compare our equations of motion (4.4) to the equations of motion by TW we rewrite Eqs. (4.4b) and (4.4c) as

\[ -\rho \partial_t^2 u_i = \partial_j \sigma_{ij} , \quad (4.22) \]

where the \( \sigma_{ij} \) are the components of the stress tensor \( \sigma \). The specifics of the \( \sigma_{ij} \) are easily gathered from Eqs. (4.24b) and (4.24c),

\[ \sigma_{ab} = 2 \left[ C_4 + \nu_4 \partial_t \right] u_{ab} , \quad (4.23a) \]

\[ \sigma_{zz} = \left[ C_1 + \nu_1 \partial_t \right] u_{zz} , \quad (4.23b) \]

\[ \sigma_{az} = \left[ C_5 + \nu_5 \partial_t - \frac{D_2}{2} + \frac{\lambda(\lambda + 1)}{2Γ} \partial_t \right] u_{az} \]

\[ + \frac{1}{2} \left[ D_2 - D_1 - \frac{\lambda + 1}{Γ} \partial_t \right] Q_a , \quad (4.23c) \]

\[ \sigma_{za} = \left[ C_5 + \nu_5 \partial_t + \frac{D_2}{2} + \frac{\lambda(\lambda - 1)}{2Γ} \partial_t \right] u_{az} \]

\[ + \frac{1}{2} \left[ D_2 + D_1 - \frac{\lambda - 1}{Γ} \partial_t \right] Q_a , \quad (4.23d) \]

where we have taken the incompressible limit. Equations (4.23) show clearly that \( \sigma \) is not symmetric and that its antisymmetric part is

\[ \sigma_{az} - \sigma_{za} = \frac{1}{Γ} \left\{ [\tau_1^{-1} + \partial_t] Q_a - \lambda [\tau_2^{-1} + \partial_t] u_{az} \right\} \]

\[ = M_{ab}(\nabla) \tilde{Ω}_b \quad (4.24) \]

where we have used Eqs. (4.4a) to obtain the last equality. Equation (4.24) reveals that the stress tensor \( \sigma_{ij} \) defined in Eqs. (4.24) is symmetric only when the Frank energy can be ignored. Of course, when the Frank energy is included, it is always possible, following the procedures in Refs. [33] and [34], to define a symmetric stress tensor \( \sigma_{ij}^{\text{sym}} \) yielding the same equations of motion as \( \sigma_{ij} \).

If we neglect the Frank energy, our stress tensor becomes identical to the symmetric stress tensor used by TW provided that we take into account Table I. Moreover, as easily can be checked, Eq. (4.4a) becomes identical to the balance of torques equation under these circumstances. Therefore, our equations of motion (4.4) are equivalent to the equations of motion by TW provided that we take the incompressible limit and neglect the Frank energy.

At this point it is interesting to compare the stability conditions which are implicit in the dissipation function. Our equations imply in the incompressible limit that \( ν_1 ≥ 0, ν_4 ≥ 0 \) and \( ν_5 ≥ 0 \). While the first two conditions can be readily identified with the conditions \( A_1 ≥ 0, A_4 ≥ 0 \) in the work of TW, the situation is less obvious for the last condition. Note that we can re-express \( ν_5 ≥ 0 \) in terms of the quantities used by TW as

\[ ν_5 = 4A_5 - \frac{\kappa^2}{2τ_1} ≥ 0 . \quad (4.25) \]

which has an identical counterpart in the work of TW. Stated in terms of the relaxation times, this condition requires that

\[ τ_1 τ_R ≥ \frac{\kappa^2}{2} \quad (4.26) \]

for ideally soft NEs.

Next we come to the comparison of the results for the modes. Our first observation is here that the massive modes in the rubbery matrix. We obtain

\[ \omega^2 = \frac{1}{2ρ} \left[ 2C_4(ω) q_1^2 + C_5^R(ω) q_2^2 \right] . \quad (4.27) \]

To the order we are working, i.e. to order \( O(q^0) \), the solutions to Eq. (4.27) and our \( ω_{t,m} \) coincide. Note that Eq. (4.27) is essentially identical to the \( t \)-direction secular equation for uniaxial solids. This can easily be checked by starting with Eq. (4.6a) and by then switching to the \( t \)-direction.

To make further contact to WT we eliminate the viscosities in Eq. (4.27) in favor of the relaxation time \( τ_R ≈ ν_1/C_1 ≈ \cdots ≈ ν_5/C_5 ≈ [ν_5 + \lambda^2/(2Γ)]/C_5 \). The subscript \( R \) indicates that \( τ_R \approx 10^{-5} - 10^{-6} s \) is of the order of the Rouse time of the polymers constituting the rubbery matrix. We obtain

\[ \omega^2 = \frac{1}{2ρ} \left[ 2C_4 q_1^2 + \tilde{C}_5^R(ω) q_2^2 \right] (1 - iωτ_R) , \quad (4.28) \]

where

\[ \tilde{C}_5^R(ω) = \frac{C_5^R(ω)}{1 - iωτ_R} \]

\[ = C_5 - \frac{D_2^3}{2D_1} \frac{(1 - iωτ_1)(1 - iωτ_R)}{(1 - iωτ_R)} . \quad (4.29) \]

Taking into account Table I we see that our \( \tilde{C}_5^R(ω) \) is identical with the renormalized form of \( C_5 \) found by TW.
and that Eq. (4.20) is in full agreement with the dispersion relation of TW for the qSH waves.

Now to the $T$-direction. To order $O(q^2)$ our results for $\omega_{T,\pm}$ coincide with the solutions of

$$\omega^2 = \frac{1}{2\rho} \left[ C_1(\omega) + 2C_4(\omega) \right] \frac{q^2}{q^2} + C_5^R(\omega) \left( \frac{q_1^2 - q_2^2}{q^2} \right)^2. \tag{4.30}$$

Note that Eq. (4.30) is essentially identical to the $T$-direction secular equation for conventional uniaxial solids. TW considered the $T$-modes only for $q_z = 0$. For $q_z = 0$ Eq. (4.30) reduces to

$$\omega^2 = \frac{1}{2\rho} \tilde{C}_5^R(\omega) q_z^2 (1 - i\omega\tau_R). \tag{4.31}$$

Using Table I find that Eq. (4.31) is in full agreement with the result of TW for the qSV waves.

Before summarizing our findings, we now briefly return to the property of NEs that makes them, as pointed out by TW, candidates for acoustic polarizers, viz. the large difference in attenuation between the $t$- the $T$-modes in the symmetry direction where $q_z = 0$. For the $t$-direction it follows immediately from Eq. (4.28) that the attenuation is proportional to $\omega$ if $C_5^R = 0$ and $q_z = 0$. Equation (4.31) has two solutions for $C_5^R = 0$, namely $\omega = 0$ and, with the proper identification $\eta_0^R = \eta_5^R$, the fast diffusive mode Eq. (4.28b). If the Frank energy is taken into account, the $\omega = 0$ mode becomes the slow diffusive mode of Eq. (4.28b). Due to their diffusiveness, the attenuation of these $T$-modes is proportional to $\sqrt{\omega}$ and hence much larger for low frequencies than the attenuation of the propagating $t$-mode. This large difference in attenuation can be used, in principle, to split the $T$-modes from the $t$-modes.

**V. SUMMARY**

Nematic elastomers exhibit the remarkable phenomenon of soft or semi-soft elasticity in which the effective shear modulus $C_5^R$ for shears in planes containing the anisotropy axis respectively vanishes or is very small. In this paper, we have explored the dynamical consequences of this elasticity. We derived dynamical equations, involving only the displacement, valid in the hydrodynamic limit in which frequencies and wavenumbers are respectively small compared to all characteristic microscopic inverse times and lengths in the system, and we determined that their mode structure is identical to that of columnar liquid crystals in the soft limit when $C_5^R = 0$. We also used the Poisson-bracket approach to derive dynamical equations, which contain non-hydrodynamic modes, for both director and displacement and verified that they reduced to those derived by Terentjev, Warner, and coworkers when contributions from the Frank free energy can be ignored. We analyzed the mode structure of these equations assuming a single relaxation time for the director, which we took to be longer than any other characteristic decay time such as the Rouse time $\tau_R$. Our equations, however, permit the introduction of frequency-dependent dissipative coefficients valid at frequencies higher than these inverse decay times.

Rheological experiments at zero wavenumber have reported frequency-dependent storage and loss moduli that are in agreement with the predictions of the semi-soft theory that includes the director [18]. It would be interesting to map out the modes of nematic elastomers directly using light scattering. It may, however, be difficult to access the true hydrodynamic limit because it applies in current NEs only for frequencies of order 100 Hz or less and because inherent sample inhomogeneities may lead to extra scattering that could mask the signals of the characteristic modes of a homogeneous system.

**Acknowledgments**

We gratefully acknowledge support by the Emmy Noether-Programm of the Deutsche Forschungsgemeinschaft (OS) and the National Science Foundation under grant DMR 00-95631(TCL).

---

[1] W. Warner and E.M. Terentjev, *Liquid Crystal Elastomers* (Clarendon Press, Oxford, 2003)
[2] H. Finkelmann, H. J. Koch, and G. Rehage, Makromol. Chem. Rapid Commun. 2, 317 (1981).
[3] P. G. de Gennes in *Liquid Crystals of One and Two-Dimensional Order*, p. 231, edited by W. Helfrich and G. Heppke (Springer, New York, 1980).
[4] E. M. Terentjev, J. Phys. Cond. Mat. 11, R239 (1999).
[5] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993).
[6] S. Chandrasekhar, *Liquid Crystals* (Cambridge University Press, Cambridge, 1992).
[7] J. Küpfner and H. Finkelmann, Macromol. Chem. Phys. 195, 1353 (1994).
[8] H. Finkelmann and H. Wermter, Polym. Mater. Sci. Eng. 82, 319 (2000).
[9] H. Finkelmann, E. Nishikawa, G. G. Pereira, and M. Warner, Phys. Rev. Lett. 87, 015501 (2001).
[10] L. Golubovi´ c and T. C. Lubensky, Phys. Rev. Lett. 63, 1082 (1989).
[11] H. Finkelmann, I. Kundler, E. M. Terentjev, and M. Warner, J. Phys. II (France) 7, 1059 (1997).
[12] G. C. Verwey, M. Warner, and E. M. Terentjev, J. Phys. II (France) 6, 1273-1290 (1996).
[13] M. Warner, J. Mech. Phys. Solids 47, 1355 (1999).
[14] O. Stenull and T. C. Lubensky, Europhys. Lett. 61, 776...
(2003). See also, X. Xing and L. Radzihovsky, Europhys. Lett. 61, 769 (2003).

[15] O. Stenull and T. C. Lubensky, to appear in Phys. Rev. E (eprint: cond-mat/0307686).

[16] X. Xing and L. Radzihovsky, Phys. Rev. Lett. 90, 168301 (2003).

[17] J. L. Gallani, L. Hilliou, P. Martinoty, and P. Keller, Phys. Rev. Lett. 72, 2109 (1994); J. L. Gallani, L. Hilliou, P. Martinoty, F. Doublet, and M. Mauzac, J. Phys. II (France) 6, 443 (1996).

[18] S. M. Clarke, A. R. Tajbakhsh, E. M. Terentjev and M. Warner, Phys. Rev. Lett. 86, 4044 (2001); S. M. Clarke, A. Hotta, A. R. Tajbakhsh, E. M. Terentjev Phys. Rev. E 65, 021804 (2002).

[19] S. M. Clarke, A. R. Tajbakhsh, E. M. Terentjev, C. Remillat, G. R. Tomlinson and J. R. House, J. Appl. Phys. 89, 6538 (2001).

[20] P. Stein, N. Assfalg, H. Finkelmann, and P. Martinoty, Eur. Phys. J. E 4, 255 (2001).

[21] A. Hotta and E. M. Terentjev, Eur. Phys. J. E 10, 291 (2003).

[22] E. M. Terentjev, I. V. Kamotski, D. D. Zakharov, and L. J. Fradkin, Phys. Rev. E 66, 052701(R) (2002); see also L. J. Fradkin, I. V. Kamotski, E. M. Terentjev and D. D. Zakharov, Proc. R. Soc. Lond. A 459, 2627 (2003).

[23] P. I. C. Teixeira and M. Warner, Phys. Rev. E 60, 603 (1999).

[24] E. M. Terentjev and M. Warner, Eur. Phys. J. E 4, 343 (2001).

[25] See for example, H. Goldstein, *Classical Mechanics* (Addison-Wesley, Reading, 1980).

[26] J. L. Ericksen, Arch. Ration. Mech. Anal. 4, 231 (1960); Trans. Soc. Rheol. 5, 23 (1961).

[27] F. M. Leslie, Q. J. Mech. Appl. Math. 19, 357 (1966); Arch. Ration. Mech. Anal. 28, 265 (1968).

[28] Groupe d’Etudes des Cristaux Liquides (Orsay), J. Chem. Phys. 51, 816 (1969).

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

[30] K. Kawasaki, in *Phase Transitions and Critical Phenomena*, vol. 2, Eds. C. Domb amd M.S. Green (Academic, New York, 1976).

[31] See, e. g., P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics*, (Cambridge University Press, Cambridge, 1995).

[32] P.C. Hohenberg and B.I. Halperin, Rev. Mod. Phys. 49, 435 (1977).

[33] D. Forster, T. C. Lubensky, P. C. Martin, J. Swift, and P. S. Pershan, Phys. Rev. Lett. 26, 1016 (1971).

[34] Dieter Forster, *Hydrodynamic Fluctuations, Broken symmetry, and Correlation Functions* (Addison Welsey, Reading, Mass., 1983); D. Forster, Phys. Rev. Lett. 32, 1161 (1974).

[35] See for example, J. Z. Xue, D. J. Pine, X. L. Wu, and P. M. Chaikin, Phys. Rev. A 46 6550 (1992).

[36] See for example, H. Riskin, *The Fokker-Planck Equation: Methods of Solutions and Applications* (Springer Verlag, Berlin, 1984), and Ref. [31].

[37] Poisson brackets of coarse-grained variables require a proper interpretation. Perhaps most naturally, they can be regarded as coarse grained versions of the corresponding microscopic Poisson brackets.

[38] H. K. Janssen, Z. Phys. B: Cond. Mat. 23, 377 (1976); R. Bausch, H. K. Janssen, and H. Wagner, *ibid.* 24, 113 (1976).

[39] C. DeDominicis, J. Physique C 37, 247 (1976); C. DeDominicis and L. Peliti, Phys. Rev. B 18, 353 (1978).

[40] H. K. Janssen, in: *Dynamical Critical Phenomena and Related Topics*, Lecture Notes in Physics, Vol. 104, ed. C. P. Enz (Springer, Heidelberg, 1979); H. K. Janssen, in: *From Phase Transition to Chaos*, ed. G. Györgyi, I. Kondor, L. Sasvári, T. Tél (World Scientific, Singapore, 1992).

[41] J. Schmidtke, W. Stille, and G. Strobl, Macromolecules 33 2922 (2000).

[42] M. Schönstein, W. Wille, and G. Strobl, Eur. Phys. J. E 5, 511 (2001).

[43] Beyond harmonic order, the left Cauchy strain tensor $\nu_{ij} = (\partial_i u_j + \partial_j u_i + \partial_k u_i \partial_k u_j)/2$ rather than the usual right Cauchy strain tensor $\omega_{ij} = (\partial_i u_j + \partial_j u_i - \partial_k u_i \partial_k u_j)/2$ couples to the director. See T. C. Lubensky, R. Mukhopadhyay, L. Radzihovsky, and X. J. Xing, Phys. Rev. E 66, 011702 (2002).

[44] See, e.g., M. Rubinstein and R. H. Colby, *Polymer Physics* (Oxford University Press, Oxford, 2003).

[45] H. Stark and T. C. Lubensky, Phys. Rev. E 67, 061709 (2003).

[46] P. G. de Gennes, in *Liquid Crystals of One- and Two-Dimensional Order*, edited by W. Helfrich and G. Høppe (Springer, Berlin, 1980).