

Dynamics of nematic elastomers

Olaf Stenull and T. C. Lubensky

Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

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We study the low-frequency, long-wavelength dynamics of soft and semisoft nematic elastomers using two different but related dynamic theories. Our first formulation describes the pure 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 nematohydrodynamics by Forster *et al.* to nematic elastomers. It treats the director explicitly and describes slow modes beyond the hydrodynamic limit.

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I. INTRODUCTION

Nematic elastomers (NE's) [1–4] 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 NE's 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 NE's. For example, temperature change or illumination can change the orientational order and cause the elastomer to extend or contract by several hundred percent [7–9]. Nematic elastomers display a soft elasticity [10–13] 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 [14–16].

In addition to unusual static properties, NE's have an intriguing dynamic mechanical behavior. Early rheology experiments on liquid crystalline elastomers [17] found no nematic effects in NE's. Later, however, several experiments [18–21] 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 NE's. This behavior has been named dynamic soft elasticity. It makes NE's interesting for device applications in areas such as mechanical vibration damping [19] (exploiting the fact that the mechanical loss is record high over a wide range of temperatures and frequencies) or acoustics where NE's 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 NE's was pioneered by Terentjev and Warner (TW) and co-workers [1,18,22–24]. References [1] and [22] present a detailed derivation of equations governing the long-wavelength, low-frequency dynamics of NE's and derive their associated mode structure, which includes nonhydrodynamic, 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 nondissipative forces are calculated as derivatives of an energy functional and dissipative forces as derivatives of a Rayleigh dissipation function. The TW work focuses primarily on the rheological response in both soft and semisoft NE's at zero wave number, and it neglects contributions to dynamical equations arising from the Frank free energy for director distortions, which are higher order in wave number 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 NE's. 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 nematodynamics, excluding the rotational inertial term that is usually discarded [28], or is missing entirely in alternative derivations [5] of these equations.

In this paper, we will present alternative approaches to deriving the equations governing the dynamics of NE's. First, we will derive the exclusively hydrodynamical equations for those variables whose characteristic frequencies vanish with wave number. These equations, like those for the hydrodynamics of smectic [5,29] and columnar [5] liquid crystals, exclude nonhydrodynamic director modes. Interestingly, the modes for soft NE's 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 nonvanishing velocity at all angles. A second pair is predominantly transverse with a velocity that vanishes for wave vector either parallel or perpendicular to the uniaxial symmetry axis, and a third pair is completely transverse with a velocity that vanishes for wave vector parallel to the symmetry axis, but is nonzero otherwise. We then derive the phenomenological equations for the slow dynamics of all displacement and director variables in NE's 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 Forster *et al.* [33,34] in their derivation of the hydrodynamics of nematics. Our dynamical equations for NE's reduce to the equations of nematohydrodynamics derived by Forster *et al.* when elastic rigidities vanish and to the purely hydrodynamics equations for NE's 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. II we briefly review the general Poisson-bracket formalism for obtaining coarse-grained dynamics. Section III focuses on the pure hydrodynamics of NE's. We set up equations of motion for the momentum density and 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, elastic displacement, and director. These are then compared to the equations of motion for uniaxial solids. We determine the mode structure. Finally, we compare our results to the results 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 [36] approach to stochastic dynamics, which provides a description of dissipative processes and noise forces. Let $\Phi_\mu(\mathbf{x}, t)$, $\mu=1, 2, \dots$, be a set of coarse-grained fields whose statistical mechanics is described by a coarse-grained Hamiltonian \mathcal{H} . The dynamical equations for $\Phi_\mu(\mathbf{x}, t)$ are first-order differential equations in time:

$$\begin{aligned} \dot{\Phi}_\mu(\mathbf{x}, t) = & - \int d^d x' \int dt' \{ \Phi_\mu(\mathbf{x}, t), \Phi_\nu(\mathbf{x}', t') \} \frac{\delta \mathcal{H}}{\delta \Phi_\nu(\mathbf{x}', t')} \\ & - \Gamma_{\mu, \nu} \frac{\delta \mathcal{H}}{\delta \Phi_\nu(\mathbf{x}, t)} + \zeta_\mu(\mathbf{x}, t). \end{aligned} \quad (2.1)$$

Here and in the following the Einstein summation convention is understood. The first term on the right-hand side is a nondissipative velocity, also known as the reactive term, that contains the Poisson bracket $\{ \Phi_\mu(\mathbf{x}, t), \Phi_\nu(\mathbf{x}', t') \}$ of the coarse-grained fields [37]. The reactive term couples $\dot{\Phi}_\mu$ to $\delta \mathcal{H} / \delta \Phi_\nu$ only if Φ_μ and Φ_ν 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 $\dot{\Phi}_\mu$ to $\delta \mathcal{H} / \delta \Phi_\nu$ only if Φ_μ and Φ_ν have the same sign under time reversal. If the noise term ζ_μ 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 [38–40] 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 ω 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 NE's. These equations apply for frequencies ω such that $\omega \tau \ll 1$, where τ is the longest nonhydrodynamic 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 $\tau \sim 10^{-2}$ s [41,42] so that the regime of applicability of hydrodynamics is quite small for current NE's. 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 ϵ mass density ρ , and momentum density \mathbf{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 \mathbf{u} take their place. The second is that an elastomer is permanently cross-linked: it is a classical rubber in which changes $\delta \rho$ in mass density are locked to changes in volume such that $\delta \rho / \rho = -\nabla \cdot \mathbf{u}$ and in which permeation 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 $5+3-1=7$ independent variables and 7 associated hydrodynamic modes. These modes are heat diffusion and, depending on direction, either six propagating sound modes, four propagat-

ing sound modes and two diffusive displacement-velocity modes, or two longitudinal sound modes and four diffusive displacement-velocity modes. In what follows, we will consider only isothermal processes so that heat diffusion can be ignored.

A. Elastic energy

To derive the hydrodynamical equations for NE's, 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_5 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 [5], curvaturelike 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 directors. We will restrict our attention to harmonic distortions.

Elastomers are permanently cross-linked pieces of rubber whose static elasticity is most easily described in Lagrangian coordinates in which \mathbf{x} labels a mass point in the unstretched (reference) material and $\mathbf{R}(\mathbf{x})=\mathbf{x}+\mathbf{u}(\mathbf{x})$, where $\mathbf{u}(\mathbf{x})$ is the displacement variable, labels the position of the mass point \mathbf{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 $\mathbf{r} \equiv \mathbf{R}(\mathbf{x})$ 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:

$$\mathcal{H} = \mathcal{H}_{\mathbf{u}} + \mathcal{H}_{\mathbf{n}} + \mathcal{H}_{\mathbf{u},\mathbf{n}}, \quad (3.1)$$

where $\mathcal{H}_{\mathbf{u}}$ is the usual elastic energy of a uniaxial solid, $\mathcal{H}_{\mathbf{n}}$ is the Frank free energy of a nematic, and $\mathcal{H}_{\mathbf{u},\mathbf{n}}$ 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 [43],

$$\mathcal{H}_{\mathbf{u}} = \int d^3x \left\{ \frac{C_1}{2} u_{zz}^2 + C_2 u_{zz} u_{ii} + \frac{C_3}{2} u_{ii}^2 + C_4 u_{ab}^2 + C_5 u_{az}^2 \right\}. \quad (3.2)$$

Here u_{ij} are linearized components of the Lagrange strain tensor \underline{u} :

$$u_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i). \quad (3.3)$$

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 NE's [14,15], 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

$$\mathcal{H}_{\delta\mathbf{n}} = \int d^3x \left\{ \frac{K_1}{2} (\partial_a \delta n_a)^2 + \frac{K_2}{2} (\varepsilon_{ab} \partial_a \delta n_b)^2 + \frac{K_3}{2} (\partial_z \delta n_a)^2 \right\}, \quad (3.4)$$

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

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

where

$$M_{ab} = (K_1 - K_2) \partial_a \partial_b + (K_2 \partial_{\perp}^2 + K_3 \partial_z^2) \delta_{ab} \quad (3.6)$$

and where $\partial_{\perp}^2 = \partial_a \partial_a$ with the Einstein convention understood. The coupling energy, finally, can be written in the form

$$\mathcal{H}_{\mathbf{u},\mathbf{n}} = \int d^3x \left\{ \frac{D_1}{2} Q_a^2 + D_2 u_{za} Q_a \right\}, \quad (3.7)$$

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

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

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 \mathcal{H} over n_a to find

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

Inserting this equation into \mathcal{H} and retaining only the dominant terms in gradients, we obtain

$$\mathcal{H}_{u-\text{el}} = \int d^3x \left\{ \frac{C_1}{2} u_{zz}^2 + C_2 u_{zz} u_{ii} + \frac{C_3}{2} u_{ii}^2 + C_4 u_{ab}^2 + C_5^R u_{az}^2 + \frac{K_1^R}{2} (\partial_{\perp}^2 u_z)^2 + \frac{K_3^R}{2} (\partial_z^2 u_a)^2 \right\}, \quad (3.10)$$

where

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

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

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

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 $(\partial_\perp^2 u_z)^2$ or $(\partial_z^2 u_a)^2$ in $\mathcal{H}_{\mathbf{u-el}}$ 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, Eq. (3.11a), 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 nonideal behavior is known as semisoftness.

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 $K_1^R (\partial_\perp^2 u_z)^2 / 2$ is needed to stabilize it. The in-plane displacements u_a are analogous to those of columnar liquid crystal, which are soft for distortions in the z direction, and a bending term $K_3^R (\partial_z^2 u_a)^2 / 2$ is needed 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 $g_i(\mathbf{x})$, and the displacement $u_i(\mathbf{x})$. These are independent variables at the reference point \mathbf{x} that satisfy the continuum generalizations of the usual relations for the momentum and displacement of a particle: $\partial g_i(\mathbf{x}) / \partial g_j(\mathbf{x}') = \delta_{ij} \delta(\mathbf{x} - \mathbf{x}')$, $\partial u_i(\mathbf{x}) / \partial u_j(\mathbf{x}') = \delta_{ij} \delta(\mathbf{x} - \mathbf{x}')$, and $\partial g_i(\mathbf{x}) / \partial u_j(\mathbf{x}') = 0$. These relations yield a nonvanishing Poisson bracket between $u_i(\mathbf{x})$ and $g_j(\mathbf{x}')$:

$$\{u_i(\mathbf{x}), g_j(\mathbf{x}')\} = \delta_{ij} \delta(\mathbf{x} - \mathbf{x}'). \quad (3.12)$$

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

$$v_i \equiv \dot{u}_i = \frac{\delta \mathcal{H}_{\text{kin}}}{\delta g_i} = \frac{1}{\rho} g_i, \quad (3.13a)$$

$$\dot{g}_i = \frac{\delta \mathcal{H}_{\mathbf{u-el}}}{\delta u_i} + \eta_{ijkl} \partial_j \partial_l v_k, \quad (3.13b)$$

where \mathbf{v} is the velocity field and $\mathcal{H}_{\text{kin}} = \int d^3x / g_i g_i / (2\rho)$ is the coarse-grained kinetic energy. η_{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 $\mathcal{H}_{\mathbf{u}}$:

$$T\dot{S} = \int d^3x \left\{ \frac{\eta_1}{2} \dot{u}_{zz}^2 + \eta_2 \dot{u}_{zz} \dot{u}_{ii} + \frac{\eta_3}{2} \dot{u}_{ii}^2 + \eta_4 \dot{u}_{ab}^2 + \eta_5^R \dot{u}_{az}^2 \right\}, \quad (3.14)$$

where the η 's are low-frequency viscosities. η_5^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 $\mathcal{H}_{\mathbf{u-el}}$. The absence of any dissipative term proportional to $-\delta \mathcal{H} / \delta u_i$ in Eq. (3.13a) reflects the tethered or cross-linked character of the elastomer. In non-cross-linked systems, this equation would contain an additional dissipative term proportional to $-\delta \mathcal{H}_{\mathbf{u-el}} / \delta u_i$ describing permeation. Second, these equations are expressed in Lagrangian coordinates, and the derivatives $\dot{\mathbf{u}}$ and $\dot{\mathbf{g}}$ 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: $\mathbf{u}_E(\mathbf{r}, t) = \mathbf{u}(\mathbf{x}(\mathbf{r}), t)$ and similarly for \mathbf{g}_E . The time derivative $\dot{\mathbf{u}} = d\mathbf{u} / dt = \partial \mathbf{u}_E / \partial t + \mathbf{v} \cdot \nabla \mathbf{u}_E$ 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. (3.13a) allows us to replace \dot{g}_i in Eq. (3.13b) with $\rho \dot{u}_i$ to produce the standard mechanical equation for a solid with dissipation:

$$\rho \ddot{u}_a = (C_2 + \eta_2 \partial_t) \partial_a u_{zz} + (C_3 + \eta_3 \partial_t) \partial_a u_{ii} + 2(C_4 + \eta_4 \partial_t) \partial_b u_{ab} + (C_5^R + \eta_5^R \partial_t) \partial_z u_{az} - K_3^R \partial_z^2 u_a, \quad (3.15a)$$

$$\rho \ddot{u}_z = (C_1 + \eta_1 \partial_t + C_2 + \eta_2 \partial_t) \partial_z u_{zz} + (C_2 + \eta_2 \partial_t + C_3 + \eta_3 \partial_t) \partial_z u_{ii} + (C_5^R + \eta_5^R \partial_t) \partial_b u_{bz} - K_1^R \partial_z^2 u_z. \quad (3.15b)$$

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. (3.11a), has a dynamical generalization with a frequency-dependent $C_5^R(\omega) = C_5 - i\omega \eta_5^R$, 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 sec-

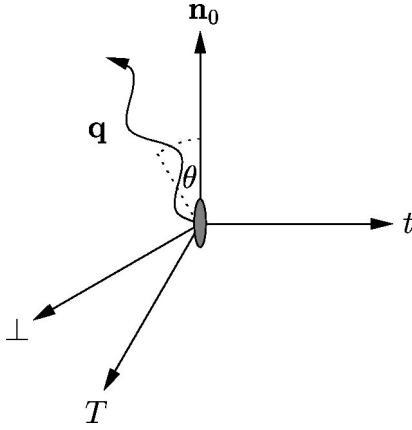


FIG. 1. Symmetry directions. The t direction is perpendicular to the plane containing the equilibrium director \mathbf{n}_0 and the wave vector \mathbf{q} of a generic excitation. The T direction is perpendicular to \mathbf{q} and the t direction.

tion, 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 NE's with $C_5^R=0$. When $C_5^R \neq 0$, the bending terms can be neglected, and the dynamical equations and associated modes are identical to those of uniaxial solids. We will return to case when C_5^R is nonzero in Sec. IV.

The sound modes have frequencies $\omega(\mathbf{q})=c(\theta)q$, where θ is the angle that \mathbf{q} makes with the z axis. The nondissipative 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 $\omega \sim -iq^2$. Thus, to obtain the true nondissipative sound-mode structure, we can ignore the bending terms. Consequentially, the nondissipative 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 $u_t = \varepsilon_{ab} q_a u_b / q_\perp$ perpendicular to both \mathbf{q} and the z axis and coupled displacements $u_\perp = q_a u_a / q_\perp$ and u_z in the plane that contains \mathbf{q} (see Fig. 1). In Fourier space, the hydrodynamic equations are

$$\rho \omega^2 u_t = C_4 q_\perp^2 u_t, \quad (3.16a)$$

$$\rho \omega^2 u_\perp = (C_1 + C_3) q_\perp q_z u_z + (C_3 + 2C_4) q_\perp^2 u_\perp, \quad (3.16b)$$

$$\rho \omega^2 u_z = (C_1 + 2C_2 + C_3) q_z^2 u_z + (C_2 + C_3) q_\perp q_z u_\perp. \quad (3.16c)$$

Thus, there is a transverse sound mode with velocity

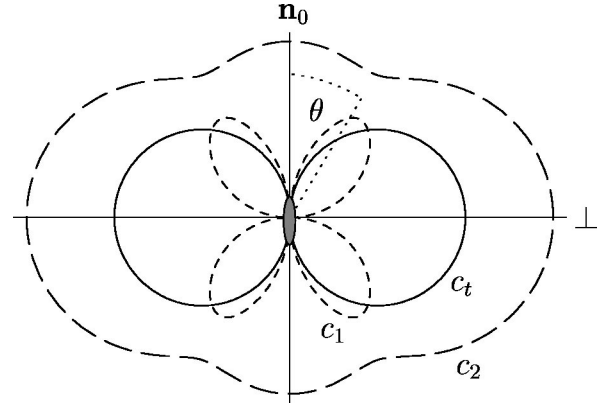


FIG. 2. Schematic polar plot (arbitrary units) of the sound velocities c_t (solid line), c_1 (short-dashed line), and c_2 (long-dashed line).

$$c_t(\theta) = \sqrt{\frac{C_4}{\rho}} |\sin \theta| \quad (3.17a)$$

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

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

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

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 $C_3 \rightarrow \infty$; its velocity is nonvanishing for all θ . The second mode is like that of a smectic-A liquid crystal [5,29] 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 nondissipative limit, we turn to a full analysis of modes in the incompressible limit. To discuss this limit, it is useful to decompose $\mathbf{u}(\mathbf{q})$ into a longitudinal part u_l along \mathbf{q} and components u_t , perpendicular to both \mathbf{q} and z , and u_T perpendicular to \mathbf{q} in the plane containing \mathbf{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

$$\rho \omega^2 u_t = (C_4 q_\perp^2 + K_3^R q_z^4) u_t - i\omega(\eta_4 q_\perp^2 + \eta_5^R q_z^2) u_t, \quad (3.18a)$$

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

These equations produce propagating modes with respective frequencies

$$\omega_{i,\pm} = \pm \sqrt{\frac{C_4}{\rho}} |q_\perp| - i \frac{2\eta_4 q_\perp^2 + \eta_5^R q_z^2}{4\rho}, \quad (3.19a)$$

$$\omega_{T,\pm} = \pm \sqrt{\frac{C_1 + 2C_4}{\rho}} \frac{|q_\perp q_z|}{q} - i \frac{2(\eta_1 + \eta_4) q_\perp^2 q_z^2 + \eta_5^R (q_\perp^2 - q_z^2)^2}{4\rho q^2}, \quad (3.19b)$$

as long as \mathbf{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

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

As in conventional nematics, $K_1 \sim K_3 \sim 10^{-6}$ dyn and $\rho \sim 1$ g/cm³. The viscosity should be larger than the 0.01 P 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

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

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

When $q_z = 0$, the t modes remain propagating soundmodes, but the T modes become diffusive with frequencies

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

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

$$\omega_{T,s} = -i \frac{2K_1^R}{\eta_5^R} q_\perp^2, \quad (3.23a)$$

$$\omega_{T,f} = -i \frac{\eta_5^R}{2\rho} q_\perp^2. \quad (3.23b)$$

Note that there is one pair of diffusive and one pair of propagating sound modes in the incompressible limit when $q_z = 0$. The attenuation (the imaginary part of \mathbf{q}) of the diffusive modes is proportional to $\sqrt{\omega}$ and that of the sound modes is proportional to ω . This is the explanation of the large difference in attenuation of the two polarizations of trans-

verse waves found by TW [22] that makes NE's candidates for acoustic polarizers. We will return to this issue in Sec. IV C.

Equations (3.21a) and (3.23a) 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 (3.21a) or, respectively, (3.23a). For $\theta \approx 0$, we get, for example, from Eqs. (3.19) and (3.21a) that the crossover is expected at an angle θ_0 such that

$$|\sin \theta_0| \approx \sqrt{\frac{\rho}{C_4} \frac{K_3^R}{\eta_5^R}} q. \quad (3.24)$$

The wave vectors in light scattering experiments on NE dynamics typically have a magnitude $q \sim 10^5$ cm⁻¹. The elastic moduli C_1 and C_4 should be comparable to the shear modulus of rubber, $C_1 \sim C_4 \sim 10^7$ dyn/cm². Hence we estimate

$$|\sin \theta_0| \approx 10^{-4}; \quad (3.25)$$

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

$$|\cos \theta_{\pi/2}| \approx 10^{-4}, \quad (3.26)$$

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 nemato-hydrodynamics [33,34] when elasticity due to network cross-linking is turned off. It must be emphasized, however, that our equations predict nonhydrodynamic modes characterized by a decay time τ that does not approach infinity with vanishing wave number q . In any real system, there are many nonhydrodynamic modes with characteristic decay times τ_α . For a dynamical theory to provide a correct description of a system over a frequency range from zero to some maximum frequency ω_M it must include contributions from all modes, both hydrodynamic and nonhydrodynamic, with characteris-

tic frequencies up to a few times ω_M . An isotropic rubber is characterized by rather long decay times τ_R of Rouse-like modes of chain segments [44]. At frequencies ω such that $\omega\tau_R \ll 1$, its viscosities become frequency independent and it is described by the hydrodynamical equations of an isotropic solid. However, when $\omega\tau_R \geq 1$, viscosities develop a non-trivial frequency dependence and hydrodynamics breaks down. In our theory for nematic elastomers, we assume that there is a single director relaxation time τ and that it is much larger than τ_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 hydrodynamic 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 $n_i(\mathbf{x})$ with $g_j(\mathbf{x}')$. This Poisson bracket is generally derived in Eulerian coordinates [34,45] 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

$$\{n_i(\mathbf{x}), g_j(\mathbf{x}')\} = -\lambda_{ijk} \partial_k \delta(\mathbf{x} - \mathbf{x}'). \quad (4.1)$$

The properties of the tensor λ_{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 λ_{ijk} must change sign with \mathbf{n} . And third, under rigid uniform rotations, the director has to obey $\dot{\mathbf{n}} = \frac{1}{2}(\nabla \times \dot{\mathbf{u}}) \times \mathbf{n}$. The only tensors and vectors available to use for the construction of λ_{ijk} are δ_{ij} , n_i , and the Levi-Civita antisymmetric tensor ϵ_{ijk} and the only tensors that satisfy the first two conditions above are $\delta_{ij}^T n_k$ and $\delta_{ik}^T n_j$ where $\delta_{ij}^T = \delta_{ij} - n_i n_j$. Thus the first two conditions imply that λ_{ijk} has two independent components, which can be expressed as parts symmetric and antisymmetric under interchange of j and k :

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

The third condition implies that the coefficient λ_2 of the antisymmetric part must be equal to -1 . There are no constraints on the value of λ , which is equal to the ratio of two dissipative coefficients of the Leslie-Eriksen theory [33]. When its absolute magnitude is positive, it determines the equilibrium tilt angle of the director under uniform shear [27].

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:

$$\dot{n}_i = \lambda_{ijk} \partial_j \dot{u}_k - \Gamma \frac{\delta \mathcal{H}}{\delta n_i}, \quad (4.3a)$$

$$\dot{u}_i = \frac{1}{\rho} g_i, \quad (4.3b)$$

$$\dot{g}_i = \lambda_{kji} \partial_j \frac{\delta \mathcal{H}}{\delta n_k} - \frac{\delta \mathcal{H}}{\delta u_i} + \nu_{ijkl} \partial_j \partial_k \dot{u}_k. \quad (4.3c)$$

The first terms on the respective right-hand sides of Eqs. (4.3a) and (4.3b) as well as the first and second terms on the right-hand side of Eq. (4.3c) stem from the Poisson brackets. \mathcal{H} is the full elastic energy of NE's as stated in Eq. (3.1). The second term on the right-hand side of Eq. (4.3a) is a dissipative term that describes diffusive relaxation. There is no dissipative contribution to Eq. (4.3b) because NE's are tethered. ν_{ijkl} is a viscosity tensor that has the same structure as η_{ijkl} . It has five independent components ν_1 to ν_5 . We use different symbols here for the viscosities than we used in Sec. III so that we can cleanly keep track of differences between the two theories.

We have assumed in Eqs. (4.3) that both Γ and ν_{ijkl} are local in time or, equivalently, that their temporal Fourier transforms are independent of frequency as they are at frequencies ω less than their respective inverse characteristic times τ_Γ^{-1} and τ_R^{-1} . At larger frequencies, however, both Γ and ν_{ijkl} do depend on frequency. In polymer gels, storage and loss moduli are proportional to $\sqrt{\omega}$ at frequencies $\omega\tau_R \geq 1$ because of the large number of closely spaced modes which in turn leads to viscosities proportional to $1/\sqrt{\omega}$ 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 $\omega\tau_\Gamma \geq 1$. There is also no reason why τ_Γ and τ_R should not be equal or nearly so. If the decay time τ for the nonhydrodynamic director modes predicted by the theory with the low-frequency approximations to Γ and ν_{ijkl} is greater than τ_R and τ_Γ , then this theory provides a correct description of the dynamics for frequencies $0 < \omega < \min(\tau_R^{-1}, \tau_\Gamma^{-1})$ including $\omega \sim \tau^{-1}$. If, on the other hand, $\tau < \min(\tau_R, \tau_\Gamma)$, the full frequency dependence of Γ must be used for frequencies $\omega > \min(\tau_R^{-1}, \tau_\Gamma^{-1})$. In all cases, however, the hydrodynamic theory of the preceding section is regained in the limit $\omega \ll \min(\tau^{-1}, \tau_R^{-1}, \tau_\Gamma^{-1})$. To keep our discussion simple, we will continue to assume that both Γ and ν_{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 the elastic constants in \mathcal{H} , except for the Frank constants K_1, K_2 , and K_3 , equal to zero. Thus we are guaranteed to obtain the well-known modes of nematic liquid crystals in this limit. One can also make the observation that in this limit Eqs.

(4.3) depend just on $\mathbf{v}=\mathbf{u}$ rather than on \mathbf{v} and \mathbf{u} . If one seeks modes in terms of \mathbf{u} rather than \mathbf{v} , one finds extra zero-frequency modes that are spurious.

Next, we condense the equations of motion into a reduced set of effective equations. We can simplify the subsequent analysis at the onset by eliminating either \mathbf{u} or \mathbf{g} with help of Eq. (4.3b). To facilitate in contact with the work of TW we choose to keep \mathbf{u} . 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. (3.8). Collecting, we obtain after some algebra the following effective equations of motion for \mathbf{Q} and \mathbf{u} :

$$\{[\partial_t + \Gamma D_1] \delta_{ab} - \Gamma M_{ab}(\nabla)\} Q_b = [\lambda \partial_t - \Gamma D_2] u_{az} + \Gamma M_{ab}(\nabla) \tilde{\Omega}_b \quad (4.4a)$$

$$\begin{aligned} \rho \partial_t^2 u_a = & -\frac{\lambda+1}{2\Gamma} \partial_t \partial_z [Q_a - \lambda u_{az}] + [C_2 + \nu_2 \partial_t] \partial_a u_{zz} \\ & + [C_3 + \nu_3 \partial_t] \partial_a u_{ii} + 2[C_4 + \nu_4 \partial_t] \partial_b u_{ab} \\ & + \left[C_5 + \nu_5 \partial_t - \frac{D_2}{2} \right] \partial_z u_{az} + \frac{D_2 - D_1}{2} \partial_z Q_a, \end{aligned} \quad (4.4b)$$

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

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

$$Q_a = -\frac{D_2}{D_1} \frac{1 - i\omega\tau_2}{1 - i\omega\tau_1} u_{az}. \quad (4.5)$$

In writing Eq. (4.5) we have used the relaxation times $\tau_1 = 1/(\Gamma D_1)$ and $\tau_2 = -\lambda/(\Gamma D_2)$. Here $\tau_1 \approx 10^{-2}$ s [1,41,42] 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, λ is given by $\lambda = -\tau_2 D_2 / (\tau_1 D_1)$. Note that λ is usually negative, but if it is positive, there are inconsistencies arising from a negative τ_2 .

Inserting expression (4.5) into the timewise Fourier-transformed equations of motion for u_a and u_z , we obtain

$$\begin{aligned} -\rho \omega^2 u_a = & C_2(\omega) \partial_a u_{zz} + C_3(\omega) \partial_a u_{ii} + 2C_4(\omega) \partial_b u_{ab} \\ & + C_5^R(\omega) \partial_z u_{az}, \end{aligned} \quad (4.6a)$$

$$\begin{aligned} -\rho \omega^2 u_z = & [C_1(\omega) + C_2(\omega)] \partial_z u_{zz} + [C_2(\omega) + C_3(\omega)] \partial_z u_{ii} \\ & + C_5^R(\omega) \partial_b u_{bz}, \end{aligned} \quad (4.6b)$$

with $C_1(\omega) = C_1 - i\omega\nu_1$, $C_2(\omega) = C_2 - i\omega\nu_2$, etc., and

$$\begin{aligned} C_5^R(\omega) = & C_5 - i\omega \left(\nu_5 + \frac{\lambda^2}{2\Gamma} \right) - \frac{D_2^2 (1 - i\omega\tau_2)^2}{2D_1 (1 - i\omega\tau_1)} \\ = & C_5^R - i\omega\nu_5^R + O(\omega^2), \end{aligned} \quad (4.7)$$

where

$$\nu_5^R = \nu_5 + \frac{\lambda^2}{2\Gamma} \left(1 - \frac{\tau_1}{\tau_2} \right)^2. \quad (4.8)$$

Note that Eqs. (4.6) are identical in form to Eq. (3.15) for uniaxial solids with $K_1^R = K_3^R = 0$. 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 η_5^R and ν_5^R . As pointed out earlier, Rouse modes will be important for $\omega\tau_R \gg 1$ and $\omega\tau_\Gamma \gg 1$, leading to a frequency dependence of the viscosities. In this regime we have to let $\nu_i \rightarrow \nu_i(\omega)$ and $\Gamma \rightarrow \Gamma(\omega)$. For $\omega\tau_R \gg 1$ and $\omega\tau_\Gamma \gg 1$, in particular, the viscosities are proportional to $1/\sqrt{\omega}$ [44].

Before we move on we point out that Eqs. (4.4) 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. t direction

By applying $\varepsilon_{ba} q_b$ (including the summation over a) to both sides of the Fourier-transformed version of Eq. (4.4a) we obtain a diagonalized equation of motion for Q_t that is readily solved with the result

$$Q_t = -\frac{D_2}{D_1} \frac{1 - i\omega\tau_2 + q^2 K_t / D_2}{1 - i\omega\tau_1 + q^2 K_t / D_1} \frac{1}{2} i q_z u_t, \quad (4.9)$$

where we have used the abbreviated notation

$$q^2 K_t = K_2 q_\perp^2 + K_3 q_z^2. \quad (4.10)$$

Application of the same procedure to Eq. (4.4b) yields

$$\begin{aligned} -\omega^2 \rho u_t = & \frac{\lambda+1}{2\Gamma} i\omega q_z \left[iQ_t + \frac{\lambda}{2} q_z u_t \right] - C_4(\omega) q_\perp^2 u_t \\ & + \frac{D_2 - D_1}{2} i q_z Q_t - \frac{1}{2} \left[C_5(\omega) - \frac{D_2}{2} \right] q_z^2 u_t. \end{aligned} \quad (4.11)$$

u_t is a purely transverse displacement that does not couple to any displacement in the plane containing \mathbf{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

$$\left[\omega^2 \rho - C_4(\omega) q_\perp^2 + \frac{\lambda(\lambda+1)i\omega - \lambda\tau_2^{-1} - 2\Gamma C_5(\omega)}{4\Gamma} q_z^2 \right] \times D_1 [1 - i\omega\tau_1 + q^2 K_t / D_1] + \frac{\lambda+1 - \tau_1^{-1} - \lambda\tau_2^{-1}}{4\Gamma} \times q_z^2 D_2 [1 - i\omega\tau_2 + q^2 K_t / D_2] = 0. \quad (4.12)$$

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

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:

$$\omega_{t,s} = -i [K_2 q_\perp^2 + K_3 q_z^2] \left\{ \Gamma + \frac{(1+\lambda)q_z^2}{2(\nu_5 q_z^2 + 2\nu_4 q_\perp^2)} \right\}, \quad (4.13a)$$

$$\omega_{t,f} = -i \frac{2\nu_4 q_\perp^2 + \nu_5 q_z^2}{2\rho}, \quad (4.13b)$$

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 ν stands ambiguously for all the viscosities. In comparing Eqs. (4.13) and (4.19) 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), $\Gamma \rightarrow \gamma^{-1}$, $\nu_1 \rightarrow 2\nu_1$, $\nu_4 \rightarrow \nu_2$, $\nu_5 \rightarrow 2\nu_3$, $q_\perp \rightarrow q_1$, and $q_z \rightarrow q_3$.

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

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

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

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

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 NE's 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. T direction

Except for the t direction, an 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 , ν_2 , and ν_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_a (including the summation over a) to the Fourier-transformed counterparts of Eqs. (4.4a) and (4.4b), we obtain

$$Q_\perp = - \frac{D_2}{D_1} \frac{1 - i\omega\tau_2 + q^2 K_\perp / D_2}{1 - i\omega\tau_1 + q^2 K_\perp / D_1} \frac{1}{2} i q_\perp u_z - \frac{D_2}{D_1} \frac{1 - i\omega\tau_2 - q^2 K_\perp / D_2}{1 - i\omega\tau_1 - q^2 K_\perp / D_1} \frac{1}{2} i q_z u_\perp, \quad (4.16a)$$

$$- \omega^2 \rho u_\perp = \frac{\lambda+1}{2\Gamma} i \omega q_z \left[i Q_\perp + \frac{\lambda}{2} (q_\perp u_z + q_z u_\perp) \right] - 2C_4(\omega) q_\perp^2 u_\perp + \frac{D_2 - D_1}{2} i q_z Q_\perp - \frac{1}{2} \left[C_5(\omega) - \frac{D_2}{2} \right] q_z (q_\perp u_z + q_z u_\perp), \quad (4.16b)$$

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. (4.16) we also need the Fourier-transformed version of Eq. (4.4c) in the incompressible limit:

$$- \omega^2 \rho u_z = \frac{\lambda-1}{2\Gamma} i \omega q_\perp \left[i Q_\perp + \frac{\lambda}{2} (q_\perp u_z + q_z u_\perp) \right] - C_1(\omega) q_z^2 u_z + \frac{D_1 + D_2}{2} i q_\perp Q_\perp - \frac{1}{2} \left[C_5(\omega) + \frac{D_2}{2} \right] q_\perp (q_\perp u_z + q_z u_\perp). \quad (4.18)$$

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 $u_\perp = -q_z u_T / q$ 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:

$$\omega_{T,s} = -i[K_1 q_\perp^2 + K_3 q_z^2] \times \left\{ \Gamma + \frac{[q^2 - \lambda(q_\perp^2 - q_z^2)]^2}{2\nu_5(q_\perp^2 - q_z^2)^2 + 4(\nu_1 + 2\nu_4)q_\perp^2 q_z^2} \right\}, \quad (4.19a)$$

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

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

By perturbatively solving the full secular equation we extract the three T direction modes for NE's. We find one massive mode

$$\omega_{T,m} = -\tau_1^{-1} - i\Gamma[K_1 q_\perp^2 + K_3]q_z^2 + i \frac{\nu_5^R - \nu_5}{2\rho} \frac{(q_\perp^2 - q_z^2)^2}{q^2} \quad (4.20)$$

and two soft modes. For nonvanishing sound velocities the soft modes have frequencies

$$\omega_{T,\pm} = \pm \sqrt{\frac{2(C_1 + 2C_4)q_\perp^2 q_z^2 + C_5^R(q_\perp^2 - q_z^2)^2}{2\rho q^2}} - i \frac{2(\nu_1 + 2\nu_4)q_\perp^2 q_z^2 + \nu_5^R(q_\perp^2 - q_z^2)^2}{4\rho q^2}. \quad (4.21)$$

When $C_5^R=0$ this result reduces, provided that the viscosities are properly identified, to Eq. (3.19b). 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 final results for the modes to those found by TW. Of course, we must take into account differences in conventions and notations in these comparisons. For guidance in identifying the 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 σ_{ij} are the components of the stress tensor $\underline{\underline{\sigma}}$. The specifics of the σ_{ij} are easily gathered from Eqs. (4.4b) and (4.4c):

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

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

TABLE I. Correspondence between quantities used by TW and quantities used in our work.

TW	This paper	TW	This paper
$2C_1$	C_1	$2A_4$	ν_4
$2C_2$	C_2	$4A_5$	$\nu_5 + \lambda^2/(2\Gamma)$
$2C_3$	C_3	γ_1	$1/\Gamma$
$2C_4$	C_4	γ_2	$-\lambda/\Gamma$
$4C_5$	C_4	ω	$-\omega$
$2A_1$	ν_1	θ_1	Q_2
$2A_2$	ν_2	θ_2	$-Q_1$
$2A_3$	ν_3	$C_5^R(\omega)$	$\hat{C}_5^R(\omega)$

$$\sigma_{az} = \left[C_5 + \nu_5 \partial_t - \frac{D_2}{2} + \frac{\lambda(\lambda+1)}{2\Gamma} \partial_t \right] u_{az} + \frac{1}{2} \left[D_2 - D_1 - \frac{\lambda+1}{\Gamma} \partial_t \right] Q_a, \quad (4.23c)$$

$$\sigma_{za} = \left[C_5 + \nu_5 \partial_t + \frac{D_2}{2} + \frac{\lambda(\lambda-1)}{2\Gamma} \partial_t \right] u_{az} + \frac{1}{2} \left[D_2 + D_1 - \frac{\lambda-1}{\Gamma} \partial_t \right] Q_a, \quad (4.23d)$$

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

$$\sigma_{az} - \sigma_{za} = \frac{1}{\Gamma} \{ [\tau_1^{-1} + \partial_t] Q_a - \lambda [\tau_2^{-1} + \partial_t] u_{az} \} = M_{ab}(\nabla) \hat{\Omega}_b, \quad (4.24)$$

where we have used Eq. (4.4a) to obtain the last equality. Equation (4.24) reveals that the stress tensor σ_{ij} defined in Eqs. (4.23) 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 σ_{ij}^S yielding the same equations of motion as σ_{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 can easily be checked, Eq. (4.4a) becomes identical to the balance of torques equation under these circumstances. Therefore, our equations of motion (4.4a), (4.4b), and (4.4c) 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 $\nu_1 \geq 0$, $\nu_4 \geq 0$, and $\nu_5 \geq 0$. While the first two conditions can be readily identified with the conditions $A_1 \geq 0$ and $A_4 \geq 0$ in the work of TW, the situation is less obvious for the last condition. Note, however, that we can reexpress $\nu_5 \geq 0$ in terms of the quantities used by TW as

$$\nu_5 = 4A_5 - \frac{\gamma_2^2}{2\gamma_1} \geq 0, \quad (4.25)$$

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

$$\tau_1 \tau_R \geq \tau_2^2 \quad (4.26)$$

for ideally soft NE's.

Next, we come to the comparison of the results for the modes. Our first observation is here that the massive modes $\omega_{t,m}$ and $\omega_{T,m}$ are unaccounted for by TW. Note that the soft modes in the t and T directions are referred to in TW as qSH waves and qSV waves, respectively.

Let us first compare the findings regarding the t direction. To foster this comparison we recast our result (4.15) as

$$\omega^2 = \frac{1}{2\rho} [2C_4(\omega)q_\perp^2 + C_5^R(\omega)q_z^2]. \quad (4.27)$$

To the order we are working—i.e., to order $O(q^3)$ —solutions to Eq. (4.27) and our $\omega_{t,m}$ 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 with TW we eliminate the viscosities in Eq. (4.27) in favor of the relaxation time $\tau_R \approx \nu_1/C_1 \approx \dots \approx \nu_5/C_5 \approx [\nu_5 + \lambda^2/(2\Gamma)]/C_5$. The subscript R indicates that $\tau_R \approx 10^{-5}$ – 10^{-6} s is of the order of the Rouse time of the polymers constituting the rubbery matrix. We obtain

$$\omega^2 = \frac{1}{2\rho} [2C_4q_\perp^2 + \hat{C}_5^R(\omega)q_z^2](1 - i\omega\tau_R), \quad (4.28)$$

where

$$\hat{C}_5^R(\omega) = \frac{C_5^R(\omega)}{1 - i\omega\tau_R} = C_5 - \frac{D_2^2}{2D_1} \frac{(1 - i\omega\tau_2)^2}{(1 - i\omega\tau_1)(1 - i\omega\tau_R)}. \quad (4.29)$$

Taking into account Table I, we see that our $\hat{C}_5^R(\omega)$ is identical with the renormalized form of C_5 found by TW and that Eq. (4.29) is in full agreement with the dispersion relation of TW for the qSH waves.

Now to the T direction. To order $O(q^3)$ our results for $\omega_{T,\pm}$ coincide with the solutions of

$$\omega^2 = \frac{1}{2\rho} \left[[C_1(\omega) + 2C_4(\omega)] \frac{q_\perp^2 q_z^2}{q^2} + C_5^R(\omega) \frac{(q_\perp^2 - q_z^2)^2}{q^2} \right]. \quad (4.30)$$

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

$$\omega^2 = \frac{1}{2\rho} \hat{C}_5^R(\omega) q_\perp^2 (1 - i\omega\tau_R). \quad (4.31)$$

Using Table I, we 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 NE's that makes them, as pointed out by TW, candidates for acoustic polarizers—viz., the large difference in attenuation between the t and 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 ω 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, $\nu_5^R = \eta_5^R$, the fast diffusive mode in Eq. (3.23b). If the Frank energy is taken into account, the $\omega=0$ mode becomes the slow diffusive mode of Eq. (3.23a). 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 semisoft 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 wave numbers 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 nonhydrodynamic modes, for both director and displacement and verified that they reduced to those derived by Terentjev, Warner, and co-workers [1,22] 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 τ_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 wave number have

reported frequency-dependent storage and loss moduli that are in agreement with the predictions of the semisoft 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 NE's 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.

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