Dynamics, dynamic soft elasticity, and rheology of smectic-*C* elastomers

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We present a theory for the low-frequency, long-wavelength dynamics of soft smectic-*C* elastomers with locked-in smectic layers. Our theory, which goes beyond pure hydrodynamics, predicts a dynamic soft elasticity of these elastomers and allows us to calculate the storage and loss moduli relevant for rheology experiments as well as the mode structure.

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Smectic elastomers [1] are rubbery materials that have the macroscopic symmetry properties of smectic liquid crystals [2]. They are sure to have intriguing properties, some of which have already been studied experimentally and/or theoretically [1]. Very recently, seminal progress has been made on smectic-C (SmC) elastomers forming spontaneously from a smectic-A (SmA) phase upon cooling. Hiraoka et al. [3] produced a monodomain sample of such a material and carried out experiments demonstrating its spontaneous and reversible deformation in a heating and cooling process. Also very recently, it was discovered theoretically that such a material exhibits the fascinating phenomenon of soft elasticity [4], i.e., certain elastic moduli vanish as a consequence of the spontaneous symmetry breaking wherefore strains along specific symmetry direction cost no elastic energy and thus cause no restoring forces.

On one hand, due to the aforementioned experimental advances, dynamical experiments on soft SmC elastomers, such as rheology experiments of storage and loss moduli or Brillouin scattering measurements of sound velocities, seem within reach. On the other hand, there exists, to our knowledge, no dynamical theory that could be helpful in interpreting these kinds of experiments. Here we present a theory for the low-frequency, long-wavelength dynamics of soft SmC elastomers with locked-in layers that goes beyond pure hydrodynamics. As in standard elastic media and nematic elastomers [5] a purely hydrodynamical theory of SmC elastomers involves only a displacement field **u** and not the Frank director **n**, which relaxes to the local strain in a nonhydrodynamic time τ_n . We go beyond hydrodynamics, by including **n** in our theory, because dynamical experiments, like rheology measurements, typically probe a wide range of frequencies that extends from hydrodynamic regime to frequencies well above it.

Smectic elastomers are, like any elastomers, permanently cross-linked amorphous solids whose static elasticity is most easily described in Lagrangian coordinates in which \mathbf{x} labels a mass point in the undeformed (reference) material and $\mathbf{R}(\mathbf{x})=\mathbf{x}+\mathbf{x}(\mathbf{x})$, where $\mathbf{u}(\mathbf{x})$ is the displacement variable, labels the position of the mass point \mathbf{x} in the deformed (target) material. Lagrangian elastic energies are formulated in terms of the strain tensor \underline{u} which, in its linearized form, has the

components $u_{ij} = (1/2)(\eta_{ij} + \eta_{ji})$, where $\eta_{ij} = \partial_j u_i$ are the components of the displacement gradient tensor η .

The elastic energy density f of the Sm \overline{C} elastomers of interest here can be divided into two parts [4],

$$f = f_{\mathbf{u}} + f_{\mathbf{u},\mathbf{n}},\tag{1}$$

where $f_{\mathbf{u}}$ depends only on \underline{u} and $f_{\mathbf{u},\mathbf{n}}$ describes the dependence of f on the Frank director \mathbf{n} including its coupling to the displacement variable. In the following we choose the coordinate system so that the z axis is parallel to the director of the initial SmA phase and the x axis is parallel to the director of tilt in the resulting SmC phase so that the equilibrium director characterizing the undeformed SmC phase is of the form $\mathbf{n}^0 = (c, 0, \sqrt{1-c^2})$ with c being the order parameter of the transition. With these conventions, $f_{\mathbf{u}}$ can be written in the same form as the elastic energy density of conventional monoclinic solids [6],

$$f_{\mathbf{u}} = \frac{1}{2}C_{xyxy}u_{xy}^{2} + C_{xyzy}u_{xy}u_{zy} + \frac{1}{2}C_{zyzy}u_{zy}^{2} + \frac{1}{2}C_{zzzz}u_{zz}^{2} + \frac{1}{2}C_{xxxx}u_{xx}^{2} + \frac{1}{2}C_{yyyy}u_{yy}^{2} + \frac{1}{2}C_{xzxz}u_{xz}^{2} + C_{zzxx}u_{zz}u_{xx} + C_{zzyy}u_{zz}u_{yy} + C_{xxyy}u_{xx}u_{yy} + C_{xxxz}u_{xx}u_{xz} + C_{yyxz}u_{yy}u_{xz} + C_{zzyz}u_{zz}u_{yz},$$
(2)

but with constraints relating the line three elastic constants. These latter constants can be expressed in terms of an overall elastic constant \overline{C} and an angle θ , which depends on the order parameter c, as $C_{xyxy} = \overline{C} \cos^2 \theta$, $C_{xyzy} = \overline{C} \cos \theta \sin \theta$, and $C_{zyzy} = \overline{C} \sin^2 \theta$. Neglecting contributions from the Frank energy, $f_{u,n}$ can be stated as

$$f_{\mathbf{u},\mathbf{n}} = \frac{1}{2} \Delta [Q_y + \alpha u_{xy} + \beta u_{yx}]^2, \qquad (3)$$

where Δ is a coupling constant and where α and β are dimensionless parameters. The variable Q_y stands for $Q_y = n_y - c \eta_{Ayx} - \sqrt{1 - c^2} \eta_{Ayz}$, where η_{Ayx} and η_{Ayz} are components of the antisymmetric part of η .

Equations (1)–(3) imply that SmC elastomers are soft under static conditions. Imagine that Q_y has relaxed locally to $Q_y = -\alpha u_{xy} - \beta u_{yx}$ so that f is effectively reduced to f_u . Then, due to the above relations among the elastic constants, defor-

mations characterized by Fourier transformed displacements $\mathbf{u} \| \hat{e}_y$ and wave vectors $\mathbf{q} \| \hat{e}_2 = (-\sin \theta, 0, \cos \theta)$ or, alternatively, $\mathbf{u} \| \hat{e}_2$ and $\mathbf{q} \| \hat{e}_y$ cost no elastic energy and hence cause no restoring forces. The effects of soft elasticity are more evident in a coordinate systems rotated though θ about the *y* axis in which $\hat{e}_{x'} = (\cos \theta, 0, \sin \theta)$, $\hat{e}_{y'} = \hat{e}_y$, and $\hat{e}_{z'} = \hat{e}_2$. In this system, $C_{x'y'z'y'}$ and $C_{z'y'z'y'}$ vanish, impyling there is no energy cost for shears $u_{z'y'}$ in the z'y' plane. If the elastomer is cross-linked in the SmC phase, these moduli become nonzero. Here, we will not consider such semisoft SmC elastomers.

Now, let us formulate our dynamical theory. Dynamical equations for \mathbf{u} and \mathbf{n} can be derived using standard Poisson-bracket approaches [7], with the result [5]

$$\dot{n}_i = \lambda_{ijk} \partial_j \dot{u}_k - \Gamma \frac{\delta \mathcal{H}}{\delta n_i}, \qquad (4a)$$

$$\rho \ddot{u}_{i} = \lambda_{kji} \partial_{j} \frac{\delta \mathcal{H}}{\delta n_{k}} - \frac{\delta \mathcal{H}}{\delta u_{i}} + \nu_{ijkl} \partial_{j} \partial_{l} \dot{u}_{k}, \qquad (4b)$$

where \mathcal{H} is the elastic energy of the system, ν_{ijkl} is the viscosity tensor, Γ is a dissipative coefficient with dimensions of an inverse viscosity, and

$$\lambda_{ijk} = \frac{1}{2}\lambda(\delta_{ij}^T n_k + \delta_{ik}^T n_k) + \frac{1}{2}(\delta_{ij}^T n_k - \delta_{ik}^T n_k),$$
(5)

where $\delta_{ij}^T = \delta_{ij} - n_i n_j$. As they stand, Eqs. (4a) and (4b) are valid for any liquid crystal elastomer with a defined director, such as, e.g., nematic, SmA and SmC elastomers. To describe SmC elastomers we have have to specify \mathcal{H} and v_{ijkl} accordingly. From the above it is clear that $\mathcal{H} = \int d^3x f$ with f as given in Eq. (1). The viscosity tensor entering here is that of a monoclinic system. It has 13 independent components and it can be parametrized, as we do, so that the entropy production density $T\dot{s}$ takes on the same form as Eq. (2) with the elastic constants C_{ijkl} replaced by viscosities v_{ijkl} and with u_{ij} replaced by \dot{u}_{ij} . The v_{ijkl} depend on the order parameter c. v_{xxxz} , v_{yyxz} , and v_{zxxz} vanish at the SmC to SmA transition and are therefore expected to be smaller than the remaining viscosities for c small [8].

A smectic elastomer is characterized, in general, by relaxation times associated with director relaxation and with other modes, which we will simply refer to as elastomer modes. For frequencies ω such that $\omega \tau_E \ll 1$, where τ_E is the longest elastomer time, the viscosities and Γ are practically frequency independent. When $\omega \tau_E \ge 1$, however, the viscosities ν_{ijkl} and Γ develop a nontrivial frequency dependence. In the following we will consider in detail only the case $\tau_n \ge \tau_E$ and $\omega \tau_E \ll 1$.

As mentioned above, Eq. (3) omits contributions from the Frank elastic energy for director distortions, which are of a higher order in derivatives than those arising from network elasticity. Without the Frank energy, our dynamical theory misses diffusive modes along certain symmetry directions where sound velocities vanish. Including the Frank energy, on the other hand, makes the equations of motion considerably more complicated. To keep our presentation as simple as possible, we will therefore, for the most part, exclude the Frank energy. When it comes to stating results for the afore-

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mentioned diffusive modes, however, we will include Frank contributions in order to present complete results.

From Eq. (4a) we can derive an equation of motion for Q_{y} . In frequency space, this equation can be written as

$$Q_y = -\alpha \frac{1 + i\omega\tau_3}{1 - i\omega\tau_1} u_{yx} - \beta \frac{1 + i\omega\tau_2}{1 - i\omega\tau_1} u_{yz},\tag{6}$$

where we have introduced the relaxation times $\tau_1 = 1/(\Gamma\Delta)$, $\tau_2 = \lambda \sqrt{1 - c^2}/(\Gamma\Delta\beta)$, $\tau_3 = \lambda c/(\Gamma\Delta\alpha)$. As we will see further below, our dynamical equations predict nonhydrodynamic modes with a decay time ("mass") τ_1 , which implies $\tau_1 = \tau_n$.

With help of Eqs. (6) and (4b) we derive effective equations of motions in terms of the displacements only, which can be cast as

$$\rho\omega^2 u_i = -\partial_i \sigma_{ii}(\omega), \tag{7}$$

with a symmetric stress tensor σ given by

$$\sigma_{\mu}(\omega) = C_{\mu\nu}(\omega)u_{\nu} + C_{\mu xz}(\omega)u_{xz}, \qquad (8a)$$

$$\sigma_{xz}(\omega) = \frac{1}{2}C_{xz\nu}(\omega)u_{\nu} + \frac{1}{2}C_{xzxz}(\omega)u_{xz},$$
(8b)

$$\sigma_{\xi}(\omega) = \frac{1}{2} C^{R}_{\xi\chi}(\omega) u_{\chi}, \qquad (8c)$$

where we use a compact notation with indices μ , ν running over *xx*, *yy*, and *zz*, and indices ξ , χ running over *xy* and *zy*. $C_{ijkl}(\omega)$ with no superscript *R* stands for $C_{ijkl}(\omega)$ $=C_{ijkl}-i\omega\nu_{ijkl}$. The superscript *R* indicates that certain elastic moduli are renormalized by the relaxation of the director. These are

$$C_{\xi\chi}^{R}(\omega) = C_{\xi\chi} - i\omega\nu_{\xi\chi} - \frac{i\omega\tau_{1}}{1 - i\omega\tau_{1}}\Delta A_{\xi\chi} = C_{\xi\chi} - i\omega\nu_{\xi\chi}^{R} + O(\omega^{2}),$$
(9)

with renormalized viscosities $\nu_{\xi\chi}^R = \nu_{\xi\chi} + \Gamma^{-1}A_{\xi\chi}$, and where $A_{xyxy} = \alpha^2 (1 + \tau_3/\tau_1)^2$, $A_{xyzy} = \alpha\beta(1 + \tau_3/\tau_1)(1 + \tau_2/\tau_1)$, and $A_{zyzy} = \beta^2 (1 + \tau_2/\tau_1)^2$.

The frequency dependence of the elastic moduli in Eq. (8) can be determined, in principle, by rheology measurements of the corresponding storage and loss moduli. The unrenormalized moduli $C_{ijkl}(\omega)$ lead to conventional storage and loss moduli $C'_{ijkl}=C_{ijkl}$ and $C''_{ijkl}=\omega\nu_{ijkl}$ that are, as in conventional rubbers, respectively, constant and proportional to ω at low frequencies. The renormalized moduli, on the other hand, have the potential for much more interesting rheology behavior. One consequence of Eq. (9) is that SmC elastomers could exhibit so-called dynamic soft elasticity [9]. To highlight this phenomenon, let us switch briefly to the rotated reference space coordinates x', y', z'. Then, Eq. (9) implies that

$$C^{R}_{y'z'y'z'}(\omega) = -i\omega\nu_{y'z'y'z'} - \frac{i\omega\tau_{1}}{1 - i\omega\tau_{1}}\Delta A_{y'z'y'z'}, \quad (10)$$

with $v_{y'z'y'z'} = \sin^2(\theta) v_{xyxy} - \sin(2\theta) v_{xyyz} + \cos^2(\theta) v_{yzyz}$ and an analogous expression for $A_{y'z'y'z'}$. $C^R_{x'y'y'z'}(\omega)$ is of the same form as Eq. (10). Thus, $C^R_{y'z'y'z'}(\omega)$ and $C^R_{x'y'y'z'}(\omega)$ vanish in the limit $\omega \to 0$ where we recover true soft elastic-



FIG. 1. Log-log plot of the reduced storage and loss moduli $C'_{\xi\chi}(\omega)/C_{\xi\chi}$ (solid line) and $C''_{\xi\chi}(\omega)/C_{\xi\chi}$ (dashed line) versus the reduced frequency $\omega\tau_n$ as given, respectively, by the real and negative imaginary parts of Eq. (9) for $\tau_n \gg \tau_E$. For the purpose of illustration we have set, by and large arbitrarily, $\nu_{\xi\chi}/(\tau_n C_{\xi\chi})=1$ and $\Delta A_{\xi\chi}/C_{\xi\chi}=10^2$.

ity. At nonvanishing frequency the system cannot be ideally soft but it can be nearly so for ω small. This type of behavior was first predicted for nematic elastomers, where it has been termed dynamic soft elasticity [9]. The storage moduli for u_{xy} and u_{yz} strains, Eq. (9), in the original coordinate system are nonzero at zero frequency. Their behavior for $\omega > 0$, like that of semisoft nematic elastomers [5,9], depends on τ_n/τ_E . If $\tau_n \gg \tau_E$, the storage moduli exhibit a step and the corresponding loss moduli an associated peak at $\omega \tau_n \sim 1$ as shown in Fig. 1; if $\tau_n \approx \tau_E$, or $\tau_n < \tau_E$, this is not the case. The storage moduli $C'_{y'z'y'z'}(\omega)$ and $C'_{x'y'y'z'}(\omega)$ in the rotated frame are zero at zero frequency. In a semisoft SmC, however, they will exhibit behavior similar to that of Fig. 1 for $\tau_n \gg \tau_F$. In nematic elastomers, there is still some controversy [10] about whether the $\tau_n \gg \tau_E$ regime has actually been observed in experiments. It would be interesting to see if it might exist in SmC elastomers, where τ_E might be shorter than it is in nematics because of the smectic layers.

To assess the mode structure of SmC elastomers, we start with an analysis of propagating sound modes in the dissipationless limit. The sound modes have frequencies $\omega(\mathbf{q}) = C(\vartheta, \varphi)q$, where $q = |\mathbf{q}|$ and where ϑ and φ are the azimuthal and polar angles of \mathbf{q} in spherical coordinates. Their sound velocities $C(\vartheta, \varphi)$, as calculated from Eq. (7) with the viscosities v_{iikl} set to zero, are depicted in Fig. 2. There are three pairs of sound modes. One of these pairs (i) is associated with the soft deformations discussed above. Its velocity vanishes for **q** along \hat{e}_{v} and \hat{e}_{2} so that when viewed in the y'z' plane it has a clover-leaf-like shape. The remaining two pairs are associated with nonsoft deformations. In the incompressible limit, these pairs become purely transverse (ii) and longitudinal (iii), respectively. In the y'z' and x'z' planes, their velocities are nonvanishing in all directions. Note that, since the velocity of pair (i) vanishes in directions where the velocities of the other modes remain finite, SmC elastomers are, like nematic elasomers [9], potential candidates for applications in acoustic polarizers.

Having found the general sound-mode structure in the nondissipative limit, we now turn to the full mode structure in the incompressible limit. In the softness-related symmetry directions the modes are of the following types: (i) nonhydrodynamic modes with frequencies

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FIG. 2. (Color online) Schematic plots of sound velocities: (a) spherical plot of mode pair (i) only; (b) and (c) polar plots in the y'z' and x'z' planes, respectively, of all three sound-mode pairs.

$$\omega_m = -i\tau_1^{-1} + iD_m q^2, \tag{11}$$

with a zero-q decay time τ_1 and diffusion constants D_m , (ii) propagating modes with frequencies

$$\omega_p = \pm Cq - iD_p q^2, \tag{12}$$

with sound velocities C and diffusion constants D_p , and (iii) diffusive modes with frequencies

$$\omega_d = -iD_d q^2 \pm \sqrt{-(D_d q^2)^2 + Bq^4},$$
 (13)

with diffusion constants D_d and bending terms *B* that are missed if the Frank energy is neglected. For $B/D_d^2 \ll 1$ the diffusive modes split up into slow and fast modes

$$\omega_s = -iB/(2D_d)q^2, \quad \omega_f = -i2D_d q^2. \tag{14}$$

Specifics of the sound velocities, the diffusion constants, and the bending terms are given in the following.

First, let us consider the case that **q** lies in the *xz* plane. In this case the equation of motion for u_y decouples from the equations of motion for u_x and u_z . This equation produces a set of transverse modes with $\mathbf{u} \parallel \hat{e}_y$. There is a nonhydrodynamic mode with

$$D_{m,y} = (4\rho)^{-1} [\sqrt{\nu_{xyxy}^R - \nu_{xyxy}} \hat{q}_x + \sqrt{\nu_{zyzy}^R - \nu_{zyzy}} \hat{q}_z]^2, \quad (15)$$

where $\hat{q}_i = q_i/q$, and there are propagating modes with

$$C_y = \sqrt{\overline{C}}/(4\rho)|\cos\theta\hat{q}_x + \sin\theta\hat{q}_z| = \sqrt{\overline{C}}/(4\rho)|\hat{q}_{x'}|, \quad (16a)$$

$$D_{p,y} = (8\rho)^{-1} [\nu_{xyxy}^R \hat{q}_x^2 + 2\nu_{xyzy}^R \hat{q}_x \hat{q}_z + \nu_{zyzy}^R \hat{q}_z^2].$$
(16b)

In the soft direction, i.e., for $\mathbf{q} \| \hat{e}_{z'}$, these propagating modes become diffusive with $D_{d,y} = D_{p,y}$ and

$$B_{y} = \rho^{-1} [\bar{K}_{1} \hat{q}_{x}^{4} + \bar{K}_{2} \hat{q}_{z}^{4} + \bar{K}_{3} \hat{q}_{x}^{2} \hat{q}_{z}^{2} + 2\bar{K}_{4} \hat{q}_{x}^{3} \hat{q}_{z} + 2\bar{K}_{5} \hat{q}_{x} \hat{q}_{z}^{3}],$$
(17)

where the \bar{K} 's are bending moduli that are combinations of the usual Frank elastic constants, the order parameter c as well as λ , α , and β . The equations of motion for u_x and u_z can be solved by decomposing (u_x, u_z) into a longitudinal part u_l along **q** and a transversal part u_T . In the incompressible limit u_l vanishes. The equation of motion for u_T produces a pair of propagating modes with

$$C_{T} = \sqrt{1/\rho} \left\{ \left[C_{xxxx} + C_{zzzz} - 2C_{xxzz} \right] \hat{q}_{x}^{2} \hat{q}_{z}^{2} + \left[C_{xxxz} - C_{zzxz} \right] \hat{q}_{x} \hat{q}_{z} (\hat{q}_{z}^{2} - \hat{q}_{x}^{2}) + \frac{1}{4} C_{xzxz} (\hat{q}_{z}^{2} - \hat{q}_{x}^{2}) \right\}^{1/2}$$
(18a)

$$D_{p,T} = (2\rho)^{-1} \bigg[\big[\nu_{xxxx} + \nu_{zzzz} - 2\nu_{xxzz} \big] \hat{q}_x^2 \hat{q}_z^2 + \big[\nu_{xxxz} - \nu_{zzxz} \big] \hat{q}_x \hat{q}_z (\hat{q}_z^2 - \hat{q}_x^2) + \frac{1}{4} \nu_{xzxz} (\hat{q}_z^2 - \hat{q}_x^2) \bigg],$$
(18b)

and $\mathbf{u} \| \hat{e}_T$ where $\hat{e}_T = (\hat{q}_z, 0, -\hat{q}_x)$.

Finally, we turn to the case $\mathbf{q} \| \hat{e}_y$. There is a pair of longitudinal propagating modes with $\mathbf{u} \| \hat{e}_y$ that is suppressed in the incompressible limit. There is a nonhydrodynamic mode with

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$$D_{m,xz} = (4\rho)^{-1} [\nu_{xyxy}^R - \nu_{xyxy} + \nu_{zyzy}^R - \nu_{zyzy}], \qquad (19)$$

where **u** lies in the *xz* plane with $u_x = \alpha(\tau_1 + \tau_3) / [\beta(\tau_1 + \tau_2)]u_z$. There is a pair of elastically soft diffusive modes with polarization $\mathbf{u} \parallel \hat{e}_{z'}$ with

$$D_{d,z'} = (8\rho)^{-1} \nu_{z'y'z'y'}^R, \qquad (20a)$$

$$B_{z'} = \rho^{-1} [\sin^2 \theta \overline{K}_6 - \sin 2\theta \overline{K}_8 + \cos^2 \theta \overline{K}_7], \quad (20b)$$

where $\nu_{z'y'z'y'}^{K}$ is the renormalized version of $\nu_{z'y'z'y'}$ and where the \overline{K} 's are once more bending moduli depending on the Frank constants, c, λ , α , and β . Finally, there is a pair of propagating modes polarized along $\hat{e}_{x'}$ with

$$C_{x'} = \sqrt{\bar{C}/(4\rho)}, \qquad (21a)$$

$$D_{p,x'} = (8\rho)^{-1} \nu^R_{x'y'x'y'}, \qquad (21b)$$

with $\nu_{x'y'x'y'}^R = \cos^2 \theta \nu_{xyxy}^R + \sin 2\theta \nu_{xyzy}^R + \sin^2 \theta \nu_{zyzy}^R$

In summary, we have presented a theory for the lowfrequency, long-wavelength dynamics of soft SmC elastomers. This theory predicts that, at least in an idealized limit, SmC elastomers possess the fascinating property of dynamic soft elasticity. Though the equations of motion are complicated, the resulting mode structure is, with respect to the softness-related symmetry directions, nicely symmetric and it has, when visualized, a certain beauty. We have calculated various dynamical quantities, such as storage and loss moduli and sound velocities, that are, in principle, accessible by experiments and we hope that our theory encourages such experimental work.

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