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 are rubbery materials that have the macroscopic symmetry properties of smectic liquid crystals. They are sure to have intriguing properties, some of which have already been studied experimentally and/or theoretically. 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. for the first time 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, 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, a purely hydrodynamical theory of SmC elastomers involves only a displacement field and not the Frank director, which relaxes to the local strain in a nonhydrodynamic time. We go beyond hydrodynamics, by including 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 crosslinked amorphous solids whose static elasticity is most easily described in Lagrangian coordinates in which labels a mass point in the undeformed (reference) material and \( R(x) = x + u(x) \), where \( u(x) \) is the displacement variable, labels the position of the mass point in the deformed (target) material. Lagrangian elastic energies are formulated in terms of the strain tensor which, in its linearized form, has the components \( u_{ij} = \frac{1}{2}(\eta_{ij} + \eta_{ji}) \), where \( \eta_{ij} = \partial_j u_i \) are the components of the displacement gradient tensor.

The elastic energy density \( f \) of the SmC elastomers of interest here can be divided into two parts

\[
   f = f_u + f_{u,n}.
\]

where \( f_u \) depends only on \( u \) and \( f_{u,n} \) describes the dependence of \( f \) on the Frank director \( n \) 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 direction of tilt in the resulting SmC phase so that the equilibrium director characterizing the undeformed SmC phase is of the form \( n^0 = (c, 0, \sqrt{1 - c^2}) \) with \( c \) being the order parameter of the transition. With these conventions, \( f_u \) can be written in the same form as the elastic energy density of conventional monoclinic solids,

\[
   f_u = \frac{1}{2} C_{xyxy} u_{xy}^2 + C_{xyzy} u_{xy} u_{zy} + \frac{1}{2} C_{zyzy} u_{zy}^2 + \frac{1}{2} C_{zzzz} u_{zz}^2 + \frac{1}{2} C_{xxxx} u_{xx}^2 + \frac{1}{2} C_{yyyy} u_{yy}^2 + \frac{1}{2} C_{zzxx} u_{xz}^2 + C_{zzzz} u_{zz} u_{xx} + C_{zzzy} u_{zz} u_{yy} + C_{zxyy} u_{xx} u_{yy} + C_{zxzz} u_{zz} u_{zz} u_{zz},
\]

but with constraints relating the three elastic constants in the first row. These latter constants can be expressed in terms of an overall elastic constant \( C \) and an angle \( \theta \), which depends on the order parameter \( c \), as

\[
   f_u = \frac{1}{2} C_{xyxy} u_{xy}^2 + C_{xyzy} u_{xy} u_{zy} + \frac{1}{2} C_{zyzy} u_{zy}^2 + \frac{1}{2} C_{zzzz} u_{zz}^2 + \frac{1}{2} C_{xxxx} u_{xx}^2 + \frac{1}{2} C_{yyyy} u_{yy}^2 + \frac{1}{2} C_{zzxx} u_{xz}^2 + C_{zzzz} u_{zz} u_{xx} + C_{zzzy} u_{zz} u_{yy} + C_{zxyy} u_{xx} u_{yy} + C_{zxzz} u_{zz} u_{zz} u_{zz}.
\]

Neglecting contributions from the Frank energy, \( f_{u,n} \) can be stated as

\[
   f_{u,n} = \frac{1}{2} \Delta (Q_y + \alpha u_{xy} + \beta u_{yz})^2
\]

where \( \Delta \) is a coupling constant and where \( \alpha \) and \( \beta \) are dimensionless parameters. The variable \( Q_y \) stands for

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Equations (1) to (3) imply that SmC elastomers are soft under static conditions. Imagine that $Q_y$ has relaxed locally to $Q_y = -\alpha u_{xy} - \beta u_{xz}$ so that $f$ is effectively reduced to $f_u$. Then, due to the above relations among the elastic constants, deformations characterized by Fourier transformed displacements $\mathbf{u} \parallel \hat{e}_y$ and wavevectors $\mathbf{q} \parallel \hat{e}_2 = (-\sin \theta, 0, \cos \theta)$ or, alternatively, $\mathbf{u} \parallel \hat{e}_2$ and $\mathbf{q} \parallel \hat{e}_y$ cost no elastic energy and hence cause no restoring forces. The effects of soft elasticity are more evident in a coordinate systems rotated through $\theta$ 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, $C_{x'y'z'}$ and $C_{z'y'z'y}$ vanish implying there is no energy cost for shears $u_{y'z'}$. If the elastomer is crosslinked in the SmC phase, these moduli become nonzero. Here, we will not consider such semi-soft 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]

$$
\dot{u}_i = \lambda_{ijk} \partial_j \hat{u}_k - \frac{\Gamma}{\delta n_i} \delta H, \\
\rho \ddot{u}_i = \lambda_{kij} \partial_j \hat{u}_k - \frac{\delta H}{\partial n_i} + \nu_{ijkl} \partial_j \partial_i \hat{u}_k,
$$

(4a)

(4b)

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

$$
\lambda_{ijk} = \frac{1}{2} \left( \delta^T_{ij} n_k + \delta^T_{ik} n_j \right) + \frac{1}{2} \left( \delta^T_{ij} n_k - \delta^T_{ik} n_j \right),
$$

(5)

where $\delta^T_{ij} = \delta_{ij} - n_i n_j$. As they stand, Eqs. (4) are valid for any liquid crystal elastomer with a defined director, like e.g., nematic, SmA and SmC elastomers. To describe SmC elastomers we have to specify $\nu_{ijkl}$ and $\nu_{ijkl}$ accordingly. From the above it is clear that $\mathcal{H} = \int d^3 x y f$ 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 parameterized, 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 $\nu_{ijkl}$ and with $u_{ij}$ replaced by $\hat{u}_{ij}$. The $\nu_{ijkl}$ depend on the order parameter $c$, $\nu_{xzzz}, \nu_{yzzz}$, and $\nu_{zzzz}$ vanish at the SmC to SmA transition and are therefore expected being 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 $\omega$ such that $\omega \tau_E \ll 1$, where $\tau_E$ is the longest elastomer time, the viscosities and $\Gamma$ are practically frequency independent. When $\omega \tau_E \gtrsim 1$, however, the viscosities $\nu_{ijkl}$ and $\Gamma$ develop a non-trivial frequency dependence. In the following we will consider in detail only the case $\tau_n \gg \tau_E$ and $\omega \tau_E \ll 1$. As mentioned above, Eq. (3) omits contributions from the Frank elastic energy for director distortions, which are 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 aforementioned 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

$$
Q_y = -\alpha \frac{1 + i \omega \tau_0}{1 - i \omega \tau_1} u_{xy} - \beta \frac{1 + i \omega \tau_2}{1 - i \omega \tau_1} u_{yz} 
$$

(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”) $\tau_1$ 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

$$
\rho \omega^2 u_i = -\partial_j \sigma_{ij}(\omega)
$$

(7)

with a symmetric stress tensor $\sigma$ given by

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

$$
\sigma_{xx}(\omega) = \frac{1}{2} C_{x\varepsilon}(\omega) u_x + \frac{1}{2} C_{\varepsilon xx}(\omega) u_x, \quad (8b)
$$

$$
\sigma_{\xi \chi}(\omega) = \frac{1}{2} C_{\xi \chi}(\omega) u_{\chi}, \quad (8c)
$$

where we use a compact notation with indices $\mu, \nu$ running over $xx, yy$ and $zz$ and indices $\xi, \chi$ running over $xy$ and $yz$. $C_{ijkl}(\omega)$ 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

$$
C_{\xi \chi}^{R}(\omega) = C_{\xi \chi} - i \omega \nu_{\chi \varepsilon} - \frac{i \omega \tau_1}{1 - i \omega \tau_1} \Delta A_{\xi \chi},
$$

(9)

$$
C_{\xi \chi}^{R}(\omega) = C_{\xi \chi} - i \omega \nu_{\chi \varepsilon}^{R} + O(\omega^2),
$$

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

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 $\omega$ 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. To highlight this phenomenon, let us switch briefly to the rotated reference space coordinates $x', y', z'$. Then, Eq. (9) implies that

$$C_{y'y'z'}^{R}(\omega) = -i\omega \nu_{y'y'z'} - \frac{i\omega\tau_1}{1 - i\omega\tau_1} \Delta A_{y'y'z'}$$  (10)

with $\nu_{y'y'z'} = \sin^2 \theta \nu_{xy} - \sin 2\theta \nu_{xyz} + \cos^2 \theta \nu_{yz}$ and an analogous expression for $A_{y'y'z'}$. $C_{y'y'z'}^{R}(\omega)$ is of the same form as Eq. (10). Thus, $C_{y'y'z'}^{R}(\omega)$ and $C_{x'x'y'}(\omega)$ vanish in the limit $\omega \rightarrow 0$ where we recover true soft elasticity. At non-vanishing frequency the system cannot be ideally soft but it can be nearly so for $\omega$ small. This type of behavior was first predicted for nematic elastomers, where it has been termed dynamic soft elasticity. 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 semi-soft nematic elastomers, depends on $\tau_n/\tau_E$. If $\tau_n \gg \tau_E$, the storage moduli exhibit a step and the corresponding loss moduli an associated peak at $\omega\tau_n \approx 1$ as shown in Fig. 11 if $\tau_n \approx \tau_E$, or $\tau_n < \tau_E$, this is not the case. The storage moduli $C_{y'y'z'}(\omega)$ and $C_{x'x'y'}(\omega)$ in the rotated frame are zero at zero frequency. In a semi-soft SmC, however, they will exhibit behavior similar to that in Fig. 11 for $\tau_n \gg \tau_E$. In nematic elastomers, there is still some controversy 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 $\tau_E$ 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 $\omega(q) = C(\theta, \varphi)q$, where $q = |q|$ and where $\theta$ and $\varphi$ are the azimuthal and polar angles of $q$ in spherical coordinates. Their sound velocities $C(\theta, \varphi)$, as calculated from Eq. (4) with the viscosities $\nu_{ijkl}$ 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 $\hat{e}_y$ and $\hat{e}_z$ so that when viewed in the $y'z'$-plane it has a clover leaf-like shape. The remaining 2 pairs are associated with non-soft 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 non-vanishing 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 elastomers, 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) non-hydrodynamic modes with frequencies

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

with a zero-$q$ decay time $\tau_1$ and diffusion constants $D_m$, (ii) propagating modes with frequencies

$$\omega_p = \pm C q - iD_p q^2$$  (12)

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

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

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

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 $u \parallel \hat{e}_y$. There is a non-hydrodynamic mode with

$$D_{n,y} = (4\rho)^{-1} \left[ \sqrt{\nu_x^R - \nu_{zyy}} \mathbf{q}_x + \sqrt{\nu_x^R - \nu_{zyy}} \mathbf{q}_z \right]^2, \quad (15)$$

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

$$C_y = \sqrt{C/(4\rho)} \left| \cos \theta \mathbf{q}_x + \sin \theta \mathbf{q}_z \right| = \sqrt{C/(4\rho)} |\mathbf{q}_s|, \quad (16a)$$

$$D_{p,y} = (8\rho)^{-1} \left[ \nu_{xyy} \mathbf{q}_x^2 + 2 \nu_{xyy} \mathbf{q}_z \mathbf{q}_x + \nu_{xyy} \mathbf{q}_z^2 \right]. \quad (16b)$$

In the soft direction, i.e. for $q_3 \parallel \hat{e}_z$, these propagating modes become diffusive with $D_{d,y} = D_{p,y}$ and

$$B_y = \rho^{-1} \left[ \tilde{K}_1 \mathbf{q}_x^2 + \mathbf{K}_2 \mathbf{q}_z^2 + \mathbf{K}_3 \mathbf{q}_x^2 \mathbf{q}_z^2 + 2 \mathbf{K}_4 \mathbf{q}_x^2 \mathbf{q}_z^2 + 2 \mathbf{K}_5 \mathbf{q}_x^2 \mathbf{q}_z^2 \right], \quad (17)$$

where the $\tilde{K}_i$ are bending moduli that are combinations of the usual Frank elastic constants, the order parameter $c$ as well as $\alpha$, $\lambda$, and $\beta$. 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

$$C_T = \sqrt{1/\rho} \left\{ [C_{xxxx} + C_{zzzz} - 2C_{xzzz}] \mathbf{q}_x^2 \mathbf{q}_z^2 \right\} \quad (18a)$$

$$D_{p,T} = (2\rho)^{-1} \left[ \nu_{xxxx} + \nu_{zzzz} - 2\nu_{xzzz} \mathbf{q}_x^2 \mathbf{q}_z^2 \right] \quad (18b)$$

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

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

$$D_{n,z} = (4\rho)^{-1} \left[ \nu_{xyy}^R - \nu_{zyy} + \nu_{zyy}^R - \nu_{zyy} \right], \quad (19)$$

where $u$ lays in the $xz$-plane with $u_x = \alpha(\tau_1 + \tau_3)/[\beta(\tau_1 + \tau_3)] |u_z$. There is a pair of elastically soft diffusive modes with polarization $u \parallel \hat{e}_z$ with

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

$$B_z = \rho^{-1} \left[ \sin^2 \theta \tilde{K}_6 - \sin 2\theta \tilde{K}_8 + \cos^2 \theta \tilde{K}_7 \right], \quad (20b)$$

where $\nu_{z'y'y'}^R$ is the renormalized version of $\nu_{z'y'y'}^R$ and where the $\tilde{K}_i$’s are once more bending moduli depending on the Frank constants, $c$, $\lambda$, $\alpha$, and $\beta$. Finally, there is a pair of propagating modes polarized along $\hat{e}_z$' with

$$C_{z'} = \sqrt{C/(4\rho)} , \quad (21a)$$

$$D_{p,z'} = (8\rho)^{-1} \nu_{z'y'y'}^R, \quad (21b)$$

with $\nu_{z'y'y'}^R = \cos^2 \theta \nu_{zyy}^R + \sin^2 \theta \nu_{zyy}^R$.

In summary, we have presented a theory for the low-frequency, 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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[1] For a review on liquid crystal elastomers see W. Warner and E.M. Terentjev, Liquid Crystal Elastomers (Clarendon Press, Oxford, 2003)
[2] For a review on liquid crystals see P. G. de Gennes and J. Prost, The Physics of Liquid Crystals (Clarendon Press, Oxford, 1993); S. Chandrasekhar, Liquid Crystals (Cambridge University Press, Cambridge, 1992).
[3] K. Hiraoka, W. Sagan, T. Nose and H. Finkelmann, Macromolecules 38 7352 (2005).
[4] O. Stenull and T. C. Lubensky, Phys. Rev. Lett. 94, 018304 (2005); forthcoming paper. See also J. M. Adams and M. Warner, Phys. Rev. E 72, 011703 (2005).
[5] O. Stenull and T. C. Lubensky, Phys. Rev. E 69, 051801 (2004).
[6] See, e.g., N. W. Ashcroft and N. D. Mermin, Solid State Physics, (Saunders, Philadelphia, 1976).
[7] D. Forster, T. C. Lubensky, P. C. Martin, J. Swift, and P. S. Persham, Phys. Rev. Lett. 26, 1016 (1971); D. Forster, Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions (Addison Wesley, Reading, Mass., 1983); D. Forster, Phys. Rev. Lett. 32, 1161 (1974).
[8] For more detailed theoretical estimates of the $\nu_{ijkl}$ one has to resort to molecular-based theories, as was done for conventional SmC’s in M. A. Ospov, T. J. Sluckin, and E. M. Terentjev, Liq. Cryst. 19, 197 (1995).
[9] E. M. Terentjev, I. V. Kamotski, D. D. Zakharov, and L. J. Fradkin, Phys. Rev. E 66, 052701(R) (2002). L. J. Fradkin, I. V. Kamotski, E. M. Terentjev and D. D. Zakharov, Proc. R. Soc. Lond. A 459, 2627 (2003).
[10] P. Martinioty et al., Eur. Phys. J. E 14, 311 (2004); E. Terentjev and M. Warner, ibid. 14, 323 (2004); O. Stenull and T. C. Lubensky, ibid. 14, 333 (2004).