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Short Communication

Layer-network coupling in smectic elastomers

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Abstract. — The complex coupling between elastic strains and smectic layer distortions in smectic A elastomers is re-analysed in terms of the penalty for the relative translations of layers with respect to the matrix in which they are embedded. This modifies the starting point of a recent theory of the elastic free energy of smectic A elastomers (Terentjev E.M. and Warner M., J. Phys. II France 4 (1994) 111-126), but leaves most of the analysis itself and the conclusions unchanged. In the large, layers move affinely with the elastic matrix, but there are layer fluctuations possible on a scale set by the mean size of chains in the network. There is a rigid constraint that uniform shear strains of the network produce corresponding uniform rotations of the smectic layer system.

New mesophase solids, in the form of rubber networks that are nematic or smectic, have remarkable elastic properties because of their liquid crystalline internal degrees of freedom. These degrees of freedom are coupled to elastic strains and lead to novel forms of mechanical instability, soft modes, piezo- and ferro-electricity, and new smectic structures. Descriptions of these new phenomena have been both molecular and continuum in nature for nematic elastomers, and limited to continuum models for smectics. Because elastomers are capable of very large deformations, many new effects appear in a highly non-linear regime and a molecular approach is preferable. In order to estimate the size of effects within continuum theory, and to see what length scales emerge, a molecular approach is also desirable. However, since there is no molecular description of smectic elastomers at this time, the continuum theory may help to understand the qualitative effects and underlying physics of such a system.

The purpose of this note is to clarify the nature of the coupling between smectic layer distortions and distortions of the polymer network in which these layers are embedded. It will turn out to a large extent that layer distortions are affine with those of the network, small fluctuations on top of uniform strains being possible. By examining layer displacements relative to the matrix in which they are embedded, we fix the lengths scales involved in the continuum
analysis. A recent analysis [1] based on relative strains rather than displacements is shown to have some shortcomings, though most of the predictions of new effects for smectic elastomers still stand.

Relative translations coupling.

If we let the z displacement (in the direction of its normal) of the smectic layer at the point \( r \) be \( U(\mathbf{r}) \), then there is clearly a penalty for its motion with respect to the \( z \) component of the local network displacement, \( V_z(\mathbf{r}) \). For small relative displacements this must penalty must be harmonic with a free energy cost of:

\[
F_d = \Lambda \int \mathrm{d}\mathbf{r}[V_z(\mathbf{r}) - U(\mathbf{r})]^2
\]

(1)

The coefficient \( \Lambda \) cannot be estimated within continuum theory, but the dimensional analysis allows the relevant magnitudes and length scales to emerge: equation (1) describes an effect based on the entropy of a rubbery network, which in continuum model has a short distance cut-off at \( R_0 \) — the spatial extent of elastically active strands. Hence the characteristic energy scale is \( k_B T \) and, taking into account the number of strands per unit volume \( N_x \), we can write

\[
\Lambda \approx \alpha N_x k_B T / R_0^2
\]

(2)

where \( \alpha \) depends on the smectic order parameter and vanishes in a high-temperature homogeneous phase, and where \( N_x k_B T \approx \mu \) is the shear modulus of the elastomer (for simplicity in the isotropic state). Since chain configurations in the rubbery state are random walks we have \( N_x \sim \langle l R_0^2 \rangle^{-1} \), with \( l \) being the step length of such a walk. Equation (1) suggests that smectic layers can fluctuate in position by \( \sim R_0 \) which may be considerably larger than \( d_o \).

The layer and network contributors to equation (1) are:

\[
U(\mathbf{r}) = U(0) + r_x \lambda_i + u(\mathbf{r})
\]

(3)

\[
V_j(\mathbf{r}) = V_j(0) + r_x \alpha_j + v_j(\mathbf{r})
\]

where \( U(0) \) and \( V_j(0) \) are the mean layer and mean network displacements, \( \lambda_i \) is the uniform part of \( \nabla_i U \) (the gradient of layer distortions) and \( u(r) \) is the fluctuating remainder, \( \lambda_{2i} \) is the \((z, i)\)th component of the tensor \( \lambda \) of uniform distortions of the network, and \( v \) is the fluctuating part. Summation over repeated indices \( i = 1, 2, 3 \) is implied. Inserting (3) into free energy (1) one obtains:

\[
\frac{F_d}{\Lambda} = \mathcal{V} [V_z(0) - U(0)]^2 + \mathcal{V} \gamma^{2/3} (\omega_i - \lambda_{2i})^2 + \int \mathrm{d}\mathbf{r} [u(\mathbf{r}) - v_z(\mathbf{r})]^2
\]

(4)

(with \( \mathcal{V} \) the volume of the system). The first term implies that there can be no uniform relative displacement of layers and network, that is there is no Goldstone mode of the relative translation in our system. The second, extremely large term constrains uniform layer strains to exactly follow the uniform elastic strains of the network. The third term is a penalty on layer fluctuations, additional to those acting on the layers of conventional smectics, and correlates layer fluctuations with mechanical fluctuations of the network.

Effect of the \( \omega_i - \lambda_{2i} \) coupling on nematic aspects of the free energy.

Underlying the smectic order is the nematic order of the component rods and hence that of the chains which the rods compose. There is another, nematic, contribution to the free energy
which occurs when the network, which fixes the configuration of chains, is rotated relative to the nematic director \([2, 3]\). Since in smectic A phases the director is perpendicular to layers, director rotation about a perpendicular axis can be described by the rotation of the layers themselves. Thus the de Gennes' term for the free energy density penalising nematic rotation, \(\nabla \cdot u\), relative to that of the matrix, \((\partial v_z/\partial x)^A\), is 

\[
\frac{b}{2} \left( \frac{\partial v_z}{\partial y} - \frac{\partial v_y}{\partial z} \right) \left( \frac{\partial v_z}{\partial y} - \frac{\partial v_y}{\partial z} \right)
\]

and the energy can be expressed as

\[
\frac{b}{2} \int dr \left[ \lambda_{zy}^s - \frac{1}{2} \left( \frac{\partial v_z}{\partial x} - \frac{\partial v_x}{\partial z} \right) \right]^2 + \left[ \lambda_{zx}^s - \frac{1}{2} \left( \frac{\partial v_z}{\partial y} - \frac{\partial v_y}{\partial z} \right) \right]^2
\]

(plus the similar term for rotation about \(y\).) According to the second term in equation (4) we must identify the uniform part \(w_i\) of \(\partial U/\partial x_i\) with \(\lambda_{zi}\), leaving behind \(\lambda_{zi}^s\) (the symmetric part) and the fluctuating parts. Whence the energy penalty becomes

\[
b \int dr \left[ \lambda_{zy}^s - \frac{1}{2} \left( \frac{\partial v_z}{\partial x} - \frac{\partial v_x}{\partial z} \right) \right]^2 + \left[ \lambda_{zx}^s - \frac{1}{2} \left( \frac{\partial v_z}{\partial y} - \frac{\partial v_y}{\partial z} \right) \right]^2
\]

that is, layer fluctuations couple to \(\lambda^s\), the symmetric part of the strain in the rubber.

The coupling (5) does not lead to new effects since \(\lambda^s\) is uniform and \(\partial u/\partial x\), and \(\partial v/\partial x\), are fluctuating. Expanding out, one obtains 

\[
b \int dr \left( \lambda_{zy}^s \right)^2 + \left( \partial u/\partial x - (\nabla v_z)^A \right)^2
\]

where the cross-coupling between \(\lambda^s\) and the spatially varying terms reduces to a surface contribution and is dropped. (Thus in [1] the non-fluctuating parts of the network-layer couplings are not as in the first part of equation (17), but are the rigid coupling of the uniform distortions \(\lambda_{zi} = w_i\).) The survivors of equation (5) above are a renormalisation of the uniaxial matrix rubber elasticity [by \(b(\lambda_{zi}^s)^2\)] and constraints on relative local layer-matrix rotations \(b(\partial u/\partial x - (\nabla v_z)^A)^2\) that are absent in conventional smectics.

The remainder of [1] should be read with the layer fluctuations \(u(r)\) in mind, as is there discussed (page 119). The coefficient \(b\) is also discussed in [1] (page 120) and by arguments similar to those above fixing \(A\), is suggested to be of the order of \(\mu\), the rubber shear modulus, that is in the range \(10^4 - 10^5 J/m^3\).

In summary we conclude that uniform smectic distortions are rigidly locked to the corresponding network strains, while undulating deformations take a complex form and involve couplings between the matrix and layer distortions. These couplings have a profound effect on the critical phenomena of elastomeric smectics and we shall return to this in future work. All piezoelectric effects discussed in [1] remain as there described.

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References

