Anomalous elasticity of disordered nematic gels

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Anomalous elasticity of disordered nematic gels

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Nematic polymer networks constitute a unique class of elastic solids as their translational and orientational degrees of freedom are coupled [1]. They can be fabricated by, for example, crosslinking a melt of liquid-crystalline polymers in the isotropic phase and then cooling it down into the nematic phase. The resulting networks often contain a high density of director textures with an orientational correlation length in the micrometer range. It has been argued that such a "poly-domain" state is stabilized by random internal stresses [2] or a random molecular field exerted by crosslinks [3,4]. In this article, I show that these disordered nematic gels should exhibit an anomalously soft mechanical response against stretching.

A simplest model free energy of disordered nematic networks reads

$$F = \int d\boldsymbol{r} \left[\mu \left(U_{ij}^2 - \alpha Q_{ij} U_{ij} - R_{ij} U_{ij}\right) + \frac{L}{2}(\partial_i Q_{jk})^2\right], \tag{1}$$

where $U_{ij} = (1/2)(\partial u_i/\partial r_j + \partial u_j/\partial r_i)$ is the strain tensor, $Q_{ij} = n_i n_j - (1/d)\delta_{ij}$ is the orientational order parameter with n_i being the director, and α is the coupling constant. The field R_{ij} represents the random internal stresses and L is the Frank constant. We assume that the gel is incompressible, $U_{ii} = 0$. The strain variable can be eliminated from eq.(1) using the mechanical equilibrium condition, $\delta F/\delta u_i = 0$, and assuming that there is no average deformation, $\langle U_{ij} \rangle = 0$. In this paper we consider only the two-dimensional case, for which the result is

$$F = \int d\boldsymbol{r} \left[-\frac{\mu}{2} \left(\alpha Q_1 + R_1^S\right)^2 + \frac{L}{2}(\partial_i Q_{jk})^2\right]. \tag{2}$$

Here, $Q_1(\boldsymbol{r})$ is a real variable defined through its Fourier transform,

$$Q_1(\boldsymbol{q}) = \sin(2\varphi) Q_{xx}(\boldsymbol{q}) - \cos(2\varphi) Q_{xy}(\boldsymbol{q}), \tag{3}$$

where φ is the azimuthal angle of the wavevector $\boldsymbol{q} = q(\cos\varphi, \sin\varphi)$. A relation parallel to eq.(3) holds between R_1^S and $R_{ij}^S = R_{ij} - (1/2)R_{kk}\delta_{ij}$. If we assume that R_{ij} has no spatial correlation, a simple dimensional argument (of Imry-Ma type) shows that the interaction between Q_1 and R_1^S renders the orientational correlation length finite at equilibrium.

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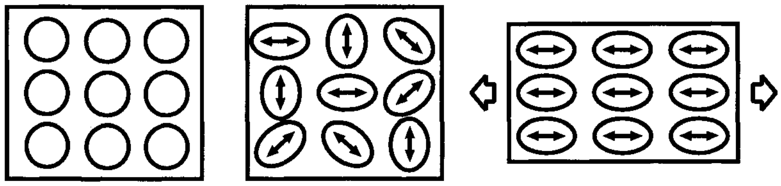

FIGURE 1. Schematic illustration of the isotropic, polydomain nematic, and monodomain nematic states from the left to right. Ellipses show the direction and magnitude of the local strain, and arrows in them indicate the director. The polydomain-monodomain transition proceeds via simultaneous rotation of the director and the strain axis.

Now let us consider the mechanical response. If the quenched disorder is enough weak and if the domain size is much larger than $(L/\mu\alpha^2)^{1/2}$, we can approximate the free energy density (averaged over the space) as

$$f = -\frac{\mu}{2}\left\langle(\alpha Q_1 + R_1^S)^2\right\rangle + \frac{L}{2}\left\langle(\partial_i Q_{jk})^2\right\rangle \simeq -\frac{\mu\alpha^2}{2}\left\langle Q_1^2\right\rangle \quad (4)$$

To reduce it, $\langle Q_1^2\rangle$ tends to be large. Its maximum value is known to be $1/4$ from the identity $\langle Q_1^2 + Q_2^2\rangle = \left\langle Q_{xx}^2 + Q_{xy}^2\right\rangle = 1/4$, where $Q_2(\boldsymbol{r})$ is defined by

$$Q_2(\boldsymbol{q}) = \cos(2\varphi)Q_{xx}(\boldsymbol{q}) + \sin(2\varphi)Q_{xy}(\boldsymbol{q}). \quad (5)$$

Thus the free energy density of the polydomain state with $\langle U_{ij}\rangle = 0$ is given by $f_{poly} = -\mu\alpha^2/8$ in the weak-disorder limit.

External strain tends to align the director in the stretching direction. At a finite value of strain the system turns into a "monodomain" state with few defects. Assuming that $U_{ij} = \text{const.}$ and $Q_{ij} = \text{const.}$, the free energy eq.(1) is minimized at $U_{ij} = (\alpha/2)Q_{ij}$. This amounts to an elongation by the ratio $\lambda_m = 1 + \alpha/4$ along the director. The minimum value $f_{mono} = -\mu\alpha^2/8$ is equal to f_{poly}; the strain-induced polydomain-monodomain transition accompanies no change of the free energy, at least to $O(\alpha^2)$. A vanishingly small mechanical stress has been experimentally observed, for instance, in [5].

The anomalous softness can be interpreted as follows. Each part of the system gets elongated by λ_m times along the director on the initial quench from the isotropic phase. The inhomogeneous distortion requires a long-ranged organization of the director field, which is characterized by the asymmetry $|Q_1(\boldsymbol{q})|^2 > |Q_2(\boldsymbol{q})|^2$. (This asymmetry produces a "four-leaf clover"-shaped pattern in the depolarized light scattering intensity [6], which resembles the experimentally observed one [3].) The elongated domains provide a mechanical Goldstone mode; the local strain axis keeps to coincide with the director during its rotation in response to stretching, which means that the elastic free energy is unchanged (see fig. 1). Some additional discussions and numerical results are presented in ref. [7].

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