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Light Scattering from Nematic Liquid Crystals in a Nonuniform Velocity Field

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Fluctuations about a nonequilibrium steady state of a nematic liquid crystal subject to small, linear shear, are studied by means of a generalized Langevin method. We find a symmetric broadening of the director power spectrum and estimate the numerical size of the effect for a typical material.

A. INTRODUCTION

Fluctuations about nonequilibrium steady states is a subject receiving considerable attention in the current literature.[1,2,3,4] The recent emergence of theoretical models for such systems has motivated our search for a system with interesting new behavior. To date, the calculations undertaken are limited to studies of the properties of ordinary fluids in the presence of a temperature or velocity gradient. In this letter, we investigate the properties of a different system which exhibits fluctuations about a nonequilibrium steady state: a uniaxial nematic liquid crystal subject to a small uniform velocity gradient. This system is of particular interest because there already exists a linear coupling of the velocity to the director through the reactive parameter λ and a consequent flow alignment of the director.[5]

We have employed a generalized Langevin method due to Tremblay, Siggia, and Arai[1] to calculate the light scattering spectrum in the presence of the shear. This technique has the advantage that it provides nonequilibrium generalizations of familiar equilibrium concepts like the

Einstein relation, the Onsager regression hypothesis and the fluctuation-dissipation theorem.

This generalized Langevin method has been developed in the context of a general set of hydrodynamic equations for modes A_α linearized about the nonequilibrium steady state in question and acted upon by Langevin forces F_α. In general, the set of hydrodynamic equations can be written

$$\dot{A}_\alpha(\mathbf{k},t) + \int \frac{d^3k'}{(2\pi)^3} M_{\alpha\beta}(\mathbf{k}, -\mathbf{k}')A_\beta(\mathbf{k}') = F_\alpha(\mathbf{k},t), \tag{1}$$

where $F_\alpha(\mathbf{k},t)$ is the Langevin force associated with the variable $A_\alpha(\mathbf{k},t)$ and a sum over repeated indices is implied. We assume the equilibrium result that $F_\alpha(\mathbf{k},t)$ is uncorrelated at unequal times or points will remain valid for the nonequilibrium states we are considering. Further, we assume that the magnitude of its correlation functions has the same form as in equilibium, with local values of the temperature and transport coefficients. To first order in the shear, Tremblay *et al.* have shown that the matrix $M_{\alpha\beta}$ can be taken to be diagonal in $\mathbf{k}$.

A simple calculation shows that time dependent correlation functions are given by

$$\langle (A_\beta(\mathbf{k},\omega)A_\gamma(\mathbf{k}',0)\rangle = [-i\omega 1 + M(\mathbf{k})]^{-1}_{\beta\alpha} \cdot D_{\alpha\nu}(\mathbf{k},\mathbf{k}')[i\omega 1 + M^\ddagger(\mathbf{k}')]^{-1}_{\nu\gamma}, \tag{2}$$

where

$$D_{\alpha\nu}(\mathbf{k},\mathbf{k}')\delta(t-t') \equiv \langle F_\alpha(\mathbf{k},t)F_\nu(\mathbf{k}',t')\rangle. \tag{3}$$

The ensemble implicit in the above Langevin method is stationary but not, in general, time reversal invariant.

B. THEORY

In a uniaxial nematic light scattering is dominated by fluctuations in the orientation of the principal axis of the dielectric tensor.[6] These are related to the fluctuating components of the director, so that the time-dependent correlation function measured by light scattering experiments in a nematic liquid crystal is that of the director.

We will consider a system where the liquid crystal is maintained in a steady state with an applied linear shear. The average local velocity of the liquid is assumed to be given by

$$\mathbf{v}^o = X\left\{\frac{X}{|X|}sz - cy\right\}\left\{c\hat{e}_z + \frac{X}{|X|}s\hat{e}_y\right\} \tag{4a}$$

where

$$c = [(\lambda + 1)/2\lambda]^{1/2} \tag{4b}$$

and

$$s = [(\lambda - 1)/2\lambda]^{1/2} \tag{4c}$$

and where X is the average rate of shear, $\hat{e}_y$ and $\hat{e}_z$ are unit vectors in the y and z directions, respectively, and where λ is the reactive parameter which couples the director and velocity. (We assume λ is such that all square roots are real.) We assume that the average density and the temperature of the liquid crystal are constant.

The equation of motion for the director is give by[7]

$$\frac{D}{Dt}n_i(\mathbf{r},t) = \frac{\lambda}{2}\delta_{il}^{\perp}n_k(\mathbf{r},t)[\nabla_l v_k(\mathbf{r},t) + \nabla_k v_l(\mathbf{r},t)] - \frac{1}{2}[\hat{n} \times (\nabla \times \mathbf{v})]_i + \frac{1}{\gamma_1}K_{ijkl}\nabla_j\nabla_l n_k(\mathbf{r},t) + \theta_i(\mathbf{r},t), \tag{5}$$

where D/Dt is the convective derivative,

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + v_j\nabla_j, \tag{6}$$

the latin letters denote Cartesian indices, and we use the summation convention for releated indices. Further, $n_i(\mathbf{r},t)$ is the director field, $\delta_{il}^{\perp}$ is the projection operator onto the plane perpendicular to $\mathbf{n}$,

$$\delta_{ij}^{\perp} = \delta_{ij} - n_i n_j, \tag{7}$$

K_{ijkl} are the Frank elastic constants for the uniaxial phase, γ_1 is the viscosity associated with the director, and θ_i is a fluctuating Langevin force satisfying

$$\langle\theta_i(\mathbf{r},t)\theta_j(\mathbf{r}',t')\rangle = 2\gamma_1^{-1}k_B T\delta_{ij}^{\perp}\delta(\mathbf{r} - \mathbf{r}')\delta(t - t'). \tag{8}$$

where k_B is Boltzmann's constant and T is the temperature.

The velocity field of Eq. (4) induces a flow alignment of the director. In the absence of an externally applied magnetic field the stable, steady state solution of Eq. (5) is

$$\mathbf{n}^o = \hat{e}_z \tag{9}$$

The time-dependent director auto-correlation function will be calculated in a coordinate frame defined by the steady state director field given above. We introduce the ordered set of basis vectors $(\hat{e}_x, \hat{e}_y, \hat{e}_z)$, where $\hat{e}_x$ is a unit vector perpendicular to $\mathbf{n}^o$ and the average velocity.

We expand the director and velocity in Eq. (5) about the steady state

$$n_i = n_i^o + \delta n_i, \tag{10}$$

$$v_i = v_i^o + \delta v_i, \tag{11}$$

and keep terms to first order in their fluctuating components and first order in the shear. In this frame, to first order, the fluctuating part of the director has only two components

$$\delta n_i \approx (\delta n_x, \delta n_y, 0). \tag{12}$$

The linearized equations of motion for the director components are

$$\frac{D^o}{Dt}\delta n_x(\mathbf{r},t) = -|X|[(\lambda^2 - 1)^{1/2}/2)]\delta n_x(\mathbf{r},t) + (\lambda/2)n_k^o[\nabla_x\delta v_k(\mathbf{r},t) + \nabla_k\delta v_x(\mathbf{r},t)] - \frac{1}{2}[\hat{n}^o\mathrm{x}((\nabla\mathrm{x}\delta\mathbf{v}(\mathbf{r},t)]_x + \frac{1}{\gamma 1}K_{xjkl}\nabla_j\nabla_l\delta n_k(\mathbf{r},t) + \theta_x(\mathbf{r},t) \tag{13a}$$

and

$$\frac{D^o}{Dt}\delta n_y(\mathbf{r}t) = -|X|(\lambda^2 - 1)^{1/2}\delta n_y(\mathbf{r},t) + \frac{\lambda}{2}n_k^o[\nabla_y\delta v_k(\mathbf{r},t) + \nabla_k\delta v_y(\mathbf{r},t)] - \frac{1}{2}[\hat{n}^o x(\nabla x\delta\mathbf{v}(\mathbf{r},t)]_y + \frac{1}{\gamma_1}K_{yjkl}\nabla_j\nabla_l\delta n_k(\mathbf{r},t) + \theta_y(\mathbf{r},t) \tag{13b}$$

where $\delta\mathbf{v}$ is the fluctuating part of the velocity and $D^o/Dt = \frac{\partial}{\partial t} + \langle\mathbf{v}^o\rangle\cdot\nabla$ and where the angular brackets denote the average velocity.

The equation of motion for the velocity is

$$\frac{D^o}{Dt}\delta v_i + (\delta\mathbf{v}\cdot\nabla)\mathbf{v}^o = -\nabla_j\delta\sigma_{ij}^R - \nabla_j\delta\sigma_{ij}^D + \zeta_i \tag{14}$$

where $\delta\sigma_{ij}^R$ and $\delta\sigma_{ij}^D$ are the reversible and dissipative parts of the stress tensor and ζ_i is a noise source given.[7]

While the time-dependent director auto-correlation function in general is quite complicated, there do exist certain geometries which simplify the power spectrum. In particular, if the scattering experiment is set up so that the wave vector is in either the x-direction or the y-direction then one component of the director decouples from the velocity and the power spectrum is trivial to calculate. From Eqs. (13a) and (13b) it is easy to see that if $\mathbf{k}$ is in the x direction, the δn_y equation decouples while if $\mathbf{k}$ is in the y direction, the δn_x equation decouples. Both power spectra exhibit the same features so we will calculate, in detail, only the case where $\mathbf{k}$ is in the y direction.

The equation of motion for δn_x becomes after we have performed a spatial Fourier transform,

$$\left\{\frac{\partial}{\partial t} + ik_y\langle v_y^o\rangle\right\}\delta n_x(\mathbf{k},t) = -\left\{\frac{|X|(\lambda^2-1)^{1/2}}{2} + \frac{K_2}{\gamma_1}k_y^2\right\}\delta n_x(\mathbf{k},t) + \theta_x(\mathbf{k},t). \quad (15)$$

This implies that the $\delta n_x - \delta n_x$ correlation function, C_{xx}, is

$$C_{xx}(\mathbf{k},\omega) = \frac{2\gamma_1^{-1}k_BT}{(\omega')^2 + \left[\frac{K_2k_y^2}{\gamma_1} + \frac{|X|(\lambda^2-1)^{1/2}}{2}\right]^2} \quad (16)$$

where ω' is the Doppler shifted frequency

$$\omega' = \omega - k_y\langle v_y^o\rangle \quad (17)$$

The result Eq. (16) shows that the effect of the shear is to increase the relaxational damping of the director mode (as would the application of an external magnetic field) and thus, in the rest frame of the liquid crystal, to symmetrically broaden the line shape. If $\mathbf{k}$ is in the x direction the $\delta n_y - \delta n_y$ correlation function has the form Eq. (16) except that no factor of one-half multiplies $|X|$ in the denominator. Finally, if we use the numbers for γ_1, λ_1, and K_2 appropriate for PAA and $k \simeq 4\pi \times 10^4 \sin\theta/2\text{cm}^{-1}$, θ a scattering angle, we find for $\theta = 2^o$ that the ratio of the additional term in the damping to the usual term is $2 \times 10^{-2}|X|$ where $|X|$ should be measured in inverse seconds.

Note added in proof:

After this manuscript was submitted we learned of similar work by H. Pleiner and H. Brand (to be published).

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