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Temperature Dependence of Normal Modes in a Nematic Liquid Crystal

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We have measured the temperature dependence of the intensity and spectrum of light scattered by fluctuations in the nematic phase of p-methoxy-benzylidene p-n-butylaniline. Our measurements are interpreted using two phenomenological theories of nematic liquid crystals.

In this Letter we report the results of an experimental study of the temperature dependence of the intensity and spectrum of light scattered by fluctuations in the ordered phase of the nematic liquid crystal p-methoxybenzylidene p-n-butylaniline (MBBA). From these measurements we determine the mean squared amplitude and time dependence of the normal modes of the ordered phase as a function of temperature. We first analyze our result using the phenomenological theory of the Orsay Liquid Crystal Group[1] to determine the temperature dependence of the elastic constants and viscosity coefficients of the model. Recently an attractively simple macroscopic model of liquid crystals has been proposed by Martin, Pershan, and Swift[2]; we analyze our data to indicate how the normal-mode displacements of this theory couple to the dielectric constant of the liquid crystal.

The order in a nematic liquid crystal of uniaxial symmetry is specified by the direction of the alignment of the liquid-crystal molecules (the optic axis) and their degree of alignment along this direction. The Cartesian dielectric-constant tensor for a uniaxial nematic may, therefore, be written

$$\delta\epsilon_{\alpha\beta}=\bar{\epsilon}\delta_{\alpha\beta}+Q(\Delta\epsilon/3)(3n_\alpha n_\beta-\delta_{\alpha\beta}), \tag{1}$$

where n_α and n_β are the Cartesian components of a unit vector (called the director) parallel to the local optic axis. An order parameter specifying the degree of alignment is[3] $Q=\frac{3}{2}\langle\cos^2\theta-\frac{1}{3}\rangle$, where θ is the angle between the long axis of a molecule and the local optic axis. The other quantities are $\bar{\epsilon}=\frac{1}{3}(\epsilon_p+2\epsilon_t)$ and $\Delta\epsilon=\epsilon_p-\epsilon_t$, where ϵ_p and ϵ_t are the dielectric constants parallel and transverse, respectively, to the optic axis of a completely ordered sample ($Q=1$). The value of Q is temperature dependent and is given approximately by the mean-field model of Maier and Saupe.[3] A uniform vector field $\vec{n}$ describes the lowest energy state of the ordered liquid.

The value of $\Delta\epsilon$ in (1) is of order unity; the order in a liquid crystal therefore has a pronounced effect upon the index of refraction, and optical methods are ideally suited to study these materials. Fluctuations in Q will scatter light and have been studied in the isotropic phase of MBBA by Stinson and Litster.[4] In the ordered liquid the strongest light scattering results from fluctuations in $\vec{n}$; these are normal modes analogous to spin waves in a ferromagnet. From the intensity and spectrum of light scattered by these fluctuations one may determine the mean squared amplitude and time dependence of the normal modes in the liquid crystal. The intensity of light scattered by these modes in p-azoxyanisole (PAA) has been studied by Chatelain[5] and the time dependence and dispersion relations determined for PAA by the Orsay Liquid Crystal Group.[6]

Our measurements were carried out by the technique of self-beating spectroscopy. Our oriented liquid-crystal sample 50 μm thick was mounted between glass microscope slides enclosed in an aluminum block whose temperature was controlled to better than 8 mdeg. The spectrum of the photocurrent was determined with a Hewlett Packard model 310A wave analyzer, and only 150 μW of laser power at 6328 Å (Spectra Physics model 119) was used in order to avoid heating the sample. Temperatures were measured with a platinum resistance thermometer. The MBBA was purified by vacuum distillation[7] and, to minimize decomposition, was maintained in a nitrogen atmosphere. The nematic-isotropic transition temperature of the sample was frequently checked and observed to stay in the range (45.45 ± 0.15)°C.

If $\vec{k}_i$ and $\vec{k}_f$ are the wave vectors of the incident and scattered light, respectively, then light is scattered by a Fourier component of the fluctuations whose wave vector is $\vec{q} = \vec{k}_f - \vec{k}_i$. Let $\vec{i}$ and $\vec{f}$ be unit vectors along the polarizations of the incident and scattered light, respectively. We studied light scattered in three configurations: (a) $\vec{n}$, $\vec{i}$, and $\vec{f}$ in the scattering plane (determined by $\vec{k}_i$ and $\vec{k}_f$); (b) $\vec{n}$ and $\vec{f}$ in the scattering plane with $\vec{i}$ normal to them; (c) $\vec{i}$ parallel to $\vec{n}$ and normal to the scattering plane with $\vec{f}$ in the scattering plane. The laboratory scattering angle was 45° for all configurations.

We now discuss our measurements using the two phenomenological models for a nematic liquid crystal.[1,2] The Orsay Group[1] analyzed its experiments using an equation of motion for the director $\vec{n}$ which was derived using the Frank elastic constants[9] and Leslie's hydrodynamic equations[10] for anisotropic fluids. Recently Martin, Pershan, and Swift (MPS)[2] have proposed a model derived directly from conservation laws for mass, momentum, and energy. In this model the director does not play an explicit role; the liquid crystal is treated as a uniaxial fluid (the axis of symmetry corresponds to the direction of $\vec{n}$) which is able to support nonuniform shears. In both models the normal modes are heavily overdamped and the spectrum of the scattered light is a Lorentzian line centered about the incident laser frequency. We refer the reader to Refs. 1 and 2 for details of these models.

We choose coordinates 1, 2, and 3 such that the optic axis lies along 3 and $\vec{q}$ is in the 1-3 plane. We studied the light which was strongly scattered by orientational fluctuations of the optic axis; these involve only the elements $\delta\epsilon_{13}$ and $\delta\epsilon_{23}$ of the dielectic-constant tensor. We find that $q_1^2 < 0.078 q_3^2$ for configuration (a) and $q_1^2 < 0.0026 q_3^2$ in configuration (b). In configuration (c), $f_2^2 < 0.0086 f_1^2$ so we observe only $\delta\epsilon_{13}$. If we ignore the q_1 components for configurations (a) and (b), then the intensity of light scattered in the three configurations may be computed from the Orsay model to be proportional to

$$\langle \delta\epsilon_{if}^2(\vec{q})\rangle = Q^2 \Delta\epsilon^2 (i_3 f_1)^2 kT/(K_{33} q_3^2), \tag{2a}$$

$$= Q^2 \Delta\epsilon^2 (i_2 f_3)^2 kT/(K_{33} q_3^2), \tag{2b}$$

$$= Q^2 \Delta\epsilon^2 (i_3 f_1)^2 kT/(K_{11} q_1^2). \tag{2c}$$

Equations (2a), (2b), and (2c) refer to our three experimental configurations, and we have used the notation of Ref. 1 for the Frank elastic constants. The temperature dependence of the elastic constants may be obtained from the temperature dependence of the intensity of the scattered light. In Fig. 1 we plot kT divided by the intensity for each configuration. The effect of the temperature variation of the refractive indices on the wave vectors, reflection at the liquid-crystal surface and the geometrical factors f_1 and f_3 has been corrected for, and the results normalized to unity at 286°K. The resulting plot gives the temperature dependence of the appropriate elastic constant divided by Q^2. Except for a slight variation for configuration (a), which is probably a result of the temperature dependence of the relatively large admixture of q_1, the elas-

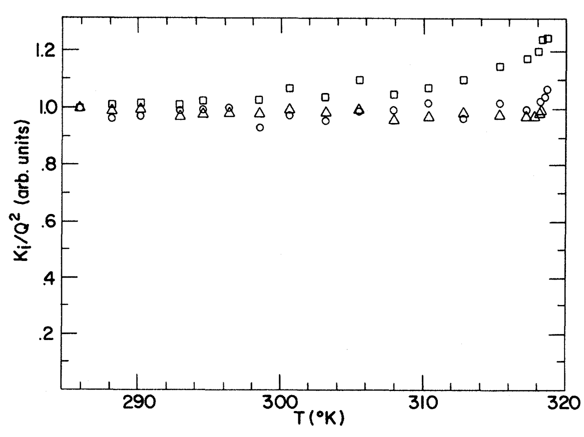

FIG. 1. The temperature dependence of normalized light intensity scattered by fluctuations in the nematic phase of MBBA. The squares, triangles, and circles correspond, respectively, to the experimental configurations (a), (b), and (c) discussed in the text.

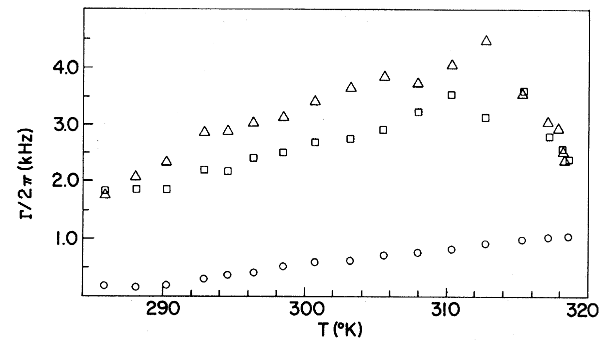

FIG. 2. Linewidth of the Lorentzian spectrum of light scattered by fluctuations in the nematic phase of MBBA. The squares, triangles, and circles correspond to experimental configurations (a), (b), and (c) discussed in the text.

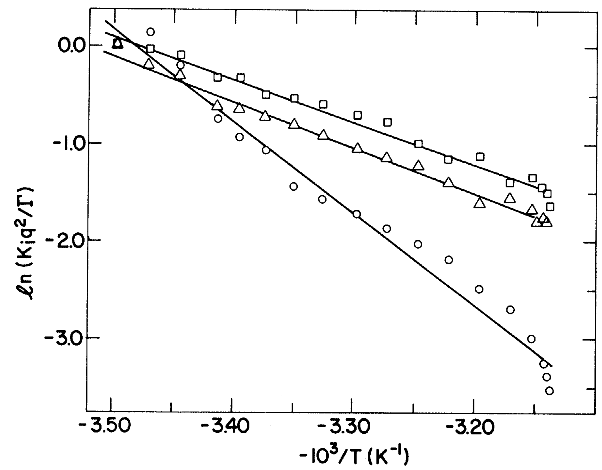

FIG. 3. Temperature dependence of the viscosity coefficients for damping of director motions in the Orsay model. The squares, triangles, and circles correspond to η_a, η_b, and η_c, respectively [see Eq. (4) of text].

tic constants are proportional to Q^2 within experimental errors. This is a striking confirmation of Saupe's theory[8] of the dependence of the Frank elastic constants on order parameter.

The authors of Ref. 2 do not give the scattering cross sections for their model, but we may obtain them as follows: When the displacements u_i are zero, the MPS 3 axis must coincide with the optic axis (director) of the liquid crystal. For small displacements, the changes we observe in the dielectric constant will, therefore, be $\delta\epsilon_{3j} = Q\Delta\epsilon(A\partial_3 u_j + B\partial_j u_3)$ where $j=1$ or 2. The intensity of the scattered light for our three configurations is proportional to

$$\langle \delta\epsilon_{if}^2(\vec{q})\rangle = Q^2\Delta\epsilon^2 A^2 (i_3 f_1)^2 kT/(M_5 q_3^2), \tag{3a}$$

$$= Q^2\Delta\epsilon^2 A^2 (i_2 f_3)^2 kT/(M_5 q_3^2), \tag{3b}$$

$$= Q^2\Delta\epsilon^2 B^2 (i_3 f_1)^2 kT/(L_5 q_1^2). \tag{3c}$$

Here we used the notation of Ref. 2 for the displacements and elastic constants.

The linewidth of the scattered light is determined by the decay times of the fluctuations. From the Orsay model, we calculate the linewidth for our configurations:

$$\Gamma_a = K_a(\vec{q})/\eta_a = K_{33} q_3^2/\eta_B, \tag{4a}$$

$$\Gamma_b = K_b(\vec{q})/\eta_b = K_{33} q_3^2/\eta_B, \tag{4b}$$

$$\Gamma_c = K_c(\vec{q})/\eta_c = K_{22} q_1^2/\gamma_1, \tag{4c}$$

where $\eta_B = \gamma_1 - \alpha_2(\gamma_2 - \gamma_1)/\alpha$. From the MPS model, we obtain

$$\Gamma_a = M_5 q_3^2/\eta_5, \tag{5a}$$

$$\Gamma_b = M_5 q_3^2/\eta_5, \tag{5b}$$

$$\Gamma_c = L_5 q_1^2/\eta_5. \tag{5c}$$

Here $\Gamma/2\pi$ is the half-width at half-maximum of the scattered spectrum, and the notation of the original references[6,2] is used for the elastic and viscosity coefficients.

In Fig. 2 we show the measured temperature dependence of $\Gamma/2\pi$ for each configuration. Both theories predict equal linewidths for (a) and (b) and agree with our results. It is also clear that the MPS elastic constants M_5 and L_5 must have different temperature dependences to be consistent with our data.

We may use our measurements to deduce the temperature dependence of the viscosity coefficients η_B and γ_1. Our intensity measurements show the Frank elastic constants are proportional to Q^2. We combined the temperature dependence of our linewidths with that of Q (known from index-of-refraction measurements[11]) to obtain the temperature dependence of η_a, η_b, and η_c as shown in Fig. 3. Since $\partial \ln\eta/\partial(1/T)$ is constant for normal liquids at constant pressure,[12] we plot in Fig. 3 $\ln\eta$ vs $1/T$ for each configuration. For most liquids $R\partial\ln\eta/\partial(1/T)$, where $R = 1.987$ cal °K^{-1} mole^{-1}, lies in the range 1.5 to 5 kcal/mole and can be up to 10 kcal/mole for associated liquids or those with elongated molecules. We find $\partial\ln\eta_b/\partial(1/T) = 8.6 \pm 0.4$ and $\partial\ln\eta_c/\partial(1/T) = 18.5 \pm 0.1$ kcal/mole. Since these viscosities (in the Orsay model) are combinations of the Leslie coefficients which presumably depend upon the order parameter Q, the large temperature dependences are not surprising. It is interesting that $\ln\eta$ remains a roughly linear function of $1/T$.

We turn now to a discussion of how the normal modes of the MPS model couple to the dielectric constant. We noted above that L_5 and M_5 must vary differently with temperature for the model to be consistent with our linewidth measurements. Since the intensity of the scattered light is proportional to Q^2A^2/M_5 or Q^2B^2/L_5 [see Eq. (3)] and is independent of temperature, we conclude that A and B also vary differently as the temperature is changed. If the symmetry axis of the MPS model always coincided exactly with the optic axis one would have $A = -B = 1$. This is not consistent with our measurements. We see, therefore, that the symmetry axis of MPS coincides with the optic axis only in an unstrained liquid crystal, and that the optic axis is coupled to the displacements u_i through the parameters A and B.

We conclude that the Orsay theory of liquid crystals is consistent with our observations. The MPS theory begins with a conceptually attractive approach, but in its present form is not complete. Unfortunately, to obtain the temperature (and frequency) dependence of the coupling between the optic axis and the strains it appears necessary to introduce a separate equation of motion for the optic axis. The theory then loses the attractive simplicity which distinguished it from earlier approaches.

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Dynamic Stabilization of Drift Waves by ac Electric Fields

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Experimental results are reported on the dynamic stabilization of drift waves in a Q-machine plasma by means of ac electric fields parallel to the magnetic field. Appreciable stabilization has been obtained both at ac frequencies around the wave frequency and at very high ac frequencies.

Dynamic stabilization effects of drift waves by ac electric fields parallel to the confining magnetic field have been theoretically predicted both at very high frequencies (above the ion plasma frequency) and high amplitudes,[1] and at frequencies comparable with the frequency of the waves and smaller amplitudes,[2,3] both in collisionless and collisional plasmas. Very recent experimental results have been reported[4,5] on high-frequency stabilization of drift waves in a discharge type plasma.

This Letter reports results of an experimental investigation on the dynamic stabilization of drift waves by ac electric fields, in a very large range of frequencies, in a fully ionized Q-machine plasma. It has been found that ac fields are effective in reducing the amplitude of the waves both at frequencies around the wave frequencies (~2 kHz) and at very high frequencies (~1 MHz).

The experiments were performed in the Fras-