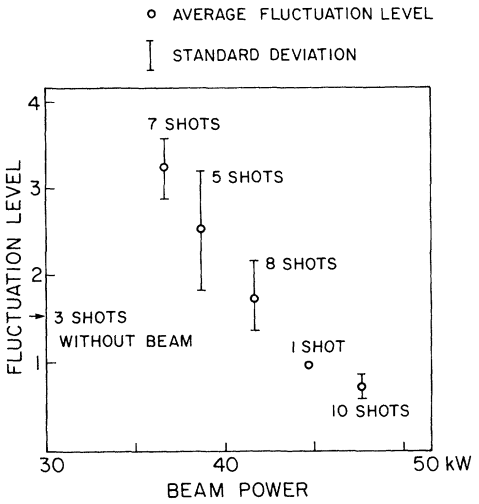

FIG. 4. High-frequency signal of floating-potential probe E-beam power.

cay. The enhanced fluctuation levels for beam powers < 42 kW are believed to be due to a combination of bulk heating of the electrons and too small a population of hot electrons. Langmuir-probe measurements near the plasma edge indicate an increase of T_e from 10 to 17 eV with E-beam injection.

We do not observe the sharp drop in floating potential that Ioffe *et al.*[3] report of their stabilization by electron-cyclotron heating. Although we have not yet measured the axial distribution of hot electrons, produced by the electron beam, we doubt that they are concentrated at the midplane. A possible stabilizing mechanism suggested by Baldwin[10] is that the hot electrons may have have a high enough ∇B precession to equal the diamagnetic drift speed and may thus short circuit the fields that drive the instability.

In summary, we report a new, and technically simple way of stabilizing ion cyclotron loss-cone instabilities. It may have important applications either used alone, or in conjunction with warm plasma injection in the stabilization of mirror-confined thermonulcear plasma.

This work was supported by the U. S. Department of Energy, Grant No. EG-77-01-4107.

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Light-Scattering Study of Two-Dimensional Molecular-Orientation Fluctuations in a Freely Suspended Ferroelectric Liquid-Crystal Film

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(Received 3 January 1978)

The dispersion relations for molecular-orientation fluctuations in a freely suspended ferroelectric smectic-C liquid-crystal film have been determined by inelastic light scattering. The smectic-C film geometry appears to be ideal for the experimental study of the space-time behavior of a two-dimensional orientational field.

The classical X-Y model is a two-dimensional model magnetic system in which spins are confined to lie in a common plane but are free to rotate in that plane under mutual interactions. It has received theoretical attention recently because in an X-Y system having only nearest-neighbor interactions, thermal orientation fluctuations suppress long-range orientational order and disallow the ordinary kind of order-disorder transition.[1] Instead a phase transition of contin-

uous order is predicted in which the high- and low-temperature states are characterized by different degrees of local order[2] and which have temperature-dependent critical exponents for $T<T_c$ and an exponentially diverging susceptibility for $T>T_c$.[3] These predictions have stimulated interest in relevant physical systems.

In this Letter we present the first experiments on a physical liquid-crystal system, a freely suspended smectic-C liquid-crystal film, which should be realistically describable as a classical continuous X-Y model. We report the results of an inelastic-light-scattering study of the amplitude and the dynamics of molecular-orientation fluctuations (spin waves) in a free smectic-C film of the optically active compound octyloxybenzylidene-β-methyl-butyl aniline (8O.5*). The experimental arrangement is sketched in Fig. 1(a).

Films were drawn in an inert atmosphere over a rectangular hole (10 mm×3 mm) in a glass microscope cover slip. As shown in Fig. 1(b), smectic films are quantized in thickness, consisting of an integral number of smectic layers, $N\geq 2$, each ~25 Å thick. Films are stable against rupture for $N\geq 2$ and are made by repeated spreading of new films until one of the desired thickness, as determined by optical reflectivity is obtained. Because of the fluid layered structure, these films are homogeneous over their area with respect to thickness and the average molecular tilt angle θ_T. Molecular orientations are characterized by the unit vector director field $\hat{n}(x,y)$ indicating the local orientation angle φ about a normal to the film plane. A free smectic-C film is a homogeneous two-dimensional physical system with cooperative interactions that is completely orientationally degenerate—in liquid-crystal parlance, a two-dimensional polar nematic.[4]

Smectic-C films containing optically active molecules such as 8O.5* will be ferroelectric, i.e., they possess a nonzero electric dipole moment per unit area, $\vec{P}_0(x,y)=P_0[\hat{n}(x,y)\times\hat{z}]$ [see Fig. 1(c)].[5] Making P_0 nonzero has two important effects on the properties of free films. Firstly,

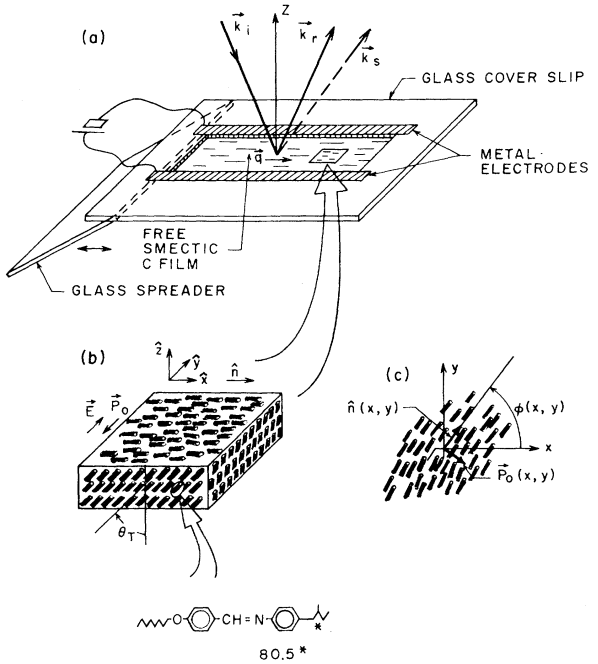

FIG. 1. Free smectic-C film geometry. (a) Film holder showing incident ($\vec{k}_i$), reflected ($\vec{k}_r$), and scattered ($\vec{k}_s$) light directions and the orienting electrodes. (b) Structure of a three-layer film showing molecular projections and a molecule of 8O.5*. (c) A portion of a film showing the molecular projections in the x-y plane, the director $\hat{n}(x,y)$, polarization $\vec{P}_0(x,y)$, and $\varphi(x,y)$.

a weak electric field ($E\sim 10$ V/cm) applied in the film plane will tend to orient $\hat{n}$ so as to minimize $-\vec{P}_0\cdot\vec{E}$, permitting the study of single-domain samples. Secondly, molecular interactions are no longer solely nearest neighbor as orientation fluctuations will produce polarization charge per unit area $\sigma_P=-\nabla\cdot\vec{P}_0$ and therefore long-range electrostatic forces. The relative magnitude of the long- and short-range interactions can be readily adjusted by manipulating P_0 via sample composition or temperature—an additional useful feature of these systems.

Orientational fluctuation dynamics may be calculated from the free energy of distortion of $\hat{n}(x,y)$ in the presence of an external electric field, $\vec{E}$:

$$F=\int dA\{\tfrac{1}{2}K_s|\nabla\cdot\hat{n}|^2+\tfrac{1}{2}K_b|\nabla\times\hat{n}|^2-\vec{P}_0\cdot\vec{E}+\int dA'(\nabla\cdot\vec{P}_0)(\nabla\cdot\vec{P}_0)'/2|\vec{r}-\vec{r}'|\}. \quad (1)$$

Here, $\nabla=\hat{x}\partial/\partial x+\hat{y}$, K_s and K_b are the two-dimensional splay and bend elastic constants, and the last term is the free-energy density arising from the polarization charge (dipole-dipole) interaction in three dimensions. Since $\nabla\cdot\vec{P}_0=P_0\nabla\cdot(\hat{n}\times z)=P_0|\nabla\times\hat{n}|$, this free-energy term will be associated only with bend distortions. Neglecting surface effects and assuming a nematiclike behavior in the elastic energy of deformation, the two-dimensional elastic constants K_s and K_b can be related to the bulk Frank

elastic constants K_{11} (splay), K_{22} (twist), and K_{33} (bend):

$$K_s = hK_{11}\sin^2\theta_T,$$
$$K_b = h[K_{22}\sin^2\theta_T\cos^2\theta_T + K_{33}\sin^4\theta_T], \quad (2)$$

where θ_T is the tilt angle. Also $\vec{P}_0 = h\vec{P}$ where $\vec{P}$ is the bulk polarization per unit volume and h is the film thickness.

Qualitative observation has been made of the temperature dependence of orientational order in the films. At temperatures where the bulk is in the smectic-C phase (37.5°C$<T<$46.8°C for 8O.5*), polarized reflection microscopy[6] shows that films are anisotropic and can be ordered by application of an electric field as expected. As the temperature is raised, an abrupt transition to an apparently isotropic state is observed. In thick films this temperature is close to the bulk $A \rightarrow C$ temperature but increases with decreasing N to ~60°C for $N=5$. For $N<5$ film rupture occurs at temperatures below this transition in 8O.5*, but it is observable in other compounds for N down to $N=2$. The order-disorder transition in the X-Y model is predicted to occur when[7]

$$K_{s,b} = (2/\pi)k_B T. \quad (3)$$

Applying Eq. (2) with $K_{\text{bulk}} \sim 5\times10^{-7}$ erg/cm, $N = 3(h = 75\times10^{-8}$ cm), $\theta_T = 30°$ we have $K_{s,b} \sim 2k_B T$ at temperatures well down into the smectic-C phase. Since θ_T decreases rapidly with increasing T, Eq. (3) can surely be satisfied. It seems likely, therefore, that the observed transition is that predicted for the X-Y model. The experiments reported here were carried out at $T = 54$°C for $2 \leq N \leq 6$ in the "ordered" phase.

For an aligned system with the director in the x direction, the Fourier transformed free energy can be written in terms of φ_{s,q_y} and φ_{b,q_x}, the amplitudes of the splay and bend normal modes with wave vector q_y and q_x, respectively. Using the equipartition theorem, the mean-square amplitudes of these fluctuations will be

$$\langle \varphi_{s,q_y}{}^2 \rangle = k_B T/(K_s q_y{}^2 + P_0 E), \quad (4a)$$

$$\langle \varphi_{b,q_x}{}^2 \rangle = k_B T/(K_b q_x{}^2 + 2\pi P_0{}^2 |q_x| + P_0 E). \quad (4b)$$

Assuming viscous damping constants η_s and η_b for the two modes respectively, the correlation functions are of the form $\langle \varphi(t)\varphi(0)\rangle = \langle \varphi^2 \rangle e^{-\Gamma t}$ where

$$\Gamma_s = \tau_s{}^{-1} = (K_s q_y{}^2 + P_0 E)/\eta_s, \quad (5a)$$

$$\Gamma_b = \tau_b{}^{-1} = (K_b q_x{}^2 + 2\pi P_0{}^2 |q_x| + P_0 E)/\eta_b. \quad (5b)$$

Note that the polarization charge interaction contributes to the bend elastic energy only [Eqs. (4b) and (5b)]. Both modes give rise to depolarized light scattering[4] and can be observed individually by properly orienting the scattering wave vector relative to the director ($\vec{q} \,||\, \hat{n}$ for bend, $\vec{q} \perp \hat{n}$ for splay).

To measure the splay or bend correlation functions, laser light (5145 Å at ~200 mW) was focused onto the film which was mounted on a rotation stage and placed inside a temperature-controlled oven.[6] Since only the scattering wave vector component in the plane of the film is experimentally relevant, its magnitude could be varied by simply rotating the sample while keeping the incident and scattered directions fixed. Metal electrodes were evaporated on the two longer sides of the hole [Fig. 1(a)] and a typical voltage of 2 V was applied across the electrodes to align the director. The depolarized scattered light was imaged onto a photomultiplier so that only one coherence area was collected. The output of the photomultiplier was analyzed using a 114-channel digital-clipped autocorrelator. The alignment of the director field was checked by polarized reflection microscopy before and after each experiment. Both the splay and bend modes gave very good single-exponential correlation functions and were fitted numerically to give the decay times of the modes.

Figure 2(a) shows the experimental results for the splay mode where Γ_s is plotted against the square of the scattering wave vector for films with 2, 3, 4, and 6 layers. All of these films show a linear dependence of Γ_s on q^2, consistent with Eq. (5a). The q-independent term $(P_0 E/\eta_s)$ is too small compared with the q^2 term to be observed in these data. From the slope of each line, the value of K_s/η_s can be obtained. It can be seen that there is a systematic increase in K_s/η_s as N decreases. Figure 2(a) also shows the inverse of the scattering-volume–corrected scattered light intensity measured for $N=3$ as a function of q^2. Since the scattered light intensity is proportional to the mean-square amplitude of the fluctuation $\langle \varphi_{s,q}{}^2 \rangle$, the q^2 dependence observed is just as predicted by Eq. (4a).

The results for the bend mode are shown in Fig. 2(b). The plot of $\Gamma_{b/q}$ versus q has the form predicted in Eq. (4b) with the slope of the curve giving K_b/η_b and the intercept $2\pi P_0{}^2/\eta_b$. Again the $P_0 E$ term is too small to be observed. Figure 2(b) shows the results for the scattered intensity and the same nonzero intercept is present. As

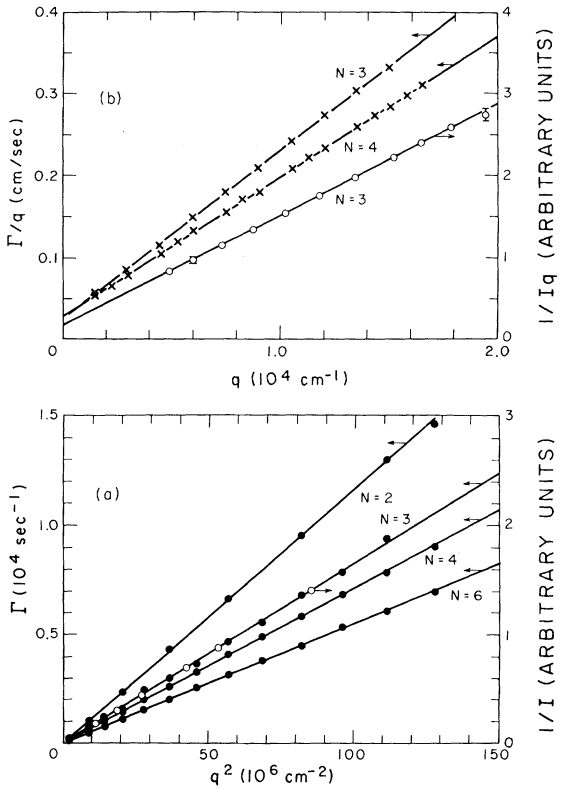

FIG. 2. (a) Decay rate (●) and intensity (○) data for the splay mode. Values of K_s/η_s (10^{-5} cm^2/sec) from slopes ($\pm \sim 2\%$): ($N=2$) 11.7; ($N=3$) 8.3; ($N=4$) 7.1; ($N=6$) 5.5. (b) Decay rate (×) and intensity (○) data for the bend mode. Values of K_b/η_s(10^{-5} cm^2/sec), from slopes ($\pm 2\%$): ($N=3$) 2.05; ($N=4$) 1.70. Values of K_b/P_0^2 (cgs) from intercepts: $(0.5\pm 0.02)\times 10^{-2}$ ($N=3$); $(4.0\pm 0.2)\times 10^{-3}$ ($N=4$); $K_b/P_0^2 = (0.8\pm 0.2)\times 10^{-2}$ ($N=3$, intensity data).

for the splay mode, K_b/η_b is observed to increase with decreasing N, whereas P_0^2/η_b does not show much thickness dependence.

The ratios K_b/η_b, K_s/η_s, and K_b/P_0^2 obtained from the data are given in Fig. 2. The measured K/η are somewhat larger than that extrapolated from typical bulk values ($\sim 10^{-5}$ cm^2/sec). This is consistent with the increase in these ratios with decreasing N and possibly indicates larger tilt angles and therefore larger two-dimensional elastic constants for the surface layers. The expected dependence of the film parameters on thickness is difficult to assess, because the tilt angle θ_T is itself strongly thickness dependent, generally increasing with decreasing N. An understanding of the film at small N will require a determination of $\theta_T(N)$ as well as $K_{s,b}(N)$, $\eta_{s,b}(N)$, and $P_0(N)$. We are currently carrying out these experiments, using ellipsometry to determine θ_T and larger electric fields combined with the exploitation of the form of Eqs. (4b) and (5b) to determine $K_{s,b}$, $\eta_{s,b}$, and P_0 absolutely.[9] The latter effort, if successful, offers the exciting possibility of testing Eq. (3) and studying the effect of the polarization on the order-disorder transition.[8]

To summarize, we have presented the first experiments on a freely suspended smectic-C film, an orientationally degenerate physical X-Y system with a variable, dipole-dipole interaction. We have determined via light scattering the dispersion relations for the two orientational fluctuation modes demonstrating the existence of a ferroelectric contribution to the bend-mode energy. Agreement with theory indicates that reliable information concerning the orientation dynamics and film parameters can be obtained, and that detailed studies of the space-time behavior of a physical X-Y system are now possible.

We would like to acknowledge many fruitful conversations with P. S. Pershan concerning this research and the following contract support: National Science Foundation Contracts No. 76-22452 and and No. DMR76-01111, and the Joint Services Electronics Program.

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[9]Work in progress at Harvard University, in collaboration with Mr. C. Rosenblatt.