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Bend periodic distortion of the texture in nematic lyotropic liquid crystals with and without ferrofluid

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The creation of a bend periodic distortion by the action of magnetic fields (**H**) in a nematic lyotropic liquid crystal is discussed. The wavelength of this distortion (P), in the limit of small deformations, allows the measurement of K_3/χ_a, where K_3 is the bend elastic constant and χ_a the anisotropy of magnetic susceptibility. In this limit, the dependence of P^{-2} with H^2 presents a linear behavior, predicted by the continuum elastic theory. The relation between P and H also allows us to determine an "effective field" in nematic samples doped with ferrofluid.

Lyotropic liquid crystals are systems composed by amphiphilic molecules and water in proper temperature and concentration conditions. It is well known[1] that the texture of nematic liquid crystals (thermotropic or lyotropic) can be distorted by magnetic fields (**H**). Two different optically aligned regions (aligned by the action of the field) can be separated by a wall.[1] The phenomenon of the distortion of the director (**n**) by the action of the magnetic field has been extensively studied for both thermotropic[2–4] and lyotropic[5] liquid crystals. More recently, a new periodic distortion of **n** was reported by Lonberg and Meyer.[6] Generally the Fredericksz transition,[1] with the formation of walls, is used to determine the ratio between the elastic constant (K) and the anisotropy of the diamagnetic susceptibility (χ_a). Values of K/χ_a for the thermotropic liquid crystals were extensively measured[1] but for lyotropic liquid crystals there are not many experiments reported.[7] Some of the difficulties of these experiments with lyotropic liquid crystals is to control the orientation of **n** in the surfaces of the sample holders, to define a rubbing direction, and after to apply a high magnetic field (about 10 kG). In this paper, we report an easy method to measure the ratio K_3/χ_a for lyotropic nematic liquid crystals (where K_3 is the bend elastic constant) from a periodic distortion of **n** induced by **H**. The periodic distorted texture is observed in a polarizing microscope. We also discuss the utilization of nematic samples doped with ferrofluids[8] in this experiment. The doping reduces the strength of H required to orient the mesophases.[8]

The sample used is a mixture of potassium laurate (29.1 wt. %), decanol (6.4 wt. %), and water (64.5 wt. %) prepared according to the conventional procedure. The phase sequence determined by optical and conoscopic observations is the following: isotropic (14.9 ± 0.5 °C) → calamitic nematic (47.2 °C) → isotropic. The experiment is performed with the sample in the calamitic nematic phase. Nematic samples are encapsuled in flat glass microslides (thickness is D, width is 2.5 mm, length is 2 cm) of different thicknesses (100, 200 μm). The laboratory frame axes are defined in Fig. 1: x is the long axis of the microslide and z is the axis normal to the biggest surface of the microslide. The magnetic field (Varian electromagnet) of strength from 2 to 17.5 kG is applied in the x-y plane and the optical microscopic observations (Wild-orthoplan-pol microscope) are made along the z axis. Samples are placed in a temperature controlled stage which maintains the atmosphere around the microslide at (25 ± 1) °C. The temperature of the microslide is measured during the experiment with high magnetic fields (H > 10 kG) to verify a possible heating of the sample produced by the electromagnet. The time necessary to form a bend distortion wall in a nematic sample in the presence of a high magnetic field is about 2 min. During this time, we observed that the temperature of the sample increases less than 0.2 °C due to the electromagnet heating. For the study of ferrofluid-doped nematic samples, **H** is obtained from small permanent magnets with strength from 100 to 1300 G. A water-base ferrofluid from Ferrofluidics Corp.[9] is used and the concentrations of magnetic grains in the sample are $C = 10^{12}$ grains/cm^3 and $2C$. A specific surface treatment procedure is not applied to the glass surfaces of the microslide. In this condition, **n** can rotate about the z axis. In fact, the geometry of the microslide favors **n** to be parallel to x, as discussed in Ref. 10 (see Appendix).

Initially, a magnetic field of 17 kG is applied along the x axis of the sample (planar geometry, **n** parallel to x) to obtain a well-oriented sample. After, the field along x is switched off and a fixed-strength-controlled magnetic field is applied along the y axis. The texture observed in a polarizing microscope shows a periodic distortion of **n** with walls formed in the direction of **H** [Fig. 2(a)].

The uniformity of the texture over large areas is critically dependent on the initial alignment of **n** parallel to the x axis. Irregularities on the inner surfaces of the mi-

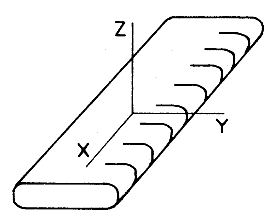

FIG. 1. Sketch of microslide cell with the laboratory frame axes x, y, and z.

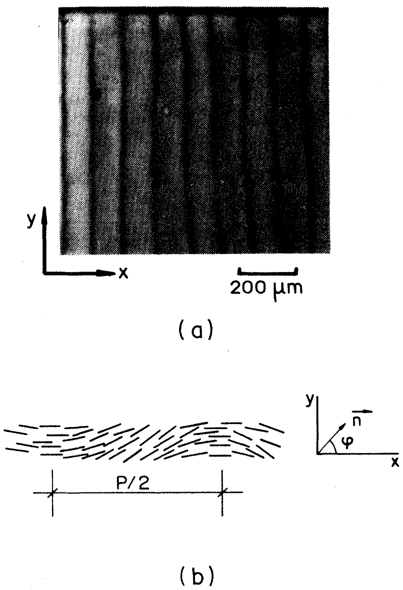

FIG. 2. (a) Lyotropic nematic phase in a microslide 200 μm thick between crossed polarizers. Magnetic field (5.3 kG) along the y axis. y axis at 0° of the light polarizing direction. (b) Sketch of the bend periodic distortion: P is the wavelength and ϕ is the polar angle between $\mathbf{n}$ and the x axis.

croslide and inhomogeneities of the magnetic field can also produce deformations on the texture. These problems can be avoided by a careful experimental manipulation. The periodic distortion of the texture remains stable for several hours. The relaxation is very slow, and after about 5 h in the presence of the magnetic field, the walls begin the process of formation of closed elliptical loops.[4] Figure 2(b) presents a sketch of the periodic deformation of $\mathbf{n}$ in the x-y plane and introduces the polar angle ϕ between $\mathbf{n}$ and the x axis.

The expression of the free-energy density taking into account the elastic deformations and the magnetic field coupling is[1]

$$F=\tfrac{1}{2}\{K_1(\nabla\cdot\mathbf{n})^2+K_2[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2+K_3[\mathbf{n}\times(\nabla\times\mathbf{n})]^2-\chi_a(\mathbf{n}\cdot\mathbf{H})^2\}\,, \quad (1)$$

where K_1 and K_2 are the splay and twist elastic constants.

The geometry of the experiment imposes that

$$\mathbf{n}=\begin{cases} n_x=\sin\phi\,, \\ n_y=\cos\phi, \quad \phi=\phi(x)\,, \\ n_z=0\,. \end{cases} \quad (2)$$

Using the usual procedure to minimize the free energy, the Euler equation for ϕ is

$$\left[k_3\left(\frac{\partial\phi}{\partial x}\right)^2-\xi^{-2}\right]\sin\phi\cos\phi-(1+k_3\cos^2\phi)\frac{\partial^2\phi}{\partial x^2}=0\,, \quad (3)$$

where $k_3=(K_3/K_1)-1$ and ξ is the usual magnetic coherence length defined by $\xi^2=K_1/\chi_a H^2$. Taking typical values of K_3, K_1, and χ_a measured[7,11,12] with lyotropics and the strength of $\mathbf{H}$ about 10^4 G, the term $k_3(\partial\phi/\partial x)^2$ can be neglected by comparing to ξ^{-2}. So, in the limit of small deformations, Eq. (3) can be written as

$$\frac{\partial^2\phi}{\partial x^2}+\frac{\phi}{\xi^2(1+k_3)}=0\,. \quad (4)$$

The solution of Eq. (4) is a periodic function with a wavelength P given by

$$P^{-2}=\frac{\chi_a}{4\pi^2K_3}H^2\,. \quad (5)$$

Figure 3 shows the experimental results of P^{-2} as a function of H^2. P does not seem to depend on sample thickness. Each value of P presented in Fig. 3 is a mean value obtained from a series of several reproducible experiments. Sometimes the observed texture is not uniform over large areas, probably due to irregularities on the inner surfaces of the microslide. In this case, no measurement of P is performed and a new sample is prepared. It can be observed in Fig. 3 that the linear behavior is obtained up to fields of about 7 kG. This fact indicates that the approximation of small distortions is limited to fields smaller than 7 kG. For higher magnetic fields, P^{-2} deviates from the linear behavior predicted by Eq. (5). This fact indicates that the approximation made in Eq. (3) to obtain Eq. (4) cannot be done.

The slope of the linear region of P^{-2} as a function of H^2 gives the ratio K_3/χ_a which, for our lyotropic systems is $(3.0\pm0.3)\times10^2$ dynes. There is no value in the literature of this ratio for the potassium laurate system. Haven,

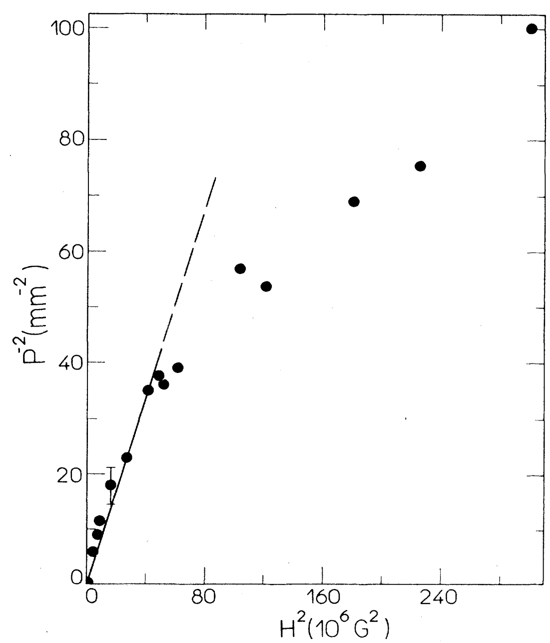

FIG. 3. Dependence of the wavelength of the periodic distortion (P) with the applied magnetic field ($\mathbf{H}$). The linear behavior is described by Eq. (5). Lyotropic calamitic nematic phase without the ferrofluid doping. The typical error is shown.

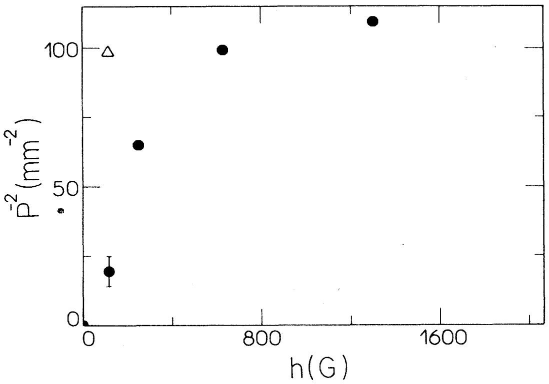

FIG. 4. Dependence of the wavelength of the periodic distortion (P) with the applied magnetic field (h). Lyotropic calamitic nematic phase doped with ferrofluid: ●, concentration of the doping $C=10^{12}$ grains/cm^3; △, concentration of the doping $2C$. The typical error is shown.

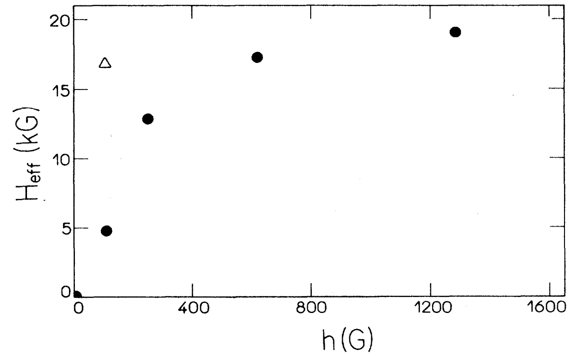

FIG. 5. Effective field H_{eff} as a function of the magnetic field h. Lyotropic calamitic nematic phase doped with ferrofluid: ●, concentration of the doping C; △, concentration of the doping $2C$.

Harmitage, and Saupe[7] reported values of K/χ_a measured in the mixture NH_4Cl-decylammoniumchloride-H_2O, very similar to ours.

To verify the influence of the ferrofluid doping in the bend periodic distortion of **n** we studied samples with two different concentrations of magnetic particles, C and $2C$. Initially samples are oriented with $H=3$ kG along the x axis (**n** parallel to x) and then a small magnetic field $\mathbf{h}(100 \leq h \leq 1300$ G) is applied along the y axis. As in the case of strong fields with samples without the ferrofluid doping, a periodic distortion of the texture is observed immediately after the field is switched on. Figure 4 shows P^{-2} as a function of h. The values of P presented in Fig. 4 are also obtained from a series of reproducible experiments. Concerning the uniformity of the texture over large areas and the relaxation process, no differences were detected between the samples with and without the ferrofluid doping. Comparing the values of P obtained with the doped samples, with the wavelength measured in samples without the doping one observes that the ferrofluid, at this concentration, enhances the distortion by the magnetic field. Using Fig. 3 as a "calibration curve," we obtain an effective field H_{eff}. H_{eff} is the field necessary to generate the same periodic deformation in samples without the ferrofluid doping. Figure 5 shows H_{eff} as a function of h. It is observed a saturation behavior of H_{eff} with h, indicating that the amplification effect of the doping is much more sensitive to variations of h for small values of h. This amplification effect is strongly dependent on the concentration of the ferrofluid doping. With samples doped with $2C$, H_{eff} for $h=140$ G is about 17 kG (with samples doped with C, $H_{eff} \simeq 5$ kG). In a previous work[13] the orientational distortion of the nematic matrix was shown to be dependent of the concentration of ferrofluid. In liquid-crystal samples doped with ferrofluid (concentration larger than the minimum concentration[14]), Brochard and de Gennes[8] predicted a gain in the coupling of order 10^3. Our experimental results (Fig. 5) indicate that this gain in the field (amplification factor) is strongly dependent of the concentration of the doping and, for a given concentration, the gain depends also on the applied field (h).

In conclusion, we present the formation (by the action of the magnetic field) of a periodic distorted texture in a nematic lyotropic liquid crystal. The wavelength of this periodic texture, in the limit of small deformations, allows the measurement of the ratio K_3/χ_a. The relation between the wavelength and H allows us also to determine an effective field in nematic samples doped with ferrofluid.

It is a pleasure to thank Dr. Ph. Martinot-Lagarde for very helpful discussions.

APPENDIX: THE GEOMETRY OF THE MICROSLIDE AND THE DIRECTOR n

A small magnetic field ($h \leq 1.3$ kG) was applied along the y axis in a previously oriented (**n** parallel to the x axis) nematic sample without the ferrofluid doping for 1 h and no distortion of the texture was observed. As discussed in Ref. 10, the geometry of the microslide favors the orientation of **n** parallel to the x axis. The existence of this favored direction to **n** can be understood taking into account that the microslide may be sketched (Fig. 1) as two half-cylindrical boundaries (diameter is D, length is 2 cm) tied by two plane surfaces (length is 2 cm, width is 2.5 mm). At the plane surfaces **n** orients parallel to the surfaces, without a particular direction. At the cylindrical boundaries, however, the condition of minimum free energy gives for the director the topological configuration of **n** parallel to the x axis (Fig. 1). These fixed directions at the boundaries break the degeneracy of **n** at the bulk. Our experiment indicates that for small magnetic fields applied in samples without the ferrofluid doping (**h** parallel to the y axis), **n** remains parallel to the x axis. In other words, the statement that **n** can rotate about the z axis is not verified in this condition. For magnetic fields $H \geq 2$ kG the bend periodic distortion of the texture is observed, indicating that the magnetic coupling between **n** and **H** is bigger than the mechanical coupling between **n** and the boundaries of the microslide. In this condition, the statement that **n** can rotate about the z axis is verified and Eqs. (4) and (5) give a good description of the phenomenon.

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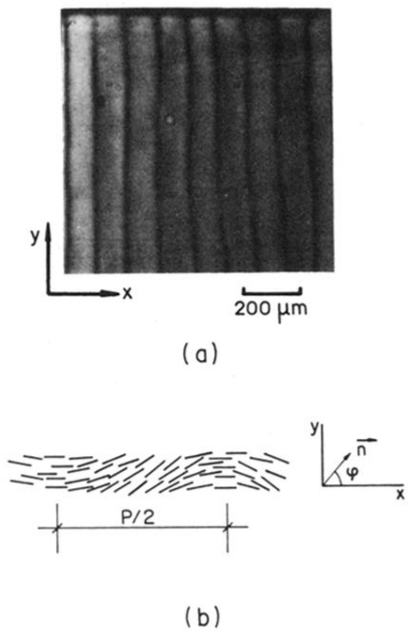

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