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Magnetic Field Dependence of Laser Light Scattering through CBOOA in Nematic Phase

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In the nematic phase of homeotropically aligned liquid crystals which have the smectic A phase at lower temperature, interference rings have been observed above some threshold magnetic field H_c which nearly coincides with the occurrence of Freedericksz transition, only for the light polarized parallel to the direction of magnetic field. This interference ring disappears with increasing magnetic field above second threshold H_s. The origin of these interference rings is explained tentatively by a periodic deformation of bulk directors.

The bend elastic constant of CBOOA estimated from the measurements of the interference ring as a function of temperature, diverges with the critical exponent (0.5 ± 0.02) near the smectic A–nematic transition temperature and agrees well with a recent suggestion of McMillan based on the mean field theory.

1 INTRODUCTION

Recently there has been considerable interest in the critical phenomena of the second order smectic A–nematic phase transition for the liquid crystal CBOOA. Various theories and experiments on the above critical phenomena have been reported extensively.[1–6] However, there remain yet unresolved phenomena. In this paper the experimental results on the critical deformation under the magnetic field above the smectic A–nematic transition temperature in CBOOA will be presented.

Observation of the interference rings of scattered light in a certain magnetic field range just above the threshold of the Freedericksz transition will be reported and a simplified model which explains these experimental observations will be proposed. Appearance of interference rings by applying an electric field in the homeotropic cell of a nematic liquid crystal has been reported[7]. and analyzed under the assumption of the spacial periodicity of the bulk director deformation.

Since it has been suggested that the smectic A–nematic phase transition in CBOOA could be second order, various theories[1,2] and experiments[3–6] concerning with the pretransitional effect at the second order smectic A–nematic transition point have been reported. Nevertheless, the reported critical exponents of both the bend and twist elastic constants do not coincide among authors. In this paper the temperature dependence of the bend elastic constant is also evaluated directly from the temperature dependence of the critical magnetic field of the observed interference rings. The estimated value of the critical exponent as a function of temperature is in good agreement with a recent suggestion of McMillan[2] based on the mean field theory.

2 EXPERIMENTAL PROCEDURE

Figure 1 shows the schematic diagram of the experimental apparatus for the light scattering measurement under the applied magnetic field. Liquid crystal samples were sandwiched between two glass slides separated with teflon spacers of various thicknesses (8–250 μm). Both glass slides were treated with lecithin surfactant in order to obtain perfect homeotropic alignment with the molecular axis perpendicular to the glass surfaces. The sandwiched sample between two glass slides is set in the magnetic field **H** whose direction is parallel ($\mathbf{S} \parallel \mathbf{H}$) or perpendicular ($\mathbf{S} \perp \mathbf{H}$) to the glass surfaces. A He–Ne laser light ($\lambda = 6328$ Å) with its polarization plane $\mathbf{E}_1$ parallel ($\mathbf{E}_1 \parallel \mathbf{H}$) or perpendicular ($\mathbf{E}_1 \perp \mathbf{H}$) to the magnetic field was shined in a direction normal

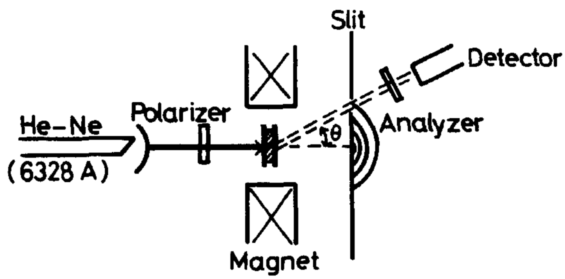

FIGURE 1 Schematic diagram of light scattering measurement under applied magnetic field.

to the sample surface. The forward scattered light was observed by a photomultiplier (HTV-R376) after passing through a pin hole of 500 μm ϕ placed about 1 m apart from the sample. The position of the pin hole and the photodetector were moved within the plane normal to the incident laser beam. The temperature of the sample was controlled within 0.05°C using a SCR thermocontroller of a PID type.

3 RESULTS AND DISCUSSION

We applied a magnetic field to impose magnetic deformation in the nematic phase at a constant temperature. For the configuration of ($\mathbf{S} \parallel \mathbf{E}_1 \parallel \mathbf{H}$), as far as the applied magnetic field H is smaller than the critical value H_c, only a usual uniaxial pattern of the forward scattered light is observed, as shown in Figure 2(a). With an increasing magnetic field, the interference ring suddenly

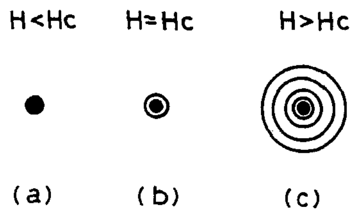

FIGURE 2 Observed interference rings near the critical magnetic field.

appears at a critical magnetic field H_c, as shown in Figure 2(b). This interference ring is a concentric circle with the center at the incident laser beam spot. As shown in Figure 2(c), with increasing magnetic field above H_c, the diameter of the interference ring becomes larger and the second or third ring appears inside the first interference ring. At the magnetic field beyond an upper limit H_s, these interference rings suddenly disappear. These interference rings are stable and reversible for both increasing and decreasing magnetic field in the nematic phase, except for the narrow temperature region around the smectic A-nematic transition. The interference rings are clearly observed only with the light polarized parallel to the direction of the applied magnetic field, but not in the case of the perpendicularly polarized light. The scattered light was also polarized parallel to the magnetic field. The interference ring was not observed in the sample cell placed in the magnetic field whose direction is perpendicular to the glass surface. Figure 3 shows the thickness dependence of the lower critical magnetic field H_c for the appearance of the first interference ring. H_c is inversely proportional to the sample thickness t. However, in the case of t above 200 μm, H_c becomes independent

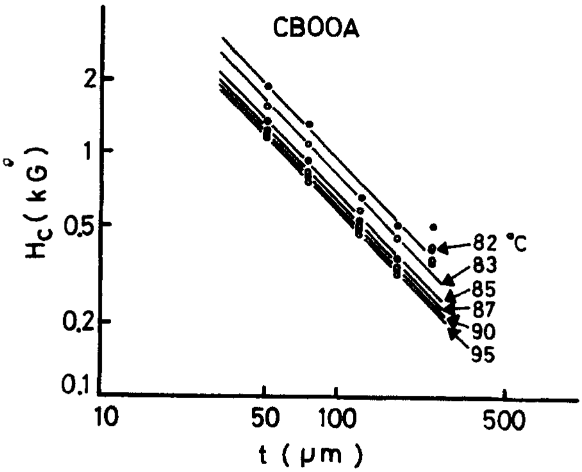

FIGURE 3 Thickness dependence of the critical magnetic field H_c at various temperatures in CBOOA.

of the sample thickness. The threshold magnetic field of the Freedericksz transition is also inversely proportional to the sample thickness. The value of H_c obtained from this experiment also agrees with the reported value[7] of the threshold of the Freedericksz transition. Figure 4 shows the magnetic field dependence of the angle θ of the first interference ring which means the angle between the scattered light direction and the incident laser beam is represented in Figure 1. As already mentioned, the interference rings are

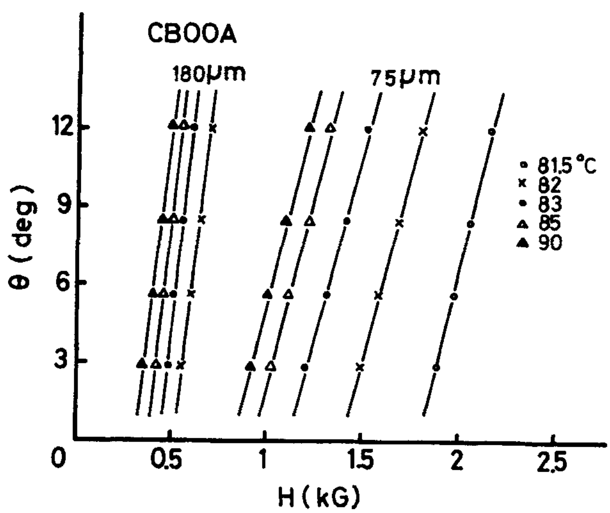

FIGURE 4 Scattering angle of the first interference ring vs applied magnetic field in CBOOA.

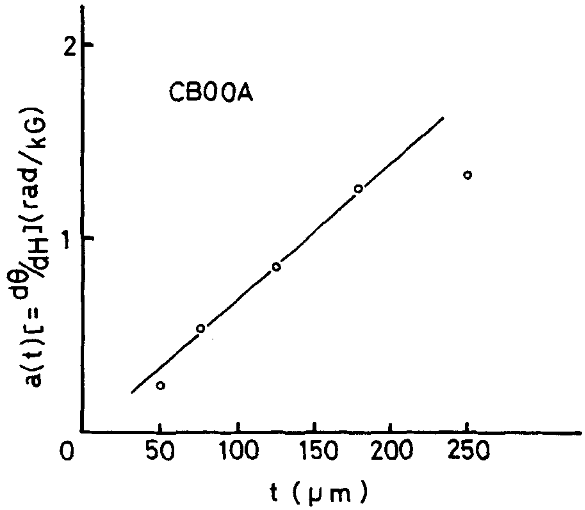

FIGURE 5 $a(t)$ as a function of sample thickness in CBOOA.

concentric circles with the center at the incident beam spot. Therefore the scattering wave vector makes the angle θ with the incident laser beam. The angle, θ, increases linearly with the magnetic field in both samples of 75 μm and 180 μm thickness with the following empirical relation.

$$\theta = a(t)\{H - H_c(t, T)\} \qquad \text{for } H \geq H_c, \tag{1}$$

where $a(t)$ is a function of the sample thickness t and the lower critical magnetic field $H_c(t, T)$ is a function of both t and temperature T. As shown in Figure 5, $a(t)$ increases linearly with increasing sample thickness between 50 μm and 180 μm, and becomes independent of the thickness above 180 μm. $a(t)$ is also insensitive to temperature. These facts may be related with the surface pinning force. The energy due to the surface pinning force seems to be dominant in comparison to the thermal agitation energy within 100 μm from the glass surface. Therefore, in the case of the sample with a thickness above 200 μm, directors near the center position of the sandwiched sample rotate freely in the direction of the applied magnetic field. Figure 6 shows the temperature dependence of the critical magnetic field in the sample with various thicknesses. The slopes of the straight lines give the value 0.26 at 75 μm and 0.24 at other thicknesses. The continuum theory[7] of liquid crystals predicts the next relation between the bend elastic constant K_{33} and the critical magnetic field H_c of the Freedericksz transition.

$$\frac{K_{33}}{\Delta\chi} = \left(\frac{H_c t}{\pi}\right)^2, \tag{2}$$

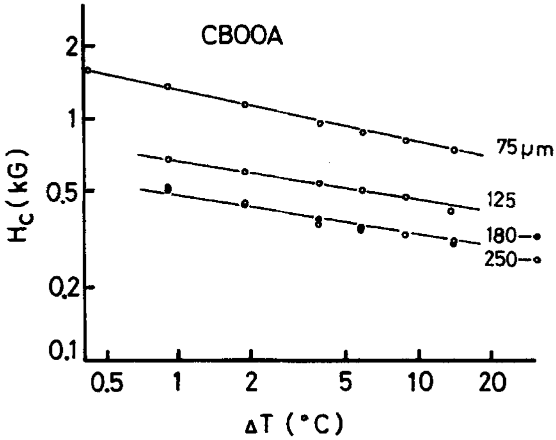

FIGURE 6 Critical magnetic field as a function of temperature above the smectic A–nematic transition temperature in CBOOA.

where $\Delta\chi$ is the magnetic anisotropy between the magnetic susceptibility in the directions parallel and perpendicular to the long axes of the molecules ($\chi_{\parallel} - \chi_{\perp}$). Theoretically, de Gennes[1] showed that in the case of a second order phase transition in liquid crystals, the expression for the bend elastic constant was given by the following equation:

$$(K_{33} - K_{33}^{\circ}) \propto (T - T_{n\cdot sA})^{-\gamma}, \qquad \gamma = 0.66, \tag{3}$$

where K_{33}° was the temperature independent ordinary nematic contribution. Using Equation (2) and the slope of the straight line in Figure 6, the critical exponent γ is estimated to be (0.5 ± 0.02) in this case. Cheung *et al.*[3] estimated for γ the value of 0.66 for the bend elastic constant in CBOOA from the temperature dependence of the viscosity determined by the measurement of the relaxation of the molecular alignment. Delaye *et al.*[4] also reported $\gamma = 0.66$ for the twist elastic constant from a light scattering measurement. However, we reported previously the value of 0.52 for γ from the measurement of the light scattering intensity in the Freedericksz transition. Cladis[6] also reported that the γ value estimated from the observation of the Freedericksz transition varies from 0.52 in the pure sample to 0.6 in the dirty sample. Our estimated value from this experiment is in good agreement with both our previous result and Cladis's value of the pure sample, being different from those of other groups. The value of $\gamma = 0.5$ coincides well with the recently predicted critical exponent within the mean field theory.[2]

All nematic liquid crystals in which interference rings are observed, for example CBOOA, EMBAC and CBOA have a smectic A phase at the lower temperature. In PAA and PAP without a smectic A phase, no such interference

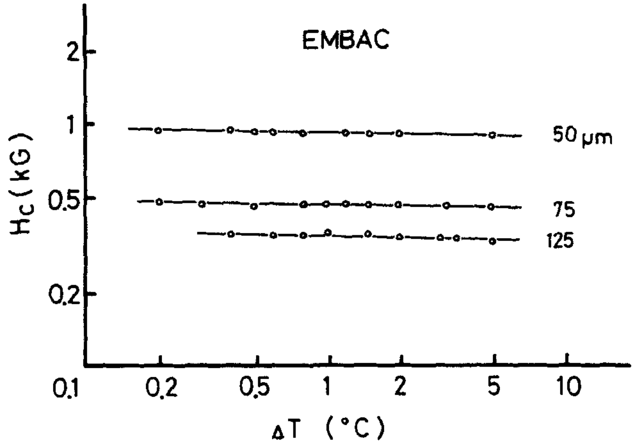

FIGURE 7 Critical magnetic field as a function of temperature in EMBAC.

ring was observed. Figure 7 shows the temperature dependence of the critical magnetic field H_c in EMBAC with various thicknesses. The critical exponent of H_c is very small compared with that of CBOOA. This fact suggests that the smectic A–nematic transition of EMBAC is a nearly first order transition, contrary to that of CBOOA.

As already mentioned, no interference ring was observed for the configuration ($\mathbf{S} \perp \mathbf{H}$) where the magnetic field $\mathbf{H}$ is perpendicular to the surface of the cell.

This fact suggests that the appearance of the interference ring is due to the magnetic deformation of directors in the central region which is aligned perpendicular to the magnetic field initially. The mechanism of the interference ring is tentatively speculated as follows. In the region $H_c \leq H \leq H_s$, the deformation of the bulk directors caused by the magnetic field seems not to be uniform contrary to the case of the usual analysis in the Freedericksz transition using the continuum theory. But the director orientations, deformated by the magnetic field, have a periodic bend deformation in a plane normal to the incident laser beam. Figure 8(a,b) show examples of probable deformations of directors. Under the above configurations, the refractive index of the extraordinary wave with polarization parallel to the applied magnetic field has a periodicity on the plane normal to the incident laser beam, resulting in the formation of two dimensional diffractive lattice for the extraordinary wave. However, it is clear that the refractive index for the ordinary wave whose polarization is perpendicular to the applied magnetic field, is uniform, resulting in the absence of the interference ring pattern as already mentioned. Let θ be the angle between the incident laser beam and the

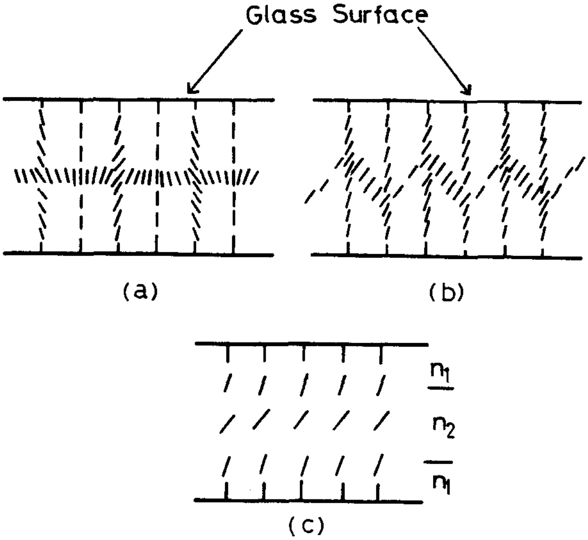

FIGURE 8 Three proposed aspects of the director configuration under the magnetic field.

first interference ring. From the consideration of one dimensional diffractive lattice, θ is expressed by the following equation:

$$\sin\theta = j\frac{\lambda}{h} \qquad (j = 1 \text{ for the first ring}), \tag{4}$$

where j, λ and h are integers, with λ representing the wave length of the incident laser beam and h represents the lattice constant of the diffractive lattice, respectively. From the above equation, one can find that θ increases with decreasing h of the periodic deformation.

Therefore the experimental fact that θ increases linearly with increasing magnetic field, shows that the spacial periodicity decreases with increasing magnetic field. If the distance h becomes the same order as the wave length λ of the incident laser light (6328 Å), the interference ring disappears, because of the limitation with ($\sin\theta \leq 1$). In this experiment, the maximum value of θ was typically 30.2° at 85°C in the sample thickness 75 μm, leading to the minimum value of $h \sim 2\lambda$. The disappearance of the ring at the upper critical field H_s is explained in terms of this mechanism. The periodicity length of the deformation of bulk directors is infinite at the threshold magnetic field H_c of the Freedericksz transition and turned to be finite above H_c being inversely proportional to the applied magnetic field. At this stage, the mechanism of originating such periodicity is not clear. However, the fact that such an effect was observed only in liquid crystals which show the smectic A phase at the lower temperature, may give some clue for the explanation. If the character of the smectic A phase is like the nematic phase at the high temperature, the configuration of Figure 8(b) seems most probable. In such a case, with increasing magnetic field the local smectic-like alignment may be reduced, returning in the decrease of the periodicity length. However, it seems there still remains another probable model to explain the observed interference rings. Even if the deformation of bulk directors caused by the magnetic field is uniform as shown in Figure 8(c), the interference rings may be originated from the multireflection of the incident laser beam in a medium with the variation of the refractive index n for the extraordinary wave. The angle of the scattered interference rings may depend on both the tilted angle and the tilted area of the directors, and increases with increasing magnetic field. In this case, when the cell is covered with the tilted region with increasing magnetic field, the interference rings should disappear, showing an upper threshold H_s. However, it is not easy to explain the observed large scattering angle by this second model.

Recently, Cladis and Torza[8] observed the new magnetic instability in the intermediate state between the smectic A and nematic states and explained the appearance of the striped texture by the periodic deformation of directors under a magnetic field. However, we observed the interference rings in the laser scattering pattern within a limited magnetic field range just above the threshold value of the Freedericksz transition as shown in Figure 9 (the

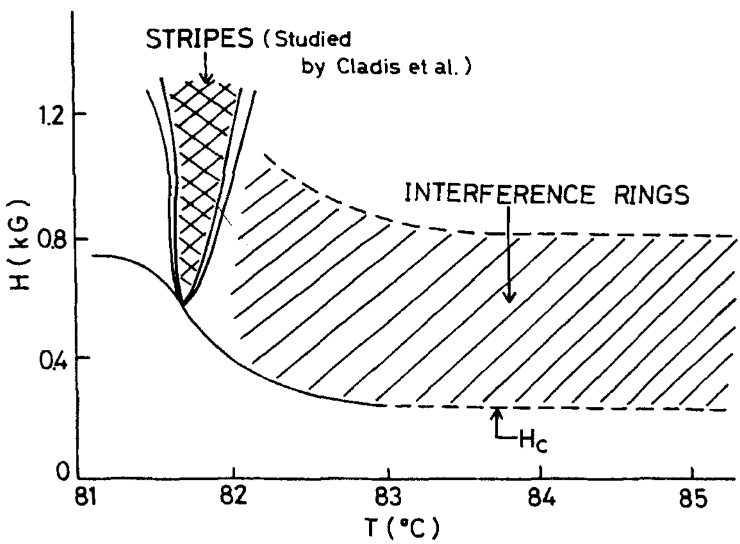

FIGURE 9 Limited magnetic field range drawn with the oblique line in which the interference rings can be clearly observed. Sample thickness is 180 μm. The striped texture studied by Cladis *et al.*, appears in the area drawn with the crossed line.

area drawn with the oblique line). In the intermediate state which was studied by Cladis *et al.*, the interference rings are unstable with the change of the magnetic field and are not clearly observed. The above mentioned magnetic effects seem to be different phenomena, because the former (studied by Cladis *et al.*) is the pretransitional phenomena at the smectic A–nematic phase transition and the latter is not as shown in Figure 9. However, both Cladis *et al.* and we proposed the same type of periodic deformation of the directors for the explanation as shown in Figure 8(b).

Until now, we can not explain completely the appearance of the concentric circular interference ring with only one dimensional periodic deformation of the directors. The two dimensional structure should be necessary in our case. We are speculating that the domains in which one dimensional periodic deformation (Figure 8(b)), are distributed randomly in the cell.

References

1. P. G. de Gennes, *Solid State Commun.*, **10**, 753 (1972).
2. W. L. McMillan, *Phys. Rev.*, **A9**, 1720 (1972).
3. L. Cheung, R. B. Meyer, and H. Gruler, *Phys. Rev. Letters*, **31**, 349 (1973).
4. M. Delaye, R. Ribotta, and G. Durand, *Phys. Rev. Letters*, **31**, 443 (1973).
5. A. Sakamoto, K. Yoshino, U. Kubo, and Y. Inuishi, *Japan. J. appl. Phys.*, **15**, 745 (1976).
6. P. E. Cladis, *Phys. Rev. Letters*, **31**, 1200 (1973).
7. P. Sheng, *RCA Rev.*, **35**, 408 (1974).
8. P. E. Cladis and S. Torza, *J. Appl. Phys.*, **46**, 584 (1975).