

Suppression of fluctuations of anisotropy in cholesteric liquid crystals by an electric field

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We observed a "red" spectral shift in the form of the Bragg band due to the increase in the order parameter S in a stabilizing field.

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In real single-domain samples of cholesteric liquid crystals (CLC) characterized by a high degree of orientation there occur thermal fluctuations of anisotropy (fluctuations of director orientation) which are defined by the order parameter S .⁽¹⁾ As is known, a stabilizing field applied to a layer of the nematic liquid crystal tends to reduce the amplitude of fluctuations and leads to an increase in S .⁽²⁾ The purpose of our work is to study the effect of fluctuations of anisotropy on the form of the selective light reflection band in CLCs. This was determined earlier without taking these fluctuations into account.^(3,4) The role of fluctuations is best established by reducing them with a stabilizing electric field applied along the axis of a spiral in a CLC with negative dielectric anisotropy.

Changes in the selective reflection band in an electric field were recorded by modulation spectroscopy methods using a scheme shown in the inset in the Fig. 1. An electric field with frequency f and intensity below the threshold of electro-hydrodynamic instabilities was applied to a sample. Changes in the sample transmittivity ΔT were detected by a photodiode and, after narrow-band amplification at a frequency $2f$, were recorded by a synchronous detector.

Samples consisted of unit-sandwiches with etched internal surfaces. The quality of liquid crystal orientation was controlled microscopically by the absence of dislocations, and the measurements were conducted within a single de Gen zone.⁽¹⁾ The liquid crystal represented a nematic-cholesteric mixture consisting of *p*-*n*-methoxybenzylidene-*p*-butylaniline (MBBA, 65 wt%), 2,3-dicyano-4-amyloxyphenylic *p*-amyloxybenzoic acid ester (DCAPABAE, 10 wt%), and cholesterylolate (25 wt%). The dielectric anisotropy of the nematic component $\epsilon_a = -4$.

The selective reflection band manifests itself in the optical transmission spectrum of a layer shown as curve 1 in Fig. 1. Curve 2 indicates a relative change in the transmission spectrum $\Delta T/T$ which exhibits a change in the selective reflection band in a stabilizing field. This spectrum is obtained by subtracting from the measured values of $\Delta T/T$ (curve 3) the level of change in the transmission (curve 4), not associated with the selective reflection band and dependent on the reduction of the total scattering background.⁽¹⁾ Thus, the selective reflection band undergoes a "red" shift in a stabilizing field; moreover, the new position of the band is shown schematically as curve 5.

We think that the red spectral shift of the selective reflection band in test samples with a high degree of orientation is due to the suppression of thermal fluctuations of anisotropy by the field. Taking into account the geometry of experiment, we see that only the conical (umbrella-shaped) mode^(1,5) is suppressed while the field has no effect on the torque fluctuations.⁽¹⁾ The presence of conical-mode fluctuations may be considered statistically and in a kinematic approximation can be interpreted as the existence in the sample volume of sections with the spiral axis deflected from the normal to a layer by a certain angle θ . For these sections the wavelength of selective (Bragg) reflection is as follows:

$$\lambda_{\max} = P \bar{n} \cos \theta \quad (1)$$

(P is the pitch of the spiral, \bar{n} is average refractive index of the CLC), i.e., smaller than for a section with the spiral axis that coincides with the normal to a layer. Application of a field reduces the amplitude of fluctuations and, consequently, θ , while it leads to extension of the wavelength that corresponds to the maximum of the selective reflection band, i.e., to the red shift of the band. The same effect is achieved by an increase of the mean refractive index \bar{n} in the field due to suppression of conical fluctuations.

We shall present estimates that give support to our model. Suppression of anisotropic fluctuations is equivalent to an increase in the field of the order parameter S that in a simple case is defined as follows:

$$S = \frac{3}{2} \overline{\cos^2 \theta} - \frac{1}{2}, \quad (2)$$

where θ is the angle of local deflection of molecules from the preferred direction (of director). An increase in the order parameter ΔS in a stabilizing field for nematic liquid crystal—calculated by de Gen⁽¹⁾ and observed by Poggi and Filippini⁽²⁾—may be described as follows:

$$\Delta S = \frac{k_B T \epsilon_a^{1/2}}{4 \pi^{3/2} K^{3/2}} E, \quad (3)$$

where k_B is the Boltzmann constant, T is absolute temperature, and K is the modulus of elasticity (single-constant approximation). If we assume that the order of ΔS for CLC remains the same, under the conditions of our experiment $\Delta \bar{S} = 2 \times 10^{-4}$. The value of the spectral shift of the selective reflection band may be expressed with the aid of Refs. 1 and 2, as follows: $\Delta \lambda_E \approx \Delta S \lambda_{\max} \approx 0.5 \text{ nm}$ (for $\lambda_{\max} = 1000 \text{ nm}$). Taking into account $dT/d\lambda = 5 \times 10^{-3} \text{ nm}^{-1}$ —a value found for a wing of the selective reflec-

tion band—the expected relative change in the transmission, dependent on the band shift, is $\Delta T/T = 10^{-3}$. Analysis of a dependence of the observed value $\Delta T/T$ on the frequency of the modulating field showed that a disagreement between the derived value and the experimental value of $\Delta T/T = 1.5 \times 10^{-4}$ at 1 kHz (see Fig. 1) is due to the relaxation of fluctuations^[5] being large in comparison to the period of the modulating field.

The following fact serves as an additional confirmation that the observed red shift of the selective reflection band in a field is indeed a result of suppression of the thermal fluctuations of anisotropy. Enhancement of the capability of single-domain samples of the original nematic liquid crystals to transmit light in a field, which can be explained only within the framework of a mechanism of fluctuation suppression,^[1] was of the same order, all things being equal, as that in CLCs.

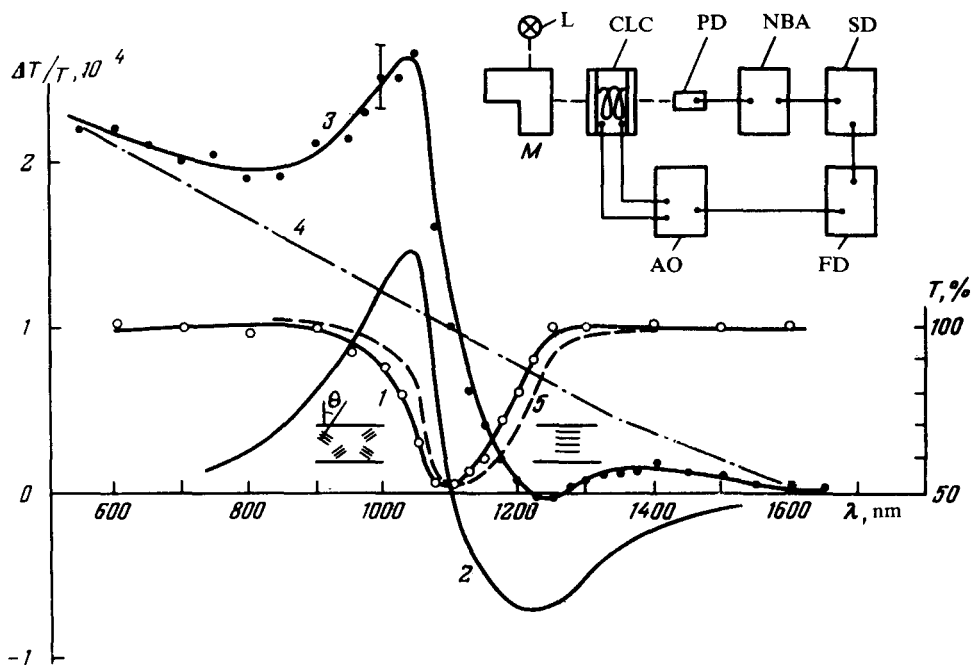


FIG. 1. Transmission spectrum T and relative change of transmission $\Delta T/T$ in a field with intensity $E = 3 \times 10^4$ V/cm and frequency $f = 1$ kHz, for a planar layer of CLC with thickness $d = 24 \pm 0.5 \mu\text{m}$ (explanation in the text). In the inset: bloc-diagram of experimental setup: L —lamp, M —monochromator, PD —Photodiode, NBA —narrowband amplifier, AO —sinusoidal voltage audio-frequency oscillator, FD —frequency doubler, SD —synchronous detector.

The observed effect of the red shift cannot be explained by assuming that the quality of planar texture improves in a field (i.e., additional “single-domainization” of sample). Actually, specific experiments showed that such an enhancement of texture quality leads to a different effect, namely the spectral narrowing of the Bragg band,^[7] an observation which is in agreement with calculations.^[8]

In conclusion, we observed an increase in the wavelength of selective reflection (a

red shift of the Bragg band) when a stabilizing electric field was applied to a CLC layer, thus leading to an increase in the CLC order parameter due to suppression of the thermal fluctuations of anisotropy.

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¹¹Lowering the temperature caused a "red spectral shift of the selective reflection band of test samples. This shift is due to an increase in the order parameter S associated with an amplitude decrease of both modes, and was analyzed in Ref. 6.

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