Incommensurability effect in nematic liquid crystal with induced gyrotropy

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Two incommensurable periods in the distribution of the director in a nematic liquid crystal with an induced gyrotropy are predicted theoretically. They have also been observed experimentally.

Intrinsic and induced cholesteric liquid crystals differ in the mechanism responsible for the spiral supermolecular structure. The nature of the close packing of the individually chiral molecules in the mesophase during orientational ordering is determined by the asymmetry and anharmonicity of the interaction forces.

In a nematic liquid crystal with an induced gyrotropy, the orientational order set by dispersion forces is locally disrupted when chiral additives are introduced. The large dimensions of the added molecules or their existence in the matrix only in the form of complexes leads to a distortion of the orientational order over a substantial distance. A superposition of the local deformations, which has been studied previously in the single-constant approximation[1] with allowance for the difference in elastic constants, leads (as we show below) to a distortion of the ideal spiral structure—more precisely, to the appearance of two incommensurate periods. This state corresponds to a minimum free energy of the liquid crystal.

Our theory is based on the free energy of a nematic liquid crystal with an induced spiral structure[1]:

$$F_d = \frac{1}{2}\,\{K_{11}(\mathrm{div}\,\mathbf{n})^2 + K_{22}(q_0 + \mathbf{n}\cdot\mathrm{curl}\,\mathbf{n})^2 + K_{33}[\mathbf{n}\times\mathrm{curl}\,\mathbf{n}]^2\}, \qquad (1)$$

where K_{11}, K_{22}, and K_{33} are the Frank elastic constants, q_0 is the wave vector of the spiral structure, which depends on the concentration of the chiral additive and which was found in Refs. 1 and 2 for the case of equal elastic constants.

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We seek the distribution of the director in the form

$$\mathbf{n} = (\cos\theta(z)\cos\varphi(z), \cos\theta(z)\sin\varphi(z), \sin\theta(z)). \quad (2)$$

The axis of the spiral structure runs along the z axis. We assume $\varphi = qz$, where q is the wave vector of the actual deformed spiral, which we assume to be an unknown parameter of the problem. Introducing the notation $\alpha = 1 - K_{11}/K_{33}$ and $\beta = 1 - K_{22}/K_{33}$, we can write the free energy as follows:

$$F_d = \frac{K_{33}}{2}\{(1-\alpha\cos^2\theta)(\frac{d\theta}{dz})^2 - \beta(q_0 - q\cos^2\theta)^2 + q_0^2 + (q^2 - 2qq_0)\cos^2\theta\}. \quad (3)$$

This is essentially the same as the free energy which is used in a study of incommensurability effects.[3,4]

With $\alpha = \beta = 0$, an extremum of functional (3) is reached for a nonlinear solution of the sine-Gordon equation[3]:

$$\frac{\theta}{2\pi} = C + \frac{1}{2\pi} am[\pm 4K(\kappa)z/l], \quad (4)$$

where $amf(z)$ is the Jacobi elliptic amplitude, and $K(\kappa)$ is the complete elliptic integral of the first kind. The unknown parameter κ, which is related to the first integral (C) of the Euler equation corresponding to (3), is found (as is q) from the minimum of free energy (3) (in this case, for $\alpha \neq 0$ and $\beta \neq 0$), in which solution (4) is used as a trial function. Determining the unknown parameters κ and q from the minimum of the free energy, we completely solve our problem of determining the actual structure of a nematic liquid crystal with an induced gyrotropy. Solution (4) corresponds to rotations θ through angles with are multiples of 2π with a period

$$l = 4\kappa K(\kappa)/(q^2 - 2qq_0)^{1/2}. \quad (5)$$

Under the condition $K_{33} > 2K_{22}(1 - K_{33}/2(K_{33} + K_{11} - K_{22}))$, this period takes on the value $l = 4[2\times(2-\alpha)(1-\beta)/(3-4\beta)]^{1/2}q_0^{-1}$, and the wave vector of the spiral structure is $q = q_0(1-2\beta)/(1-\beta)$. A solution of this type corresponds to a periodic array of static solitons in the orientation of the director, while the spiral structure is retained.

Experimentally, this result is manifested in two-band lasing with distributed feedback on the basis of a nematic liquid crystal with an induced gyrotropy.

A laser of this type is an oriented layer (15–30 μm thick) of a liquid crystal of planar texture, activated by an emitting dye of the phenolenone class with a concentration of 0.5% by weight. As matrices we used single components (MBBA and 5TsB) and the multicomponent substance ZhK-654. A gyrotropy was produced in the single-component nematic liquid crystals by means of a nonmesogenic, optically active additive; in the case of the ZhK-654, we added cholesteryloleate. Oriented layers of the liquid crystals with impurities were pumped by the second harmonic of an Nd^{3+} laser ($\lambda = 0.53\ \mu$m) operating in single pulses ($\tau_p = 30$ ns). The maximum pump intensity was 8 MW/cm^2; the beam diameter in the plane of the liquid-crystal layer was 0.5 mm.

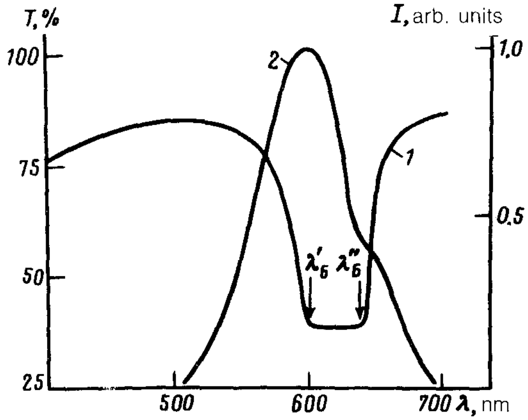

FIG. 1. 1—Transmission spectrum of a mixture of ZhK-654 with 36% cholesteryloleate for linearly polarized light; 2—fluorescence spectrum of the impurity dye. The layer thickness is 30 μm.

A distinctive feature of the output spectra of a distributed-feedback laser using an induced spiral structure is that the Bragg wavelengths (λ_B) differ from those determined from the selective reflection spectra. In a distributed-feedback laser based on cholesteric mesogens, the wavelengths λ_B are the same, within the experimental errors, in the emission and in the selective reflection spectra.

Figure 1 shows the transmission spectrum of a mixture of ZhK-654 and 36% of cholesteryloleate (curve 1). For this position of the spectrum of selective absorption, in the fluorescence band of the dye impurity (curve 2), the lasing occurs in two bands simultaneously, and the values of λ_B (the central and more intense lines of the emission band) are 599.2 and 640 nm. Correspondingly, the values of λ_B found from the selective absorption spectrum as the average at the half-intensity level[6] is 620 nm.

In the induced spiral structure due to a nonmesogenic, optically active additive, the content of the latter required to achieve the same spiral pitch is far lower than that which causes a large width of the selective absorption spectrum. The result is a degradation of the conditions for simultaneously emission in two bands. Figure 2 shows selective absorption spectra as the concentration of the optically active additive was varied. At an additive concentration of 6.4%, the value of λ_B in the emission spectrum of the distributed-feedback laser based on MBBA is 577.2 nm. This wavelength corresponds to the short-wavelength edge of the plateau of the selective absorption band. An increase in the additive concentration to 7.2% leads to emission with $\lambda_B = 594.7$ nm, which corresponds to the long-wavelength edge of the selective absorption band for this concentration. In the nematic liquid crystal with the same optically active additive we find a similar situation.

In summary, the lasing in the distributed-feedback laser occurs in two bands, which correspond to the edges of the plateau in the selective absorption spectrum. According to the operating principle of a distributed-feedback laser, lasing is possible only on a periodic structure whose Bragg frequency falls in the region of an amplification of the lasing impurity. Any distortions of an aperiodic nature (a gradient of the pitch, a scatter in the axes of the spiral over the thickness, or a heating of the substance by the pump light) will lead to only a broadening of the output line. The appearance of

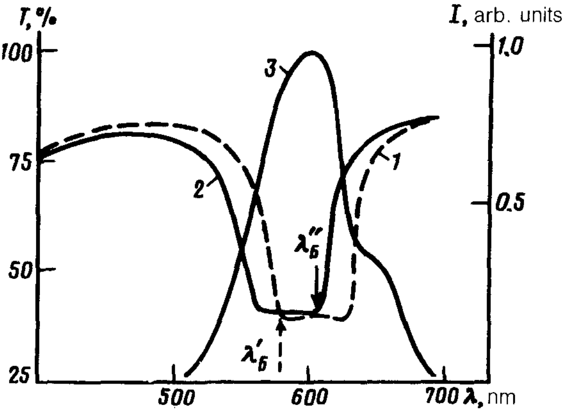

FIG. 2. Transmission spectrum of MBBA for various concentrations of the optically active additive. 1—$c = 6.4\%$; 2 = $c = 7.2\%$, linearly polarized light; 3—fluorescence spectrum of the impurity dye. The layer thickness is 15 μm.

two lines at the edges of the selective absorption band is evidence of a new modulation of the spiral structure.

Taking into account the constancy of the width of the plateau in the selective absorption spectrum of the mixture of MBBA with the optically active additive in the concentration range used here (Fig. 2), we find $\Delta\lambda_B = 52$ nm with $\lambda'_B = 577.2$ nm; this result corresponds to $\lambda'_B/\lambda''_B \approx 0.917$.

Estimates based on the elastic constants[7] of MBBA ($K_{11} = 6\times10^{-7}$ dyn, $K_{22} = 4\times10^{-7}$ dyn, and $K_{33} = 7.5\times10^{-7}$ dyn) yield $lq \approx 0.925$, in good agreement with experiment.

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