Dynamic effects in the blue phases of liquid crystals

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Dynamic effects associated with changes in the intensity of diffraction bands and a rotation of the polarization plane of light have been observed in the blue phases in an alternating electric field. These dynamic effects lead to a modulation of the intensity and of the polarization plane of the light.

The local dielectric anisotropy of the cubic blue phases[1] gives rise to several unique physical properties. Nontrivial electrooptic effects are observed: a giant electrostriction,[2,3] a change in the orientation of single crystals,[4] and phase transitions in an electric field.[3-5] The relaxation times of these effects are[6] $\tau_1 \gtrsim 1$ s. In addition to a macroscopic restructuring, however, an external agent can also cause a local change in the order, which should be characterized by considerably shorter relaxation times. Our purpose in the present study was to learn more about the dynamic effects in the blue phases.

Measurements were carried out in the blue phases[1] BPI and BPII in a mixture of the chiral nematic crystal 4-cyan-4(2-methyl)butyl-phenyl (52% by weight) and the nematic liquid crystal 4-cyan-heptyloxydiphenyl, with a positive dielectric anisotropy. Single crystals of the blue phases were grown in flat cells with an electrically conducting coating. In the resulting single crystals, the [110] direction in BPI and the [100] direction in BPII ran perpendicular to the plane of the cell. The temperature interval of BPI is 27.30–27.67 °C, and that of BPII is 27.67–27.82 °C.

It was found that the application of an alternating electric field to the blue phase causes a modulation of the intensity of light transmitted through the sample. The basic part of the alternating component of the light has a frequency twice that (ν) of the electric field. Figure 1 shows, for the two circular polarizations, the spectrum of the ratio of the amplitude of the alternating component of the signal ($I_{2\nu}$) to the intensity of the constant component (I_0). The plus sign corresponds to an increase in the signal with increasing field. At $\lambda < 630$ nm, the intensities $I_{2\nu}(\lambda)$ of the two circular polarizations are similar. In this wavelength region, the temporal modulation of the intensity should be attributed to a decrease in the refractive index in the electric field[7] and a change in the conditions for interference of the light reflected from the surface of the sample. The wavelength shift of the interference spectrum that results[7] leads to a temporal modulation of the light intensity. The modulation amplitude (Fig. 1) corresponds to $\Delta n/n \approx 3 \times 10^{-3}$ in a field $E = 2.4 \times 10^4$ V/cm.

In the region of the diffraction band, a substantially higher peak of an alternating signal was found for the natural circular polarization (Fig. 1). The sign of the effect implies a decrease in the intensity of the diffraction peak at the maximum of the voltage applied to the sample. In an electric field there is a shift of the diffraction band, which characterizes the time scale of the relaxation to an equilibrium value of the cell
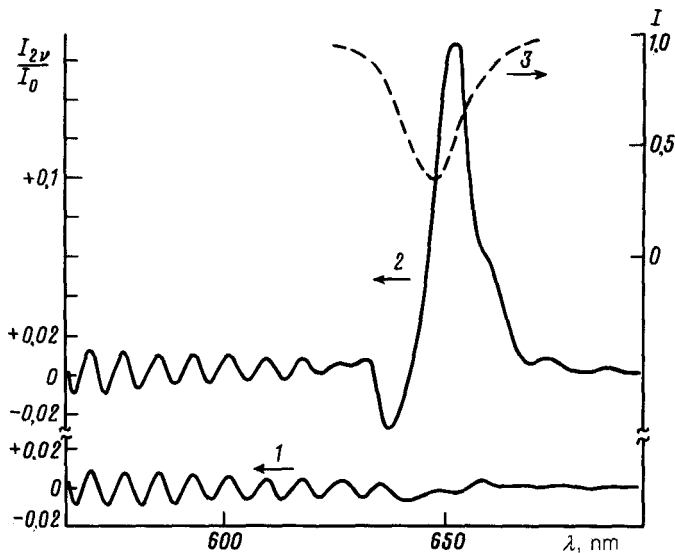
FIG. 1. *1, 2*—Spectra of the amplitude modulation $I_{2\nu}/I_0$ of the right- and left-hand circularly polarized light; *3*—diffraction band in the transmission spectrum I. The thickness of the sample is $d = 12\,\mu$m; $U = 20$ V; $\nu = 200$ Hz; $T = 27.5$ °C.

parameter,[6] $\tau_1 \gtrsim 1$ s. At the time scale $t \sim 1/\nu$, however, the diffraction intensity does not remain constant. The maximum of the effect lies on the long-wave side of the center of the diffraction line. The magnitude of the effect is quadratic in the field. The relaxation time is $\sim 10^{-4}$ s.

For unpolarized or circularly polarized light, the intensity of the alternating component (the amplitude modulation) goes through minima outside the diffraction band (Fig. 1). It was found that linearly polarized light emerging from an analyzer is modulated, even at the wavelengths corresponding to the minima of the amplitude modulation. Figure 2 shows the magnitude of the alternating component of the intensity as a function of the angular position of the analyzer at the minima of the amplitude modulation [$I_{2\nu}^{\min}(\varphi)$]. This modulation is not a consequence of an electric-field-induced birefringence,[4,8] since the phase of the alternating component varies in the opposite direction when the analyzer is rotated through the position corresponding to complete blocking ($\varphi = 0$; Fig. 2). The behavior of the magnitude of the alternating component as a function of λ (Fig. 3) is quite different from the behavior of the amplitude modulation (Fig. 1). The effect (Figs. 2 and 3) stems from a cyclic rotation of the polarization plane of the light. The magnitude of the effect is proportional to $\sin 2\varphi$.

Away from a minimum of the amplitude modulation, we observe a superposition of two effects, which stem from the change in the refractive index and the rotation of the polarization plane. In this case the dependence is asymmetric with respect to $\varphi = 0$ (Fig. 2), since the phase of the alternating signal for the modulation of the polariza-
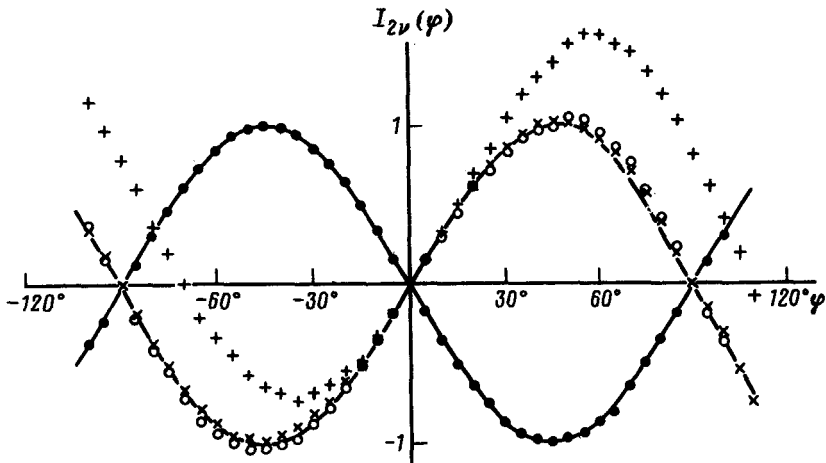
FIG. 2. Intensity of the alternating component of the signal versus φ at the minima of the amplitude modulation [x—$I_{2v}^{\min}(\varphi)$, BPI, $\lambda = 604$ nm; O—$I_{2v}^{\min}(\varphi)$, BPII, $\lambda = 635$ nm] and at maximum [$+$ — $I_{2v}^{\max}(\varphi)$, BPI, 601 nm] (arbitrary units). O—($I_{2v}^{\max}(\varphi) - b\sin^2(\varphi)$), $d = 12\,\mu$m, $v = 200$ Hz. Solid lines) $I = \pm \sin 2\varphi$.

tion plane reverses when $\varphi = 0$ is crossed ($I_{2v}(\varphi) \sim \sin 2\varphi$), while that for the amplitude modulation does not change ($I_{2v}(\varphi) \sim \sin^2\varphi$). If the modulation due to the change in the refractive index is subtracted from the resultant effect, the φ dependence becomes similar to the dependence becomes similar to the dependence $I_{2v}^{\min}(\varphi)$ (Fig. 2).
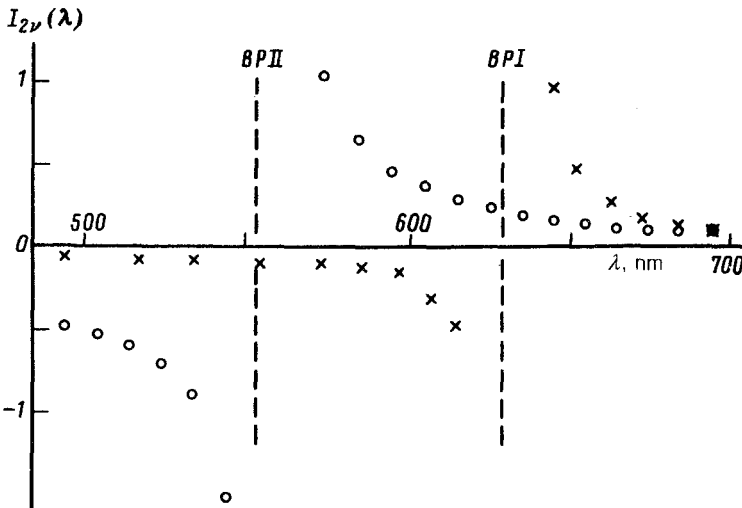
FIG. 3. Spectrum of the alternating component of the signal during modulation of the polarization plane in BPI ($\times$, $T = 27.6$ °C) and BPII (O, $T = 27.7$ °C) (arbitrary units). Dashed lines— Positions of the diffraction lines of the transmission spectrum. $d = 12\,\mu$m, $v = 200$ Hz.

The change in the cell parameters a of the single-domain sample in a field may occur through a coordinated restricting of the entire sample or the formation and annihilation of defects. Macroscopic restructurings of this sort are characterized by relaxation times $\tau_1 \gtrsim 1$ s. The difference between the spectral positions of the maxima of the alternating component of the signal and of the diffraction band, $\lambda_{\rm BP}$ (Fig. 1), indicates that there is a cyclic shift of the band in the alternating field, in addition to the change in the diffraction efficiency. This shift has an anomalous sign ($\lambda_{\rm BP}$ decreases at the maximum of the sinusoidal electric field for substances with a positive dielectric anisotropy for the [110] and [100] reflections of BPI and BPII, respectively) and has a short relaxation time ($\sim 10^{-4}$ s). The periodic change in $\lambda_{\rm BP}$ in the alternating field might be a consequence of a decrease in the refractive index in the electric field. A change in n leads to a shift of $\lambda_{\rm BP}$ at a fixed size of the unit cell ($\lambda_{\rm BP} = 2dn$). The optical properties (the diffraction intensity, the rotation of the polarization plane, and the refractive index) are determined by the squared modulus of the Fourier harmonic ϵ_τ of the order parameter of the blue phases.[9] The change in ϵ_τ at constant values of the reciprocal-lattice vectors τ occurs at the level of the unit cell and does not require a macroscopic restructuring of the overall sample. The relaxation time of ϵ_τ within a unit cell can be estimated from the expression[8] $\tau_2 \sim \eta/(Kq^2)$, where η is the viscosity, K is the Frank modulus, and $q = 2\pi/a$. With $\eta \sim 1$ P, $K \sim 5 \times 10^{-7}$ dyn, and $q \sim 2 \times 10^5$ cm^{-1}, we find $\tau_2 \sim 0.5 \times 10^{-4}$ s, which is close to the measured relaxation time.

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