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Interference Oscillations in the Dynamics of the Optical Response of Polymer Dispersed Nematic Liquid Crystals

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Abstract—The dynamics of the optical response of a polymer-dispersed nematic liquid crystal under the action of electric field pulses was experimentally studied for film samples differing in the size of nematic droplets. The optical signal relaxation curve exhibits an oscillating character, with the number of oscillations determined by the transverse size of the nematic droplets. The interference character of the signal oscillations is confirmed by an analysis of the oscillating response within the framework of the anomalous diffraction approximation and by a comparison with the dependence of the transmitted light intensity on the applied voltage measured in a static regime. *© 2002 MAIK "Nauka/Interperiodica".*

Previously [1–6], we discovered and studied oscillations in the dependence of the optical transmission of polymer-dispersed nematic liquid crystal (PDNLC) films on the applied voltage. These oscillations are caused by interference of the light transmitted between the liquid crystal (LC) droplets and the light scattered from the droplets in the forward direction. Reorientation of the LC director inside the droplets under the action of an electric field applied to the PDNLC film leads to a change in the phase shift between the interfering light rays and, hence, to oscillations in the dependence of the transmitted radiation on the applied voltage. The number of oscillations (extrema) on the transmitted intensity curve is proportional to the transverse (i.e., normal to the film plane) size of the droplets. The experiments reported in [1–6] were performed in a static regime, whereby the voltage sweep time (10 s and above) was significantly greater than the characteristic time of reorientation of the nematic droplets (1–10 ms). Under these conditions, the oriented structure of the nematic droplets passes, in the course of increasing field strength, through a sequence of equilibrium states from a bipolar configuration of the LC director to the state of saturation, in which the LC director is oriented parallel to the field nearly the whole droplet volume.

Technical applications, such as spatial light modulators used in flat displays and the data recording, storaging, and processing devices, typically employ discrete addressing based on the high-frequency pulsed electric field action upon discrete elements of a shutter. In this context, it is important to study the contribution of the interference component to the optical response of PDNLC films measured in a dynamic regime, in which case the time of electric signal switching is much smaller as compared to the characteristic time of reorientation of the nematic droplets.

The samples of PDNLC films were prepared by a conventional method based on the phase separation of a homogeneous solution of the liquid crystal and prepolymer components, initiated by photocuring of the polymer matrix [7]. The composition components were an optical adhesive of the NOA-65 type (Norland Products Inc.) and a nematic mixture of cyanobiphenyl derivatives [8] taken in a 1 : 1 mass ratio. The initial solution was placed into a gap between glass plates with transparent electrodes, the film thickness being determined by 10-μm spacers. During the subsequent treatment, the total power of a mercury lamp was varied from 1 to 10 mW/cm^2. The sample was separated into several regions, each processed using a certain photocuring regime (temperature and radiation intensity). By varying these technological parameters, it was possible to change morphological characteristics of the final sample structure. As a result, the average nematic droplet size in various regions of a sample film varied from 1 to 10 μm. Within a certain region of the composite film, the deviation of the nematic droplet size from the average did not exceed 40%.

Investigations of the sample texture by means of a polarization microscope showed that the internal oriented structure of the nematic droplets corresponds to a bipolar configuration of the LC director. The symmetry axes, connecting two poles of the droplet, are oriented predominantly in the film plane and exhibit random azimuthal orientations. It should be noted that the symmetry axes of small droplets may slightly deviate from the film plane, because the shape of such droplets is close to spherical. Large droplets are significantly oblate, with the transverse size being 1–2 μm smaller than the lateral dimensions (in the film plane). In droplets of this shape, a minimum of the elastic energy is attained

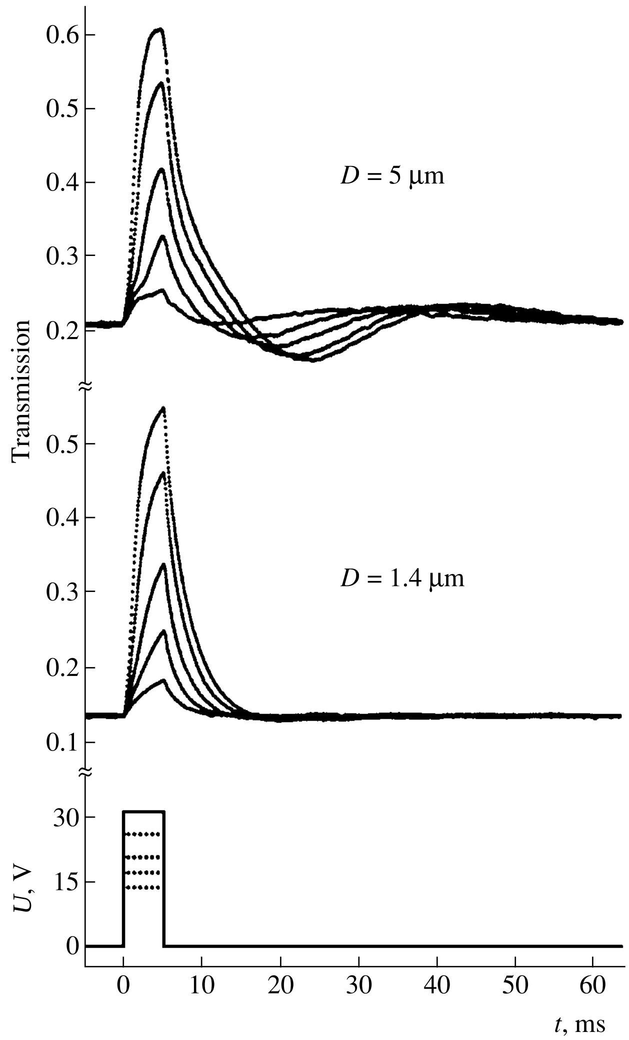

Fig. 1. Oscillograms of the optical transmission signal from two regions of a composite polymer film containing nematic droplets with different average size D, observed in response to a single control voltage pulse (bottom diagram) with an amplitude of 14.0, 17.5, 21.0, 26.3, and 31.5 V (bottom to top).

for the symmetry axes parallel to the long axes of droplets [9] that aligned in the film plane.

The electrooptical characteristics were studied using the monochromatic radiation of a He–Ne laser operating at $\lambda = 0.633$ μm. The volt–contrast characteristics were measured using a slowly varying (~1 V/s) amplitude of the alternating (500 Hz) voltage applied to the cell electrodes. We studied the dynamics of the optical response of the PDNLC films as a function of the amplitude of single rectangular electric pulses with a duration of 5 ms.

Figure 1 presents oscillograms of the optical response for two PDNLC film regions differing in nematic droplet size and shows the shape of the control electric pulse. The sample containing small nematic droplets exhibits monotonically decaying, nearly exponential curves of the transmitted light intensity variation after switching off the applied voltage. The curves of signal relaxation observed for the sample with large nematic droplets exhibit a significantly different nonmonotonic behavior, whereby the transmitted light intensity decreases, exhibits a minimum, increases again, passes through a local maximum, and then gradually decreases to the initial level. The position of minimum on the time scale, as well as the total relaxation time, depend on the electric pulse amplitude. This is explained by the fact that a more pronounced transformation of the oriented droplet structure requires a longer relaxation time. At the point of minimum, the transmitted light intensity is lower than that in the initial state (before the application of electric pulse). It is interesting to note that the depth of the minimum (i.e., the difference of optical transmission in the initial state and at the point of minimum) increases with the control pulse amplitude.

For comparison, Fig. 2 shows variation of the transmitted light intensity in response to the applied voltage for the same sample regions measured in a static regime. The curve for the PDNLC containing small nematic droplets exhibits a classical S-like shape [7]. No threshold behavior with a bending point corresponding to the Fréedericksz critical field is observed in this case, which is explained, as noted above, by the presence of nematic droplets with the symmetry axes oriented at an angle with respect to the film plane. Violation of the condition of orthogonality of the axes of a bipolar LC director configuration and the applied field direction [2–4] is just what leads to a thresholdless shape of the volt–contrast characteristic. This situation is typical of composite films in which the nematic droplet size is significantly smaller than the film thickness. In contrast, the orthogonality condition holds better for the film containing large droplets, as manifested by a threshold character of the optical transmission variation in this sample region (Fig. 2). This curve displays additional minimum and maximum resembling those observed in the dynamic regime (Fig. 1).

Now let us use an analytical approach developed previously [2–5] for explaining the observed interference oscillations of the volt–contrast characteristic. The relative phase shift of the interfering light rays is determined by the ratio of the difference in the optical paths to the light wavelength: $\Delta nD/\lambda$, where $\Delta n = n_{lc} - n_p$, n_{lc} is the refractive index for an extraordinary ray in the liquid crystal and $n_p \simeq 1.52$ is the refractive index of the polymer matrix. The refractive index for the extraordinary ray, depending on the mutual orientation of the electric vector of the light wave and the LC director, varies in the given nematic composition from $n_{lc}^{min} \simeq 1.52$ to $n_{lc}^{max} \simeq 1.72$. In the saturation state (corresponding to the maximum transmission in Figs. 1 and 2), the LC director is perpendicular to the electric field vector of the light beam, $\Delta n = n_{lc}^{min} - n_p = 0$, and, hence, the

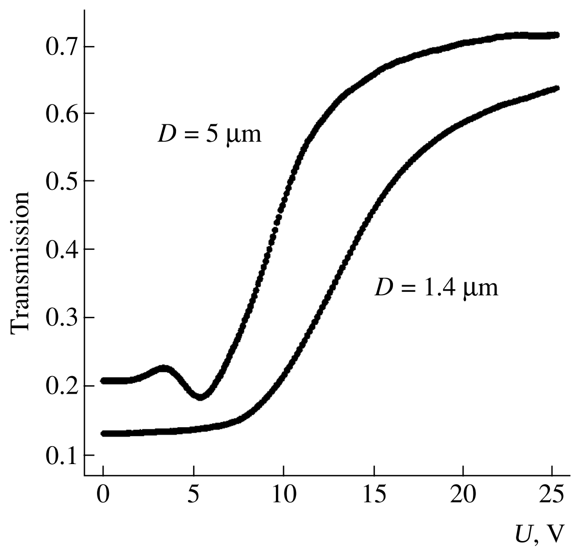

Fig. 2. Variation of the optical transmission depending on the applied voltage, measured in the static regime for the same regions of the composite film as in Fig. 1.

phase shift is zero. As the applied voltage decreases (Fig. 2), the birefringence increases and, at a certain point, the optical path difference ΔnD reaches $\lambda/2$. This condition corresponds to the first minimum in the volt–contrast characteristic. As the applied voltage droplets further, the path difference crosses a level of $\Delta nD = \lambda$, which corresponds to the first local maximum. The relation $\Delta nD = 3\lambda/2$ corresponds to the second minimum in the transmission, and so on.

The transverse size of small nematic droplets amounts on the average to 1.4 μm. In this case, the maximum possible optical path difference is $\Delta n^{max}D = 0.2 \times 1.4$ μm = 0.28 μm < $\lambda/2$. For such small LC droplets dispersed in the polymer matrix, no oscillations can be observed in the optical transmission curves (Fig. 1 and 2).

For large nematic droplets with a transverse size of 5 μm, the optical path difference is $\Delta n^{max}D = 0.2 \times 5$ μm = 1.0 μm ≈ $3\lambda/2$. Therefore, relaxation of such droplets into the initial state after switching off the electric pulse (Fig. 1) or with decreasing applied voltage (Fig. 2) will result in the transmission sequentially passing through the first minimum, maximum, and second minimum, and attaining saturated initial (zero field) state, in agreement with the experimental observations.

Thus, we reported for the first time the results of investigation of the oscillating character of the optical response relaxation observed upon application of a rectangular electric pulse to the film of a polymer-dispersed nematic liquid crystal. An analysis of the experimental results presents convincing evidence of the interference nature of these oscillations. It should be noted that the dynamic pattern of the optical response of composite films in the general case can be extremely complicated as a result of the interplay of various effects including, for example, the influence of an electric field of the spatially separated impurity ions [10, 11], the formation of defects and domains [12, 13], restructurization of the droplet–polymer interphase boundary [8, 14], etc. In this context, the results presented above show the importance of taking into account the interference effects during complex analysis of the dynamic characteristics of composite films.

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