

Photostimulated transformation of molecules—a new type of “giant” optical nonlinearity in liquid crystals

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A novel type of “giant” optical nonlinearity in liquid crystals is observed and investigated. It is manifest near the intrinsic absorption edge and is characterized by anomalously large cubic nonlinearity constants $\epsilon_3 \approx (10^{-2}-10) \text{ cm}^3/\text{erg}$ for relaxation times $\tau \approx (10^{-1}-10) \text{ sec}$. The complete set of independent components of the nonlinear susceptibility tensor χ_{ijkl} is determined for the MBBA liquid crystal. It is shown that the nonlinearity is due to photostimulated transformations of the molecules, in which the final form possesses a polarizability that differs from that of the initial form. Some characteristics of the phototransformation of MBBA molecules are determined by comparing the experimental data with calculations. These characteristics are the lifetime, relative change of the polarization during the phototransformation, and the diffusion coefficient.

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INTRODUCTION

Nonlinear optical properties of liquid crystals are being actively investigated of late.¹ Particular interest attaches to the investigation of the so-called “giant” nonlinearity of nematic liquid crystals (NLC). Anomalously high values of the parameters of the cubic nonlinearity in these media,^{1) $\epsilon_3 \approx (10^{-2}-10) \text{ cm}^3 \cdot \text{erg}^{-1}$, have been observed in experiments on the self-focusing of light,²⁻⁴ nonlinear optical activity,⁵ and when methods of nonlinear active spectroscopy⁶ and dynamic holography⁷ are used. The strong cubic nonlinearity observed in Refs. 2–6 was due to a high-frequency analog of the Kerr effect (rotation of the NLC director \mathbf{n} by the electromagnetic-wave field), whose large value in the liquid-crystal phase is due to the hindering of the relaxation processes compared with the isotropic phase, owing to the strong orienting interaction between the molecules. This effect was observed in the mesophase at field intensities $\mathbf{E} \approx (10^2-10^3) \text{ V/cm}$, as against 10^5 V/cm in the isotropic phase.⁸}

The possibility of recording dynamic holographic gratings in a nematic mixture of cyanbiphenyls⁷ ($\epsilon_3 \approx 10 \text{ cm}^3 \cdot \text{erg}^{-1}$) by light beams with ordinary polarization, at a field intensity $\approx 1 \text{ V/cm}$ much lower than the threshold value of the Fréedericksz transition in these media,⁹ points to the existence of at least one more mechanism of the giant nonlinearity of NLC.

We have investigated a new type of “giant” nonlinearity of a nematic liquid crystal [*p*-methoxybenzylindene-*p*-butylaniline (MBBA)] by determining the characteristics of diffraction and self-diffraction of radiation by optically induced dynamic holographic gratings. The use of this procedure, which makes it easy to register refractive-index changes of the order of 10^{-5} , has made possible to measure the acting radiation at low densities ($I \approx 10^{-1} \text{ W/cm}^2$). The data obtained indicate that near the edge of the intrinsic absorption of MBBA there appears an extremely strong cubic nonlinearity due to reversible phototransformations of the liquid-crystal molecules. From the experimental dependences we determined certain characteristics of the phototransformed form: its lifetime, diffusion coefficient,

change of polarizability of the phototransformed form compared with the initial one.

1. DYNAMIC HOLOGRAPHIC GRATINGS BASED ON PHOTOSTIMULATED TRANSFORMATIONS OF MOLECULES

It is known that the action of light can cause reversible structural transformations of molecules.¹⁰ Included among these photostimulated transformations are the formation of hydrogen bonds, dimerization of molecules, and their trans-cis isomerization.¹¹ It is also known that the MBBA molecules have several metastable states corresponding to different relative locations of the benzene rings,¹² thus pointing to the possibility of such a photo-isomerization. The polarizability of the new form in all these transformations can differ substantially from that of the initial one, and this leads macroscopically to optically induced changes of the refractive index of the medium.

Assume that two coherent light beams with intensities I_1 and I_2 intersect in the volume of an NLC at an angle 2θ and produce a spatial intensity variation

$$I^{o,e}(x) = I_0^{o,e} [1 + m \cos(2\pi x/\Lambda)], \quad (1)$$

where

$$m = 2(I_1 I_2)^{1/2} / (I_1 + I_2), \\ I_0 = I_1 + I_2, \quad \Lambda = \lambda / 2 \sin \theta.$$

The indices “o” and “e” designate the polarization of the writing waves. The number of phototransformations of the molecules per unit time is $k^{o,e} \alpha^{o,e} I^{o,e} / h\nu$, where $\alpha^{o,e}$ is the absorption coefficient and $k^{o,e}$ is the quantum yield of the process. The molecules of the new type can go over spontaneously into ordinary molecules in a characteristic time T ; they can also diffuse with a diffusion coefficient D that can depend on the direction. Both processes should lead to erasure of the refractive-index-variation grating.

Assuming that the period Λ of the diffraction grating is large compared with the cell thickness d , which in turn is much less than the transverse dimensions of the writing beams, and assuming also that the NLC sample

is optically thin ($d \ll \alpha^{-1}$), we can find the spatial distribution of the molecules of the photostimulated form $N(x, t)$ by solving the one-dimensional diffusion equation

$$\frac{\partial N}{\partial t} = k^{o,e} \alpha^{o,e} \frac{I^{o,e}}{h\nu} - \frac{N}{T} + D_x \frac{\partial^2 N}{\partial x^2}. \quad (2)$$

Confining ourselves to the spatial harmonic that coincides with the exciting interference field, we find that the induced change in the refractive index is described in the stationary case by the expression

$$\Delta n^{o,e} = \frac{2\pi}{\langle n \rangle} \Delta \beta^{o,e} k^{o,e} \alpha^{o,e} T \frac{I_0^{o,e}}{h\nu} \left(1 + \frac{m}{1 + D_x q^2 T} \cos qx \right), \quad (3)$$

where $\Delta \beta^{o,e}$ is the difference between the polarizabilities of the MBBA molecule in the phototransformed and initial forms and $|\vec{q}| = 2\pi/\Lambda$ is the modulus of the vector of the holographic grating (the spatial frequency).

The optically induced change Δn is proportional in this model to the light power absorbed by the NLC. The cubic-nonlinearity constant

$$\varepsilon_2 = k^{o,e} \alpha^{o,e} \Delta \beta^{o,e} \langle n \rangle \frac{1}{h} \left(\frac{1}{T} + D_x q^2 \right)^{-1} \quad (4)$$

is a function of the light polarization and depends on the spatial frequency. Its dispersion is determined by the product $\Delta \beta^{o,e} \alpha^{o,e}$, i.e., it depends on the absorption spectra of the initial and phototransformed forms.

In the case of small phase advances, the efficiency of the holographic grating, or the ratio of the intensity of the radiation diffracted in the first order I_d to the intensity of the reading beam I_r , is determined by the square of the phase advance over the thickness d of the grating¹³:

$$\eta = I_d / I_r = (\pi \Delta n^{o,e} d / 2\lambda)^2. \quad (5)$$

It follows from (3) and (5) that the diffraction efficiency of a dynamic hologram should increase in proportion to the square of the intensity of the writing radiation, while the intensity of the diffracted radiation in self-diffraction is proportional to the cube of this intensity. The constant ε_2 of the cubic nonlinearity should be expressed here in terms of measurable quantities:

$$\varepsilon_2 = \frac{2\lambda \langle n \rangle}{\pi d} \left(\frac{I_d}{I_r^3} \right)^{1/2}. \quad (6)$$

It is seen from (3) and (5) that the efficiency of holographic recording is determined by the absorption coefficient, and since in the case of NLC the coefficients for waves with different polarization can differ substantially, the writing efficiency should depend on the polarization of the writing beams. Most single-component liquid crystals [MBBA, *p*-azoxyanisole (PAA), 4, 4'-di-*n*-heptyloxyazoxybenzene (HOAB) and others] have positive absorption dichroism, and the largest nonlinearity should be observed in them when the writing radiation has extraordinary polarization.

The stationary value of the diffraction efficiency, as seen from relations (3) and (5), is a function of the period of the holographic grating. The efficiency does not depend on the convergence angle of the writing beams at small θ ($\Lambda \gg \sqrt{DT}$), and decreases at large θ ($\Lambda \ll \sqrt{DT}$):

$$\eta \sim \left(\frac{\Lambda^2}{\Lambda^2 + 4\pi^2 DT} \right)^2. \quad (7)$$

The writing and erasure of the grating, as follows from the solution of Eq. (2), are purely exponential processes and are described respectively by the equations

$$I_d = I_{d0}(1 - e^{-2t/\tau}), \quad I_d = I_{d0}(e^{-2t/\tau}), \quad (8)$$

where I_{d0} is the stationary value of the intensity of the diffracted radiation, defined by expression (5). The erasure time $\tau = 2(1/T + Dq^2)^{-1}$ does not depend in the approximation considered on the light intensity. Expression (8) points to different mechanisms of grating erasure at high and low spatial frequencies. In the case of small periods ($\Lambda \ll \sqrt{DT}$) the grating should vanish because of the decrease, with time, of the modulation coefficient of the refractive index, owing to diffusion of the phototransformed molecules. The total number of phototransformed molecules can remain unchanged in this case. In the case of long periods ($\Lambda \gg \sqrt{DT}$) the erasure time is determined by the lifetime of the phototransformed molecules and should not depend on the period Λ .

2. EXPERIMENT

The experimental layout for the writing of the holograms is shown in Fig. 1. A helium-cadmium ($\lambda = 0.44 \mu\text{m}$) or argon ($\lambda = 0.456, 0.46, 0.488 \mu\text{m}$) laser, operating in the lowest transverse mode, was used for the writing. The holograms were written in accordance with the standard two-beam scheme; after passing through a beam splitter, two beams of equal intensity were guided by a long-focus lens ($F = 1 \text{ m}$) to the plane of the liquid crystal. The grating vector \vec{q} was perpendicular to the director of the liquid crystal. Since diffraction of the Raman-Nath type¹³ was realized in all the experiments, the writing of the holograms could be judged both by the diffraction of the testing beam, which was incoherent with respect to the writing beam, and by the appearance of non-Bragg self-diffraction orders. The polarizers P_1 and P_2 were used to measure the intensity and polarization of the writing radiation. The polarizers P_3 and P_4 made it possible, in the case of an unpolarized radiation source, to establish independently the polarization of the writing and testing radiations. The polarizer P_5 was used to investigate the polarization state of the diffracted radiation. The medium for writing the holograms was a plane-oriented nematic MBBA liquid crystal placed in a 65- μm cell with orienting covers and kept at a temperature 22 °C.

The diffraction efficiency of the holograms reached its stationary value within several tenths of a second, and the grating was most effectively written by radiation with extraordinary polarization. The dependence of the

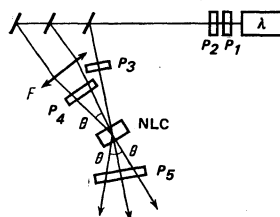


FIG. 1. Layout of experiment.

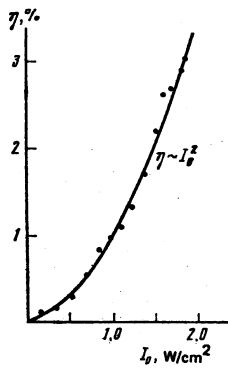


FIG. 2. Dependence of the diffraction efficiency η on the intensity I_0 of the writing beams.

stationary diffraction efficiency on the intensity of writing radiation with extraordinary polarization (the exposure characteristic) is shown in Fig. 2. In the intensity range up to 2 W/cm² the MBBA crystal, as expected, behaves as a medium with cubic nonlinearity. When radiation with $\lambda = 0.456 \mu\text{m}$ and high power $I_0 > 4 \text{ W/cm}^2$ is recorded, saturation of the diffraction efficiency is observed, and at $I_0 > 20 \text{ W/cm}^2$ the efficiency even decreases with increasing intensity.

The spectral dependence of the parameter of the cubic nonlinearity is shown in Fig. 3a. In accordance with (3) and (4), the spectral variation of $\epsilon_2(\lambda)$ correlates with the absorption spectrum of the MBBA nematic phase.

The dependence of the stationary diffraction efficiency on the grating period (the frequency-contrast characteristic) is shown in Fig. 4. The diffraction efficiency increases nonlinearly for small periods and has a tendency to saturate with increasing Λ in accord with expression (7). The oscillations on the general monotonic curve exceed the measurement error. When the experiment is repeated multiply at various points of the sample, the oscillations are preserved, although their period and arrangement may change.

An investigation of the kinetics of the grating decay has shown that the initial erasure section, all the way to intensity values $I_d = 0.25 I_{d0}$, is well described by an exponential function with a characteristic time of the order of several tenths of a second and dependent on the period of the holographic grating (Fig. 5). At small periods, the time constant increases with increasing period, and at large it approaches a constant value, which equals, within the framework of the one-dimensional treatment, half the lifetime of the phototransformed form [see (8)]. The subsequent erasure stage

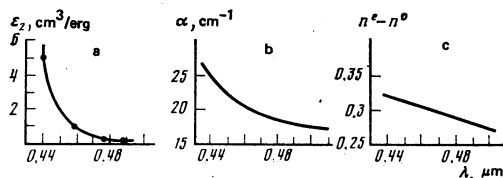


FIG. 3. Spectral dependences for the liquid crystal MBBA: a) of the cubic-nonlinearity parameter $\epsilon_2(\lambda)$; b) of the absorption coefficients $\alpha(\lambda)$; c) of the birefringence ($n^e - n^o$) (λ).

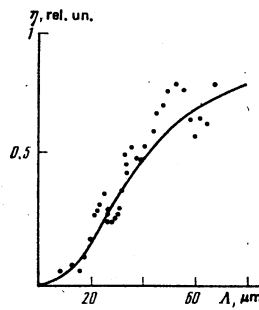


FIG. 4. Dependence of the diffraction efficiency η on the period of the holographic grating (solid line—calculation based on the values $D = 1.3 \times 10^{-7} \text{ cm}^2 \cdot \text{sec}^{-1}$ and $T = 1.6 \text{ sec}$).

is characterized by a much longer relaxation time ($\tau > 10 \text{ sec}$), but owing to the low level of the signal relative to the scattered light its detailed analysis is difficult.

Figure 6a shows the dependence of the intensity of the diffraction of the testing radiation with extraordinary (curve 1) and ordinary (curve 2) polarizations on the angle φ between the direction of the NLC director and the intensity vector of the writing field. At a fixed value of the angle φ , unequal changes take place in the extraordinary and ordinary refractive indices. The grating is written most effectively, as follows from (3), by the radiation with extraordinary polarization. Within the framework of the proposed model, the dependence obtained is due to the dependence of the absorption coefficient on the angle φ . The ratio of the values of curves 1 and 2 at a given angle φ should then be constant at $(\Delta\beta^e\alpha^e/\Delta\beta^o\alpha^o)$ [see (3)]. Figure 6b shows the dependence of this ratio on the angle φ . It is seen that it remains constant within the limits of error.

To complete the description of the experimental results, it must be pointed out that despite the good reproducibility of the results for each given NLC, the values obtained for two different samples could differ by approximately 30%.

3. DISCUSSION OF RESULTS

As follows from the preceding section, the experimental data agree well with the predictions of the model of photostimulated transformations of NLC, and at the same time both quantitative and qualitative deviations are observed from the model of self-induced rotation of the molecules. First, the writing is observed at pow-

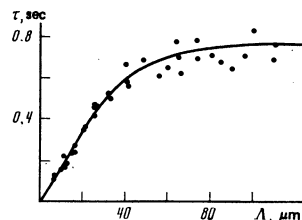


FIG. 5. Dependence of the characteristic erasure time τ of a holographic grating on the period Λ of the interference pattern (solid line—calculation based on the values $D_{\perp} = 1.3 \times 10^{-7} \text{ cm}^2 \cdot \text{sec}^{-1}$ and $T = 1.6 \text{ sec}$).

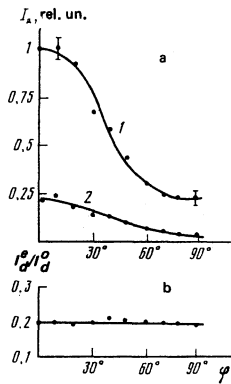


FIG. 6. a) Dependence of the intensity of the diffraction of the testing extraordinary radiation I_d^e (curve 1) and ordinary I_d^o radiation (curve 2) on the angle φ between the NLC director and the polarization of the writing radiation; b) dependence of the ratio I_d^e/I_d^o on the angle φ .

er densities that are patently insufficient to reorient the molecules (the energy of the optical action $\Delta\epsilon |\bar{E}|^2 / 4\pi \approx 10^{-5}$ erg/cm³ is considerably lower than the energy of the elastic interaction of the NLC molecules $\pi^2 K_{11} / d^2 \approx 10^{-1}$ erg/cm³, where K_{11} is the Frank elastic modulus).¹⁴ Second, the effect is a maximum when the crystal is acted upon by beams with extraordinary polarization ($E \parallel n$), when the twist is identically equal to zero. Third, the sign of the nonlinear refractive index does not depend on the polarization of the exciting light, a fact established in experiments on which polarization holograms were written in MBBA. Fourth, the orientational model presupposes for the nonlinearity parameter a spectral dependence² $\Delta\epsilon(\lambda)$ (see Fig. 3a) that differs substantially from that obtained in experiment.

It should be noted that in the thermal writing mechanism, the characteristics of the diffraction should be qualitatively similar to those observed in our experiment. In the stationary regime, the cubic nonlinearity constant is in this case

$$\epsilon_2^T = \frac{c\alpha^{0,e}}{4\pi Kq^2} \left(\frac{\partial n}{\partial T} \right)_p^{0,e}, \quad (9)$$

where K is the thermal conductivity coefficient, $(\partial n / \partial T)_p^{0,e}$ is the thermo-optical constant, and c is the speed of light. The characteristic grating erasure time is determined by the rate of thermal diffusion

$$\tau_T = 2/D_T q^2. \quad (10)$$

The quantities calculated on the basis of (9) and (10) at $q = 6 \times 10^3$ cm⁻¹ and at the values typical of MBBA, namely $K = 2.1 \times 10^4$ (erg·cm⁻¹·sec⁻¹·deg⁻¹), $D_T = 1.5 \times 10^{-3}$ cm²·sec⁻¹ (Ref. 15), $(\partial n / \partial T)_p^{0,e} \approx 2 \times 10^{-4}$ deg⁻¹ (Ref. 16), and $\alpha \approx 25$ cm⁻¹ ($\lambda = 0.44$ μ m), namely $\epsilon_2 \approx 5 \times 10^{-5}$ cm³/erg and $\tau \approx 5 \times 10^{-5}$ sec, turn out to be lower by respectively three and five orders than those obtained in our experiment. So large a quantitative discrepancy allows us to exclude the thermal nonlinearity mechanism from consideration.⁵⁾ We arrive thus at the conclusion that the optical nonlinearity is indeed due to photostimulated transformation of the NLC molecules.

It is phenomenologically customary to describe a medium with cubic nonlinearity by a fourth-rank non-

linear-susceptibility tensor: $P_i = \chi_{ijkl} E_j E_k E_l$. Since the change of the refractive index is due to absorption of light, it follows that introduction of such a tensor is strictly speaking incorrect. However, if we express the material polarization, responsible for the diffraction effects connected with the phototransformations, in the form

$$P_i \sim \alpha_{ij} E_j \Delta \beta_{kl} E_k E_l, \quad (11)$$

where α_{ij} is the NLC absorption tensor and $\Delta \beta_{kl}$ is the tensor of the difference of the susceptibilities of the phototransformed and initial forms, we can introduce formally a nonlinear susceptibility tensor $\chi_{ijkl} = \alpha_{ij} \Delta \beta_{kl}$, the rules for whose use are contained in expression (11) and differ from those traditionally applied to nonlinear-susceptibility tensors.¹⁷

Since nematic crystals have a symmetry characterized by the $D_{\infty h}$ group, the tensor α_{ij} can be diagonalized, with $\alpha_{11} = \alpha_{22}$ and $\alpha_{33} \neq \alpha_{11}$. If after the phototransformation the crystal remains nematic with the same axis and symmetry group, then the tensor $\Delta \beta_{kl}$ has a structure similar to the tensor α_{ij} , and the tensor χ_{ijkl} has only four independent components:

- 1) $\chi_{1111} = \alpha_{11} \Delta \beta_{11} = \alpha_{22} \Delta \beta_{22} = \alpha_{11} \Delta \beta_{33} = \alpha_{22} \Delta \beta_{33}$,
- 2) $\chi_{3333} = \alpha_{33} \Delta \beta_{33}$,
- 3) $\chi_{1133} = \alpha_{11} \Delta \beta_{33} = \alpha_{22} \Delta \beta_{33}$,
- 4) $\chi_{3311} = \alpha_{33} \Delta \beta_{11} = \alpha_{33} \Delta \beta_{22}$.

In a coordinate system in which the z axis coincides with the NLC director n , the first two components determine the diffraction of a testing beam having the same polarization as the writing waves, whereas the last two describe diffraction with differing polarizations. Therefore, the experimental data (see Fig. 6) can be used to determine the ratios of all four independent components of the nonlinear-susceptibility tensor

$$|\chi_{1111}| \approx 0.19 |\chi_{3333}|, \quad |\chi_{1133}| = |\chi_{3311}| \approx 0.44 |\chi_{3333}|$$

and, knowing ϵ_2 , to determine their absolute values ($\lambda = 0.44$ μ m, $\Lambda = 13$ μ m):

$$|\chi_{3333}| \approx 0.63 \text{ cm}^3 \cdot \text{erg}^{-1}, \quad |\chi_{1111}| \approx 0.12 \text{ cm}^3 \cdot \text{erg}^{-1}, \quad |\chi_{1133}| = |\chi_{3311}| \approx 0.28 \text{ cm}^3 \cdot \text{erg}^{-1}.$$

The diffraction intensity is proportional to the square of χ_{ijkl} , therefore the signs of the tensor components cannot be determined from measurements of the diffraction efficiency. However, when the gratings are written by radiation whose polarization has ordinary and extraordinary components, the diffraction intensity is proportional to the square of a linear combination of the corresponding components, so that it is possible to ascertain whether they are of equal sign. An analysis of the curves shown in Fig. 6 has shown that the components χ_{1111} and χ_{3311} are of the same sign. The same holds also for the pair χ_{1133} and χ_{3333} . It must be stated here that the signs of both pairs can differ. This result agrees with the conclusions that follow from the characteristics of the polarization writing in MBBA and with the predictions of the considered nonlinearity model.

A comparison of the experimental results with calculation makes it also possible to determine certain other characteristics of the phototransformed form.

As indicated above, at low spatial frequencies the

grating erasure time is independent of the grating period and is determined by the lifetime of the phototransformed form. Therefore, starting from the experimental results shown in Fig. 5, this time can be estimated at $T = (1.6 \pm 0.3)$ sec.

It should be indicated, however, that this estimate is valid if it is assumed that the cell walls do not influence the relaxation of the phototransformed form. The measured lifetime can also be connected with the deactivation of the phototransformed form on the cell walls, since the diffusion length becomes comparable with the cell thickness at the spatial frequencies corresponding to saturation of the contrast vs time characteristic.

Knowing the lifetime of the phototransformed form and using relation (18), we can determine the diffusion coefficient of the excited MBBA molecule $D_1 = (1.3 \pm 0.3) \times 10^{-7} \text{ cm}^2 \cdot \text{sec}^{-1}$. The solid line in Fig. 5 shows the calculated dependence of the grating lifetime on its period under the assumption that $T = 1.6$ sec and $D_1 = 1.3 \times 10^{-7} \text{ cm}^2 \cdot \text{sec}^{-1}$. The obtained values of T and D_1 were used also to calculate the frequency-contrast characteristic of the hologram (solid line in Fig. 4), which likewise agrees with experiment.

Change of the molecule polarizability by the phototransformation. The physical cause of the change of the refractive index under phototransformation is the change of the polarizability of the new form compared with the polarizability of the initial NLC molecule. If it is assumed that the concentration of the excited molecules is so low that it does not change the local field in the liquid crystal then, assuming this field in the form proposed by Vuks,¹⁸ we obtain the following expression for the relative increment $\Delta\beta^{o,e}$ of the polarizability of an individual MBBA molecule following phototransformation

$$\frac{\Delta\beta^{o,e}}{\beta_0^{o,e}} = \frac{2n^{o,e}}{(n^{o,e})^2 - 1} \frac{N_0}{N} \Delta n^{o,e},$$

where $\beta_0^{o,e}$ is the polarizability of the unexcited MBBA molecule, N_0 is the concentration of the main MBBA molecules, and $N = \alpha I / (1/T + D_x q^2)$ is the stationary concentration of the phototransformed MBBA molecules. Substitution of the numerical values for $\lambda = 0.44 \mu\text{m}$ yields $\Delta\beta/\beta_0 \approx 9 \times 10^{-2}$.

We note that to explain certain experimental data it is necessary to make additional assumptions concerning the character of the phototransformations of the MBBA molecules. Thus, we have not observed self-diffraction above the phase-transition point. One can expect either a decrease of the probability of the phototransformation in an isotropic liquid, or else a substantial shortening of the lifetime of the new form.

An explanation is needed also for the variation of the frequency vs contrast characteristic of the holograms. The probable cause of the oscillations may be a pure interference effect, but the quality of the interferometer made up of the planes of the cell is insufficient to obtain oscillations of the observed magnitude. It is possible that the form of curve is connected with various

boundary conditions at different points of the sample, owing to the varying profile of the orienting surface.

CONCLUSION

The aggregate of the experimental data allows us to state that the observed nonlinearity mechanism is connected with the photostimulated transformation of the NLC molecules.

The MBBA crystal is characterized by anomalously large values of the cubic-nonlinearity parameter ($\epsilon_2 \approx 5 \times 10^{-2} \text{ cm}^3 \cdot \text{erg}^{-1}$ for $\lambda = 0.44 \mu\text{m}$ and $\Lambda = 13 \mu\text{m}$) and by a sensitivity to the polarization of the writing radiation. This makes its use promising as a dynamic medium for writing ordinary and polarization holograms by continuous radiation from low-power lasers.

The considered nonlinearity mechanism is not an exclusive feature of either MBBA or of the class of nematics. Dynamic gratings with similar properties were observed also in a mixture of cyanobiphenyls⁷ and MBBA colored with methylene red.¹⁰ The photoinduced cis-trans transition of the liquid-crystal molecule in the smectic phase leads to a local phase transition into a nematic phase,¹⁹ and this also manifests itself in a large change of the optical constants.

The description of the nature of the phototransformed form in each concrete case calls for a special research. It is quite possible that for different liquid crystals they turn out to be different. It is clear even now, however, that by applying light it is possible to produce molecules of new form and to regulate their concentration. Extremely promising in this connection, is, e.g., the possibility of controlling the structure of liquid-crystal polymers during the stage of their polymerization.

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¹⁾ $\epsilon_2 = 2\Delta n \langle n \rangle / |E|^2$, where Δn is the optically induced change of the refractive index and $\langle n \rangle = (1/3)(2n^o + n^e)$ is the average refractive index, and n^o and n^e are the refractive indices of the ordinary and extraordinary waves. E is the light-wave field strength.

²⁾ The thermo-optical constants of MBBA are increased by several orders of magnitude near the phase-transition point.¹⁵ This gives ground for hoping to be able to write effectively thermal gratings in the pre-transition region.

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