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SELF-ACTION OF A LIGHT BEAM IN NEMATIC LIQUID CRYSTALS IN A DC ELECTRIC FIELD

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The aberrational self-action of a light beam in nematic liquid crystals (an undoped nematic host and samples doped with amino acids and dyes) in the presence of an external dc electric field has been studied experimentally. The aberrational pattern and the dynamics of its development were found to be dependent on the liquid crystal composition and experimental conditions and practically independent of the polarization of light. Unlike the self-action in the absence of a dc field, the aberrational pattern can have a complex asymmetric structure. The self-action observed can be explained by the surface photorefractive effect.

Introduction. The light-induced reorientation of the director of nematic liquid crystals (NLCs) results in an aberrational self-action of a light beam [1 – 3]. In transparent NLCs, the high efficiency of this self-action is due to the "giant" orientational nonlinearity of transparent NLCs, which exceeds the Kerr nonlinearity of liquids by nine orders of magnitude [4]. Adding a small amount ($\sim 0.1\%$) of light-absorbing dopants into a nematic host can increase further the orientational nonlinearity by two orders of magnitude and even change the sign of nonlinearity [5].

External low-frequency alternating fields affect the self-action of light in NLCs, suppressing or enhancing it depending on the sign of dielectric anisotropy and the experimental geometry [6, 7].

The aberrational pattern is in this case a system of concentric rings, which can be elongated perpendicularly to the direction of the incident radiation polarization (the degree of

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elongation is no more than 40%) [8]. Because the distribution of intensity and the polarization in the aberrational pattern carry information on the deformed director field, the self-action of light is a highly convenient and informative tool in nonlinear optics of liquid crystals [9 – 11].

Considerable interest is aroused by the light-induced orientational phenomena in NLCs subjected to a dc electric field [12 – 16]. These photorefractive type phenomena are due to redistribution of the charge carriers in an NLC cell produced by the combined action of the dc electric field and the light wave (the charges are either initially present in the nematic host or generated upon excitation of absorbing impurities). The corresponding orientational nonlinearity can greatly exceed the "giant" orientational nonlinearity.

So far the light interaction with NLCs in a dc field has only been studied for plane light waves (the transverse dimension of the beam is much larger than the crystal thickness), mainly with the aid of multiwave processes. We report here on the observation and investigation of an efficient self-action of a narrow light beam in such NLCs.

1. Experimental samples and experimental conditions. We studied a pure (undoped) nematic mixture ZhKM-1277 and this host doped with small amounts of various impurities such as amino acids and dyes.

The nematic matrix ZhKM-1277 is a mixture of biphenyls and esters. The dielectric anisotropy of this material is positive ($\Delta\varepsilon = 12.1$ at the frequency of 1 kHz and the temperature $t = 20\,^\circ C$); the extraordinary and ordinary refractive indices are, respectively, 1.71 and 1.52 ($\lambda = 589\,nm$, $t = 20\,^\circ C$).

The nematic host was doped with amino acids (asparagine (Asp) and tyrosine (Tyr)) which can change the ion concentration in solutions [17]. The dyes used were methyl red (MR), which can affect the electric conductivity [18], and rhodamine 6G (R6G), exhibiting (according to [12]) photoconductivity.

The experiments were carried out on five planar aligned samples: (1) ZhKM-1277 with the thickness $L = 50\,\mu m$; (2, 3) ZhKM-1277+0.2 wt% (2) Asp or (3) Tyr ($L =$(2) 50 or (3) 100 μm); (4) ZhKM-1277+0.1 wt% MR ($L = 100\,\mu m$); and (5) ZhKM-1277+0.05 wt % R6G ($L = 100\,\mu m$). The homeotropically aligned 100-μm ZhKM-1277+0.05 wt % R6G cell was also studied (sample 6). Planar alignment of the samples was obtained by buffing the polyimide coating of the glass substrates of the cells. Homeotropic alignment was obtained with the aid of chromium stearyl chloride. A dc electric field was applied to the ITO electrodes deposited on glass substrates.

The experimental setup was similar to that used in [19]. The radiation of a solid-state LASOS-GL laser ($\lambda = 532\,nm$), an argon LASOS laser ($\lambda = 515\,nm$), or an argon-krypton ILM-120 laser ($\lambda = 647$, 515, 488 nm) was focused by a lens ($f = 14\,cm$, the size of the laser beam waist was about 100 μm) into an NLC cell. To change the polarization of the incident light beam, we used a double Fresnel rhomb. The plane of the NLC cell was vertical; the

NLC director **n** was in the horizontal plane. The angle α of the light incidence on NLC was changed by rotating the cell about the vertical axis (the angle α is taken as positive if the cell is rotated counterclockwise and as negative for the clockwise rotation). In the above geometry, the extraordinary wave is excited in NLC if the polarization of the incident light is horizontal. A screen to observe the changes in the light beam in the course of its interaction with NLC was placed behind the cell. The sign of the self-action was determined by the analysis of the aberrational-pattern transformation upon transverse displacement of the NLC cell with respect to the light beam [11]. The polarity of the applied dc voltage will be taken as positive if the input (for the light beam) cell surface is the anode.

2. Experimental results.

2.1. Undoped nematic matrix ZhKM-1277. At the normal incidence of a horizontally polarized light beam (with power $P = 1 - 10\,mW$) on an NLC, no aberrational pattern is observed without a dc field. When a dc field ($U \sim 2\,V$) is applied to the cell, the ring-shaped aberrational pattern of the light beam self-defocusing is developed within $\sim 1\,min$. Then the pattern collapses slowly within $\sim 10\,min$. The time of aberrational-pattern relaxation after switching off the light or dc electric fields is $1 - 2\ min$. The aberrational pattern is not fully symmetric with respect to the vertical line passing through its center because one of the halves is somewhat elongated along the horizontal.

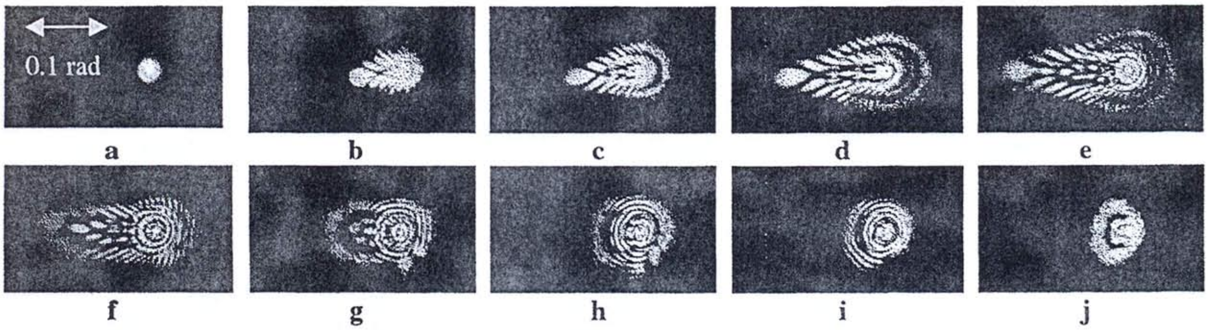

Fig. 1. Aberrational pattern dynamics in the light beam passing through sample 1 (NLC ZhKM-1277) under a simultaneous action of dc and light fields ($L = 50\,\mu m$, $U = +2\,V$, $\alpha = +40°$, $\lambda = 515\,nm$, $P = 5\,mW$). Time intervals between snapshots are 5-10 s.

In the case of oblique incidence, the observed self-action of the light beam is much more complex. The dynamics of the self-action development is shown in Fig. 1. After the application of a dc field, a "petal" elongated along the horizontal is formed within several seconds (Fig. 1b). The center of the wide part of the "petal" coincides with the center of the beam prior to voltage application. Then, the petal increases in dimensions and, along with it, the second structure (Figs. 1c – 1e) appears. This structure consists of rings elongated toward the "petal". The sign of the self-action of the second structure is negative (self-defocusing). Further, the petal itself transforms into the ring structure (Figs. 1f – 1g), its

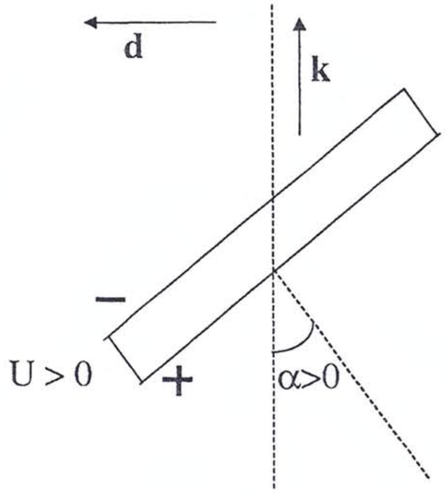

Fig. 2. Schematic representation of beam deflection (view from above): **k** is the wave vector of the light beam incident on the NLC, α is the angle of light beam incidence on the NLC, U is the dc voltage applied to the NLC, **d** is the aberrational-pattern elongation vector. The change in the sign of α or U results in the change in the direction of the vector **d**.

center moves to the center of the second structure (Figs. 1h – 1i). Finally, both ring systems merge together (Fig. 1j).

The elongation direction of the self-action pattern (i.e., the direction from the center of the second (ring-shaped) system to the petal center) depends, in the case of oblique incidence, on the sense of the crystal rotation (the sign of the angle α) and the applied voltage polarity. For $\alpha > 0$ and $U > 0$, the pattern is elongated to the left (Fig. 2).

The rate of development and the size of the aberrational pattern depend on the beam power P and voltage U. At a low voltage ($U \sim 1\,V$), the pattern becomes asymmetric (as in Fig. 1b) and then remains unchanged. As the beam power increases to $P = 10\,mW$, the steady-state asymmetric pattern increases in size. This pattern does not change upon a further increase in the power P. For $U \sim 2\,V$ and the beam power higher than 8 mW, the asymmetric pattern transforms rapidly ($\sim 10\,s$) into the ring-shaped one. In all the above cases, the polarization of the aberrational pattern was horizontal (that is, coincident with the linear polarization of the incident radiation).

The light beam self-action efficiency increases with decreasing wavelength of light. Thus, for $U = 2\,V$ and $P = 4\,mW$, no aberrational pattern was observed for the red line $\lambda = 647\,nm$. For the green line $\lambda = 515\,nm$, the horizontal divergence of the pattern was 0.3 rad; for $\lambda = 488\,nm$, this divergence was 0.6 rad.

We investigated the dependence of the light self-action on the incident light beam polarization. The rotation of the polarization plane from the horizontal direction was not found to affect the shape, polarization (horizontal), and the development of the aberrational-pattern. This rotation only changes the pattern brightness and results in the occurrence of a bright spot with vertical polarization at the point corresponding to the position of the light beam at the initial moment of irradiation. The aberrational pattern intensity is maximum for the horizontal radiation polarization (extraordinary wave) and vanishes for the vertical polarization (ordinary wave). The intensity of the vertically polarized bright spot shows an opposite polarization dependence. If the ordinary wave is incident on the NLC, the aberrational pattern is not seen. However, the pattern can easily be visualized at any moment by slightly (by $\sim 5°$) rotating the polarization plane of the incident radiation.

It should be emphasized that the clearly pronounced asymmetry of the aberrational pattern of the self-action of light and independence of its shape of the polarization of incident light are related to the effect of the dc field. In the case of orientational self-action of light beams in transparent and absorbing crystals in the absence of a dc field, the aberrational pattern is a system of rings and, in the experimental geometry considered, the divergence and ring number of this pattern decrease monotonically upon rotation of the polarization plane from the horizontal to the vertical direction.

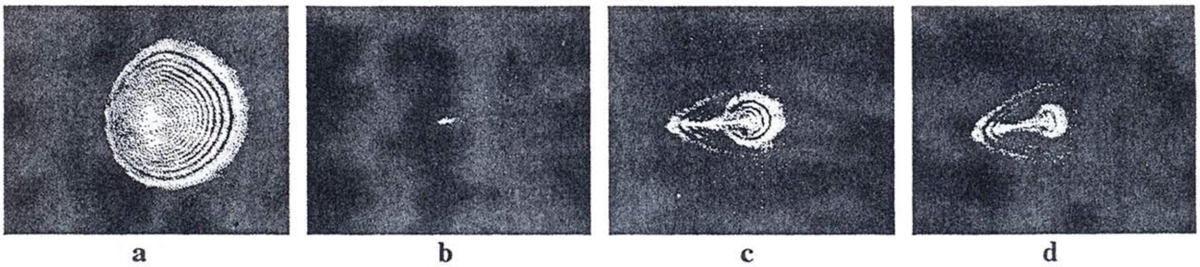

Fig. 3. Aberrational patterns for sample 3 (ZhKM-1277+0.2% Asp) developed in 1 *min* after the application of a dc field for (a, b) normal and (c, d) oblique ($\alpha = +40°$) incidence of the light beam on the crystal: (a, c) extraordinary wave and (b, d) ordinary wave ($P = 5\,mW$, $\lambda = 515\,nm$, $U = +2\,V$).

2.2. Nematic host ZhKM-1277 doped with amino acids. For these samples, no significant difference from an undoped host was noticed. For example, Fig. 3 illustrates independence of the aberrational-pattern shape of the incident-light polarization.

For sample 3, we measured the dependence of the horizontal dimension of the steady-state aberrational pattern on P and U. It was found that, for each angle α, the same dimension of the pattern can be obtained at different combinations of P and U. This is evident from Fig. 4 which shows the isolines of the horizontal divergence of the pattern. It follows from Fig. 4 that the nonlinear-optical response of NLC in dc field increases in passing from the normal to the oblique incidence of the light beam and grows rapidly with U.

2.3. Nematic host ZhKM-1277 doped with the dye "methyl red". A specific feature of sample 4 is that the ring-shaped structure was only observed for both the normal and oblique incidence of the light beam on NLC subjected to a dc field. However, as in the above cases, the pattern developed independently of the light polarization. Thus, for $U = +2\,V$, $\alpha = 40°$,

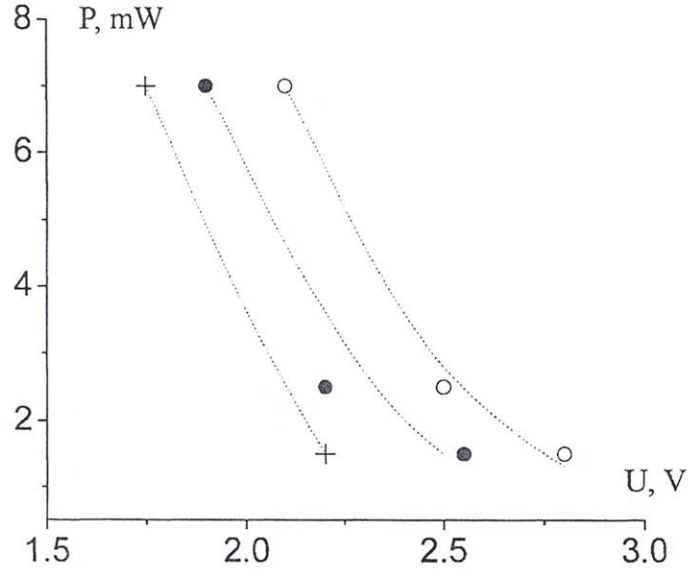

Fig. 4. Isolines of angular dimension θ_h of the aberrational pattern of the light beam self-action for sample 4 (ZhKM-1277+0.2% Tyr): (+) $\theta_h = 35°$, $\alpha = +50°$; (•) $\theta_h = 18°$, $\alpha = +50°$; (○) $\theta_h = 35°$, $\alpha = 0°$.

and $P = 3\,mW$ the number N of the self-defocusing rings increases from 3 to 12 for horizontal beam polarization (extraordinary wave) and from 0 to 9 for vertical polarization (ordinary wave). The initial three rings in the field of the extraordinary wave occurred already prior to the application of the dc field; the characteristic time of their formation was $\sim 10\,s$. The appearance of these rings can naturally be related to the orientational nonlinearity of NLC due to changing the intermolecular forces upon light absorption by the "methyl red" molecules.

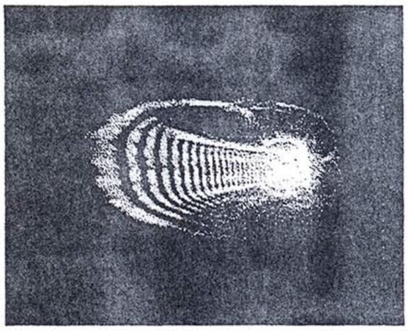

Fig. 5. Aberrational pattern for sample 5 (planar aligned ZhKM-1277 + 0.05% R6G) at $\alpha = +40°$, $P = 5\,mW$, $\lambda = 515\,nm$, and $U = +4\,V$.

2.4. Nematic host ZhKM-1277 doped with the dye "rhodamine 6G" (planar alignment). The aberrational pattern for sample 5 (ZhKM-1277+0.05 wt % R6G) is qualitatively similar to that for the undoped host. However, for oblique incidence the pattern is somewhat different (Fig. 5). An additional point to emphasize is that to obtain the well-developed pattern, the voltage should be higher ($U = 4\,V$) than for the indoped host ($U = 2\,V$). At the same time, the aberrational pattern was observed for very low radiation powers. Thus, for the beam power $P = 0.02\,mW$ we observed six rings of the aberrational self-defocusing ($U = 6\,V$, $\alpha = 0°$).

2.5. Nematic host ZhKM-1277 doped with the dye "rhodamine 6G" (homeotropic alignment). No aberrational pattern was observed for the normal incidence of the light beam on sample 6. No apparent influence of a dc field was found.

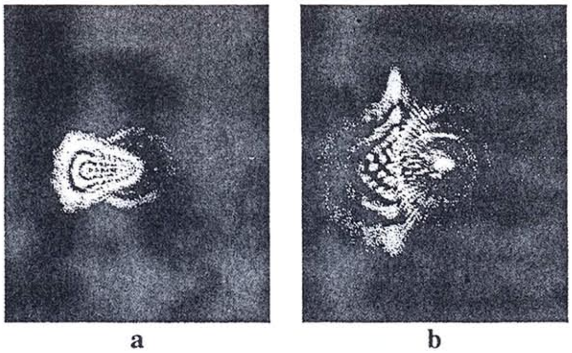

Fig. 6. Aberrational patterns for sample 6 (homeotropically aligned ZhKM-1277 + 0.05% R6G): (a) $P = 1\,mW$ and (b) $P = 5\,mW$ ($\alpha = +40°$, $U = +3\,V$, $\lambda = 515\,nm$). Voltage was applied in 2 *min* after starting the illumination.

For the oblique incidence ($\alpha = 40°$) of the light beam (extraordinary wave), the pattern of the aberrational self-focusing is first set in ($N = 1$ at $P = 1\,mW$ and $N = 5$ at $P = 5\,mW$). Then, this pattern collapses within $\sim 2\,min$. A subsequent application of a dc field gives rise to an asymmetric pattern (Figs. 6a and 6b). For $P = 1\,mW$, the rotation of the polarization plane (after collapse of the initial orientational ring) does not affect the pattern development. For $P = 5\,mW$, the pattern in the ordinary wave is qualitatively similar to that in the extraordinary wave. However, the size of this pattern is slightly smaller and the polarization plane rotation results in an increase of the pattern within several seconds.

Discussion. The occurrence of the aberrational pattern is related to the director reorientation, which is confirmed by the characteristic times of its establishment and relaxation. The low radiation power required for the development of the aberrational pattern and independence of the pattern of the polarization rule out the orientational phenomena due to the action of light on the induced dipoles [1 – 2] and due to changing the intermolecular forces [3]. One can assume the discussed orientational phenomenon to belong to the photorefractive phenomena – the director reorientation is due to the action of dc electric fields produced by electric charges whose occurrence is conditioned by the effect of the field of light.

In our experimental conditions, the appearance of the ions in the bulk crystal under the action of light [12] must not play an important role in the light-induced director reorientation because a strong effect was observed not only in NLCs doped with absorbing dyes (R6G and

MR), but also in the transparent nematic host. Moreover, the efficiency of the bulk charge generation must depend on the radiation polarization (because of absorption dichroism). At the same time, the aberrational pattern, as was noted above, depends weakly on the polarization. It should also be noted that the ion generation itself would result in an additional screening of the external field, which, in planar NLCs, would give rise to the self-focusing of the light beam. It only remains to assume, therefore, that the role of light reduces to changes in the concentration of the ions localized near nematic – polymeric coating interface under the action of an external dc electric field and screening this field. To confirm the presence of the screening effect, we additionally measured the Freedericksz transition threshold in dc and ac fields. For an undoped nematic host the threshold of the Freedericksz transition under the dc field was four times as high as that under the ac field (3.2 and 0.8 V, respectively).

The asymmetry of the aberrational pattern for an oblique incidence can apparently be explained by the difference in the surface charge densities near the anode and cathode under the action of light. In this case, the distribution of the resulting electric field in the NLC bulk (and, correspondingly, the director field) will be asymmetric with respect to the light beam axis. This explanation is supported by the change in the direction of the beam deflection with a change in the voltage polarity and in the sense of the cell rotation. The manifestation of aberrational-pattern asymmetry for both the planar and homeotropic samples indicates that the asymmetry effect is characteristic of diverse orientants.

The decrease in the self-action efficiency with increasing light wavelength can be explained in the assumption that the change in the surface charge density is due, for example, to the photoinduced injection of charge carriers from the orientant into the nematic layer. The nematic host doping with amino acids exerted no noticeable influence on the aberrational pattern because the small amount of the impurity that we managed to dissolve did not change much the concentration of the charge carriers in the NLCs. The lack of pronounced asymmetry in the case of NLC doping with methyl red needs further investigation.

Conclusions. A simultaneous action of a light beam and a dc field on nematic liquid crystals can result in the formation of a complex aberrational pattern in both planar and homeotropic samples. In this case, the NLC orientational nonlinearity exceeds the conventional “giant” optical nonlinearity by two or three orders of magnitude. The aberrational pattern and its behavior are qualitatively different for NLCs of different composition, but in all cases the aberrational pattern is independent of the light beam polarization. The elongation direction of the asymmetric aberrational pattern depends on the voltage polarity and the sign of the angle of light wave incidence. The phenomenon observed is likely to be due to the surface photorefractive effect, that is, to the change in the charge concentration at the interfaces between the NLC and the orienting surfaces.

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31 August 2005