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The interaction of light with a nematic liquid crystal (NLC) doped with dendrimers containing terminal light-absorbing azobenzene fragments and a low-molar-mass dye identical to the fragments was studied in detail. Both the dendrimers and the dye induce orientational nonlinearity in a transparent nematic host. For the fifth-generation dendrimer, the nonlinearity is negative (director is rotated away from the light field direction). For the first-generation dendrimer and the dye, the nonlinearity sign depends on the light propagation direction with respect to the NLC director, light polarisation and external electric field. The results obtained show that complication of the molecular structure of the dopant, i.e. passing from the 'free' dye molecule to the first- and fifth-generation dendrimers, results in an increase of the efficiency of the orienting effect of the light.

Keywords: nematic liquid crystal; dendrimer; optical nonlinearity; light-induced effect; self-phase modulation

1. Introduction

In recent years much attention has been paid to liquid crystalline materials containing supramolecular (nanoscale and high-molar-mass) admixtures. Such dopants affect, for example, the elastic, optical, dielectric and viscous properties of nematic liquid crystals (NLCs) (*1–5*). Nonlinear optical characteristics are also sensitive to supramolecular admixtures. Thus, incorporation of small amounts of fullerenes (*6–8*), nanotubes (*8*) and CdSe particles (*9*) increases the photorefractive nonlinearity of a nematic host. Comb-shaped polymers with light-absorbing azobenzene side chains were found to induce orientational nonlinearity, which is (at equal absorption coefficients) higher than the nonlinearity induced in the same nematic host by a dye similar in structure to the absorbing moieties of the polymers and the nonlinearities of other low-molar-mass dye–nematic systems (*10*). Dye-doped NLCs allow one to study a number of nonlinear phenomena (self-phase modulation, stimulated orientational scattering, bulk and surface photorefractivities, development and interaction of solitary waves, etc.) at low light beam power. These phenomena made orientational nonlinearity of NLCs attractive for various applications in nonlinear optics and photonics.

Quite recently, the synthesis and characterisation of a new class of macromolecular compounds, such as photochromic dendrimers, have been reported (*11–17*). The heavily branched structure of dendrimer molecules manifests itself in a set of unique properties, including the specific features of their photophysical (*11, 12*) and photochemical behaviour (*13–17*).

In this paper, the results are presented of a detailed investigation of the interaction of light with NLCs doped with dendrimers containing 8 and 128 light-absorbing azobenzene terminal fragments (G1 and G5, respectively) and the low-molar-mass dye D identical to the fragments. Initial results of this study were reported previously (*18*).

2. Experimental

The molecular structures of the substances investigated, i.e. the low-molar-mass dye (D) and dendrimers of the first (G1) and the fifth (G5) generations, are shown in Figure 1. The synthesis, phase behaviour and photo-optical properties of these substances in dilute isotropic solutions and thin films have been described previously (*15*). It can be seen from Figure 1 that the terminal fragments R of the dendrimers are identical to the dye D.

The liquid crystalline material ZhKM-1277 (NIOPIK, Russia) was used as a nematic host. The nematic phase embraces a wide temperature range from $-20°$ to $60°C$. This material has a positive dielectric anisotropy; the refractive indices are 1.71 and 1.52 ($\lambda = 589$ nm) for the extraordinary and ordinary waves, respectively.

The concentrations of the dopants in the guest–host mixtures were 0.1 wt % (G5), 0.5 wt % (G1) and 0.5 wt % (D). We investigated the interaction of light with 100 μm thick homeotropic and planar cells. To produce the cells we used substrates coated with conducting layers of indium tin oxide. To obtain homeotropic
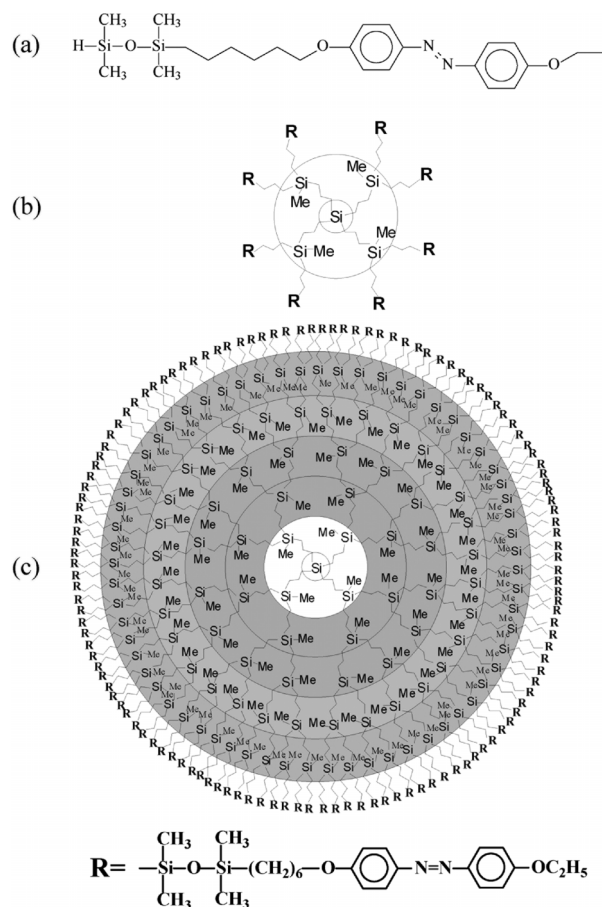
Figure 1. Chemical formulas of (a) the azobenzene dye D and the dendrimers (b) G1 and (c) G5 of the first and fifth generations.

and planar alignments, layers of chromium stearyl chloride and rubbed polyimide, respectively, were used.

The absorption of the dye and dendrimers is maximal in the UV region (at about 350 nm) and decreases with increasing wavelength (*13, 15*). The spectral dependences of the absorption coefficients in the range from 400 to 550 nm for the mixtures studied are presented in Figure 2. In this range, absorption is due to the dopants; at shorter wavelengths, the contribution of nematic host becomes dominant.

Solid-state ($\lambda = 473$ and 532 nm) and argon ($\lambda = 515$ nm) lasers were used as light sources. The linearly polarised light beam was focused by a lens ($f = 16$ cm) into an NLC cell, which could be rotated about the vertical axis, thus changing the angle of incidence, α. The latter is taken as positive if the cell is rotated anticlockwise and negative in the opposite case. The NLC director was in the horizontal plane. A double Fresnel rhomb was used to rotate the polarisation plane of the incident beam.

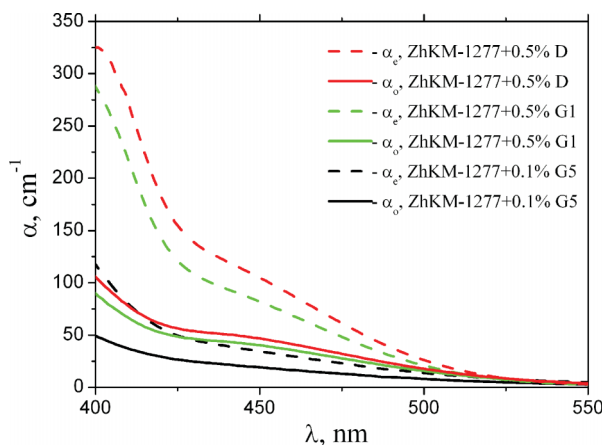The effect of light beam on liquid crystalline systems was studied using the phenomenon of self-phase

Figure 2. Spectral dependences of the absorption coefficients α_o and α_e of the liquid crystalline mixtures.

modulation. The latter results in the formation of a characteristic ring system in the cross-section of the beam passed through the NLC cell (*19–21*). From the number of rings, one can find the nonlinear phase shift and thus the change in the refractive index magnitude due to the light–NLC interaction. The transformation of the aberration pattern upon transverse displacement of the cell makes it possible to determine the sign of the refractive index variation (*22*).

3.   Results and discussion

Illumination of NLC cells doped with the dendrimers and azo dye resulted in formation of an aberration pattern. The pattern was of orientational origin, as evidenced by the dynamics of its development and relaxation: the time of development was $\tau_d \sim 20$–60 s (depending on the beam power and the incidence angle) and the relaxation time, $\tau_r \sim 15$ s. Typical aberration patterns are shown in Figures 3(a) and 3(c).

The determination of the self-action sign showed that the self-defocusing of the light beam (negative nonlinearity) occurred for the homeotropic and planar (Figure 3(b)) samples with G5 dendrimer, the homeotropic sample with G1 dendrimer and the homeotropic sample with dye D. The corresponding decrease in the refractive index of the extraordinary wave is obviously due to the director rotation normally to the light field **E**. On the other hand, self-focusing was observed for the planar samples with G1 (Figure 3(d)) and D (positive nonlinearity, the refractive index increases due to the director rotation parallel to **E**).

The manifestation of the self-defocusing clearly indicates the relationship between the orientational phenomena and the absorbing dopants.

The effect of changing the sign of orientational nonlinearity (observed for G1 and D dopants) has
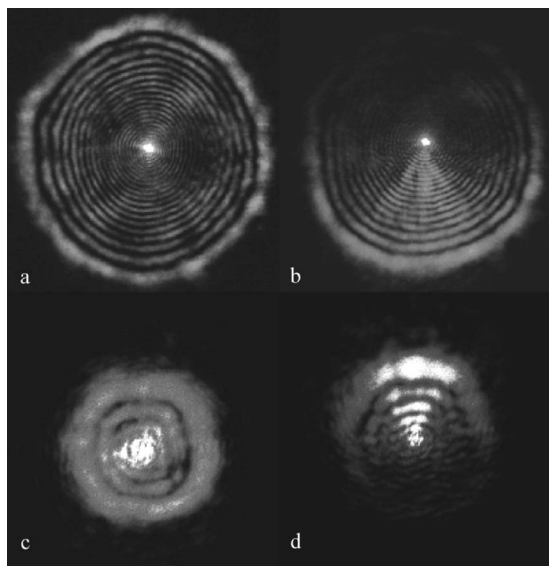
Figure 3. Typical shapes of the aberration patterns in the cross-section of the light beam ($\lambda = 473$ nm, $\alpha = 50°$) passed through planar samples of (a, b) ZhKM-1277+0.1% G5 ($P = 2.1$ mW) and (c, d) ZhKM-1277+0.5% G1 ($P = 4.3$ mW). (a, c) The steady-state patterns and (b, d) the transformations of the patterns upon fast shift of the NLC cell upwards (the determination of the self-action sign). Lightening of the (b) lower and (d) upper parts of the patterns indicates the director rotation normal or parallel to the light field, respectively.

been observed previously for a number of low-molecular azo dyes (*22–24*). According to Janossy and Szabados (*24*), the reason for this phenomenon is the presence of two (*trans-* and *cis-*) isomers of the azo compounds. The *trans*-isomers induce negative nonlinearity in the NLC, whereas for the *cis*-isomers it is positive. Before illumination, most dopant molecules are in the *trans* state. Excitation of *trans* molecules gives rise, with some probability, to conversion into the *cis* state. A similar process also takes place for the *cis*-isomers. As a result, a certain equilibrium ratio of the isomer concentrations is established. It is important that this ratio does depend on the angle between by the light field **E** and the NLC director **n**. The reason for such a dependence is the difference in the order parameters of the isomers, and with it the probabilities of a molecule excitation for a given light polarisation. Angular dependence of the isomer concentrations may result in the appearance of a critical angle β_c between wavevector **k** of the light wave and NLC director **n** such that at $\beta < \beta_c$ director rotates perpendicular to the light field **E** and at $\beta > \beta_c$, parallel to the light field **E**. It should be noted that the photoalignment of azobenzene mesogens under polarised light, the process responsible for the light-induced birefringence and dichroism in LC polymers (*25*), should not play an important role in our experiment because the

azobenzene moiety concentration is very low (no more than 0.5 wt %) and the viscosity of low-molar-mass nematics is much lower than that of the LC polymers.

The dependences of the aberration-ring number N on the light-beam power at oblique incidence on NLC cells are shown in Figure 4. The N value increases and saturates with light beam power P. As can be seen from Figure 4(a), the value of the nonlinear response induced by 0.5% G1 is about two times higher than that induced by 0.1% G5 and about three times higher than the corresponding value for 0.5% of 'free' dye molecules.

Figure 5 shows the dependences $N(P)$ for different angles of light incidence on planar NLC doped with 0.1% G5. The threshold light beam power ($P_{th} = 2.1$ mW) is clearly manifested at normal incidence. It should be noted that no self-action was developed at normal incidence on homeotropic and planar NLCs with G1 and D because the director **n** in these cases is already aligned in the directions to which the light field **E** rotates it.
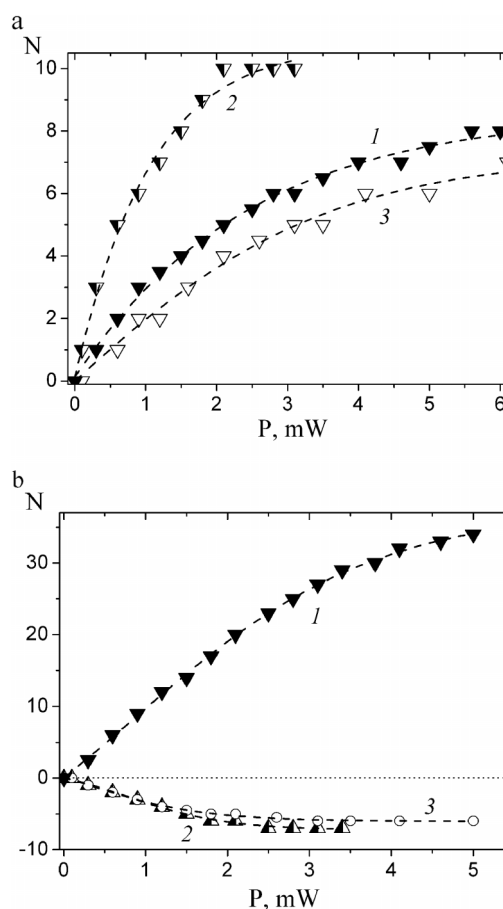

Figure 4. Dependences of the aberration-ring number N on the light beam power P ($\lambda = 473$ nm, $\alpha = 50°$) for (a) homeotropic and (b) planar samples of ZhKM-1277 doped with (1) 0.1% G5, (2) 0.5% G1 and (3) 0.5% D. Positive sign of N corresponds of self-defocusing, whereas the negative one indicates self-focusing.
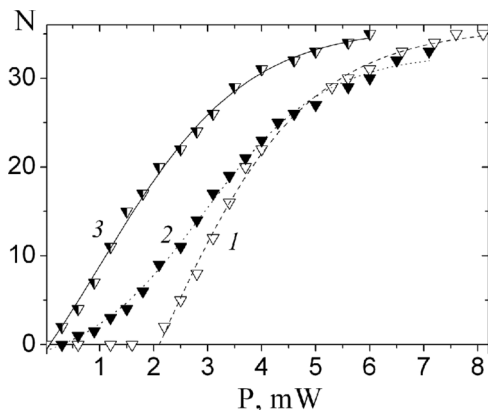
Figure 5. Dependences of the aberration-ring number N on the beam power P (λ = 473 nm) for a planar sample of ZhKM-1277 + 0.1% G5 at different angles of light incidence α = (1) 0°, (2) 20° and (3) 40°.
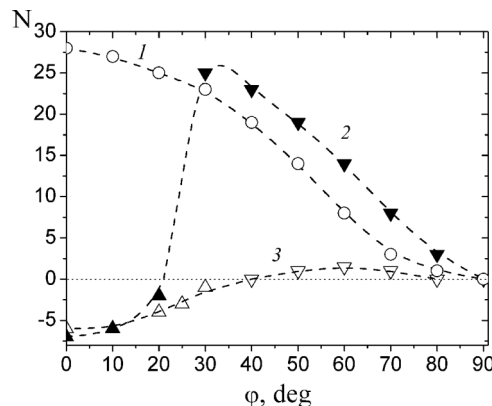

Figure 6. Dependences of the aberration-ring number N of the self-defocusing (circles and inverted triangles) and self-focusing (triangles) on the angle φ of the polarisation plane rotation (λ = 473 nm, P = 3.1 mW, α = 50°) for planar cells of ZhKM-1277 doped with (1) 0.1% G5, (2) 0.5% G1 and (3) 0.5% D. Changing the sign of N illustrates the transition from self-focusing to self-defocusing.

In the saturation region (Figures 4 and 5) the ring number N approaches the maximum value N_{sat}, corresponding to the complete director reorientation normally or parallel to the light field. For instance, N_{sat} = 35 at normal incidence on planar sample ZhKM-1277+0.1% G5 (curve 1 in Figure 5). The maximum possible ring number estimated for Δn = 0.19 is N_{max} = 40.

It should be emphasised that the characteristic features of the light-induced director reorientation (threshold at normal incidence and saturation) are the same as in the case of optical Freedericksz transition in transparent homeotropic NLCs (*19, 26*).

Figure 6 shows the dependences of the ring number N on the polarisation plane rotation angle φ (0° corresponds to *p*-polarisation; 90° to *s*-polarisation) for the light beam transmitted through NLC cells. It can be seen that for the planar sample ZhKM-1277+0.1% G5 (Figure 6, curve 1) the transition from the horizontal (extraordinary) polarisation (φ = 0°) to the vertical (ordinary) one (φ = 90°) results in the collapse of the aberration pattern (N decreases from 28 to 0). The polarisation dependences for planar samples ZhKM-1277 doped with 0.5% G1 (Figure 6, curve 2) and 0.5% D (Figure 6, curve 3) are quite different; polarisation rotation gives rise a change in the self-action sign (self-focusing is replaced by self-defocusing). This effect is also a consequence of the photoinduced isomerisation.

From Figure 6 we estimated the critical angle β_c with the aid of the relation (*22, 27*)

$$\varphi_c = \arccos\left(\frac{\sin\beta_c}{\sin\beta}\right), \qquad (1)$$

where β is the angle of light refraction and φ_c is the angle at which the polarisation dependence changes its

sign. For NLC doped with G1 dendrimer β_c = 56° and for NLC doped with the dye D, β_c = 43°. These critical values β_c can be reached neither in planar nor in homeotropic samples because of the light refraction at the air–glass interface. It can be seen that the range of negative nonlinearity, $0 < \beta < \beta_c$, increases with complication of molecular structure, i.e. when passing from dye D to dendrimers G1 and G5 (for G5 the nonlinearity is always negative, independent of geometry).

The principal difference is also manifested in the action of low-frequency (ν = 3 kHz) electric field on planar samples doped with the dendrimers (Figures 7(a)–7(b)) and azo dye (Figure 7(c)); in the case of NLC doped with G5 the nonlinear optical response is always negative, whereas for G1- and D-doped NLCs the $N(U)$ dependence can change its sign. Moreover, the $N(U)$ dependences are non-monotonic; their shapes depend on the sign of angle α, i.e. the sense of the NLC cell rotation about the vertical axis. Similar dependences were previously observed and analysed for the sign-inversion nonlinearity induced by azo dye DEANAB (*28*).

For ZhKM-1277+0.1% G5 (Figure 7(a)), the dependences at positive (curve 2) and negative (curve 1) α have a little difference. For both cases, at first, low-frequency voltage results in increasing aberrational pattern due to larger torque on the beam axis with respect to the torque on beam periphery. Indeed, the torque induced by the electric field is governed by the expression $\Gamma_{elect} = \frac{\Delta\varepsilon}{4\pi}(\mathbf{nV})[\mathbf{n} \times \mathbf{V}]$, i.e. $\mathbf{\Gamma}_{elect} \sim \sin 2\psi$, where ψ is the angle between director $\mathbf{n}$ and electric
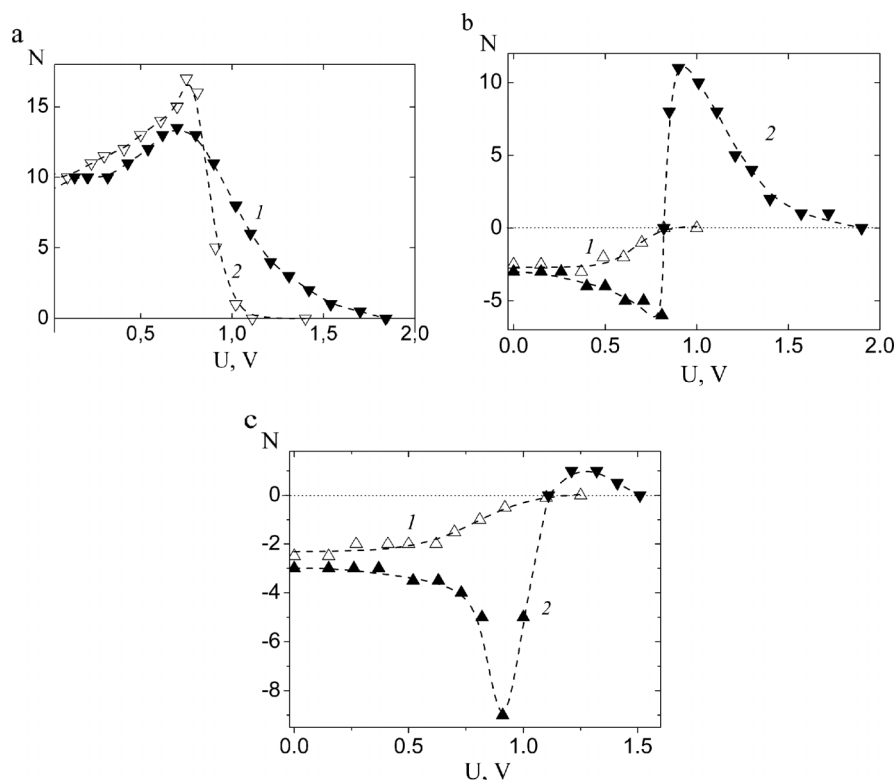
Figure 7. Dependences of the aberration-ring number N of the self-defocusing (inverted triangles) and self-focusing (triangles) on low-frequency ($\nu = 3$ kHz) electric voltage for planar samples of ZhKM-1277 doped with (a) 0.1% G5, (b) 0.5% G1, and (c) 0.5% D for $\alpha = -40°$ (1) and $\alpha = +40°$ (2). The change in the sign of N illustrates the transition from self-focusing to self-defocusing. The positive and negative signs of the angle α correspond to the counterclockwise and clockwise rotation of the NLC cell about the vertical axis.

field **V**; on the beam axis ψ is larger than ψ on the beam periphery due to orienting action of the light field. Further increase in voltage suppresses aberrational rings owing to smoothing of the director orientation (the director in the middle plane of the cell becomes normal to the substrates throughout the entire sample). Differences in pattern behaviour at positive and negative angles of light incidence are due to the pretilt. Figure 8 explains this phenomenon. At positive α (Figure 8(a)), director at the beam periphery $\mathbf{n}_1$ rotates due to low-frequency voltage along with director on the beam axis $\mathbf{n}$ which rotates due to light action. At negative α (Figure 8(b)), director at the beam periphery $\mathbf{n}_1$ rotates in opposite sense with respect to director on the beam axis $\mathbf{n}$ which results in larger N.

Application of the low-frequency field to planar samples doped with G1 and D (Figures 7(b) and 7(c)) lead at positive α to changing the self-action sign (curves 2). At first, the number of aberrational rings of self-focusing grows (director on the beam periphery rotates in opposite sense with respect to director on the beam axis). Then due to the simultaneous action of external voltage and elastic forces director jumps to the orientation shown on Figure 8(a), the

angle β between $\mathbf{n}$ and $\mathbf{k}$ becomes less than the β_c, and self-defocusing reveals. At negative α (curves 1), no variation of the self-action sign (self-focusing) was observed. In this case, director on the beam axis rotates along the director at the beam periphery. This reorientation can result in increasing N at the beginning, but later low-frequency field suppresses the aberrational pattern due to smoothing of director field.

In homeotropic NLCs, an ac field decreases N (suppresses the director field deformation) independent of the sample composition.

We also studied the influence of a dc electric field on the light beam self-action in NLCs doped with dendrimers G1 and G5. The self-action in this case was observed even for the ordinary (vertical) polarisation (to be more precise, a small deviation from the vertical polarisation was required to visualise the pattern). The phenomena manifested were similar for both dendrimer generations. The aberration pattern had the ring-shaped structure (Figure 9(a)) with a minor asymmetry, which was easy to observe at shifting the cell in transversal horizontal direction. Shifting the cell left (with respect to the beam) makes the right part of
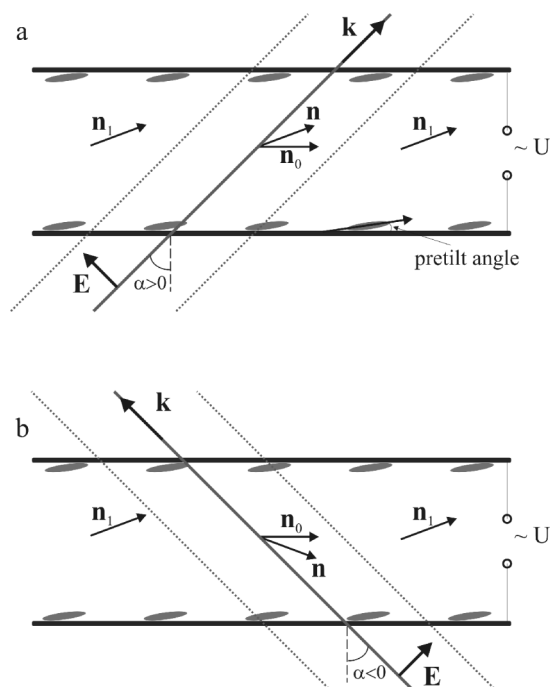
Figure 8. The geometry of light interaction with NLC in the presence of the external low-frequency field for (a) positive and (b) negative α: $\mathbf{n}_0$ is the unperturbed director, $\mathbf{n}$ is director on the beam axis, $\mathbf{n}_1$ is the director at the beam periphery. The beam refraction at the NLC boundaries is not shown.

the pattern brighter (Figure 9(b)), while shifting the cell right highlights an internal asymmetric structure (Figure 9(c)). Such behaviour is similar to the self-action in NLCs due to surface photorefractivity (light-induced elimination of the external dc screening by ions present in nematic mixtures) (*29*). The degree of the pattern asymmetry in our case is much smaller than that for undoped host. Detailed analysis and simulation of the asymmetric patterns for NLCs in a dc field will be presented elsewhere.

We estimated the enhancement factor η of the nonlinear optical response due to the dopants as compared to the pure nematic host. To this end we determined the ratio of the light-beam powers at which an equal number of rings was observed for pure and doped cells at the same experimental conditions. In order to obtain larger N ($\sim$ 10–15) for pure samples we carried out the measurements with short-focal-length lens ($f = 8$ cm). For the negative dye-induced nonlinearity the comparison was made for the cells of 'opposite' alignment (e.g. the homeotropic pure host cell and the planar doped cell).

Comparison of dependences $N(P)$ measured at $\alpha = 50°$ and $\lambda = 473$ nm for the homeotropic and planar samples ZhKM-1277+0.1% G5 with the corresponding dependences for undoped NLCs yielded $\eta = -55$ and -65, respectively. These values coincide within the limits of experimental error ($\sim$15%) of our method. For the homeotropic and planar samples doped with G1 and D we found $\eta = -70$ and 36 (G1) and $\eta = -11$ and 19 (D), respectively.

The quantity η is obviously proportional to the dopant concentration and is thus inconvenient for comparison of the nonlinearities of different liquid crystalline systems. We shall use for this purpose the factor $\eta_\alpha = \eta/(\alpha_\parallel + 2\alpha_\perp)$, proportional to the ratio of η to the absorptivity $\alpha_{av} = (\alpha_\parallel + 2\alpha_\perp)/3$ averaged over the director orientation.

For homeotropic and planar samples we obtained $\eta_\alpha = -1.08$ and -1.28 cm, respectively, for doping with G5, $\eta_\alpha = -0.64$ and 0.33 cm for G1 doping and $\eta_\alpha = -0.08$ and 0.14 cm for D doping.

The nonlinearity parameter $\eta_\alpha = -0.94$ cm (G5, planar alignment) is smaller than the parameter $\eta_\alpha = -2.3$ for nematic host doped with comb-like polymer (*10*) by approximately a factor of two. Nevertheless, this value is higher than the values reported in literature for liquid crystalline systems doped with
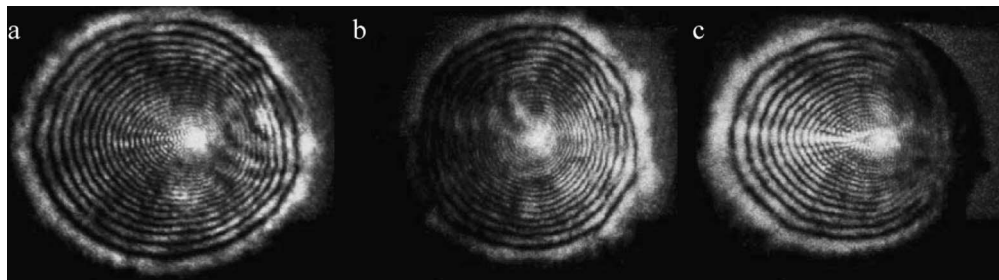

Figure 9. Typical shapes of the aberration patterns in the cross-section of the light beam ($\lambda = 515$ nm, $\alpha = +40°$) passed through planar samples of ZhKM-1277+0.1% G5 ($P = 5$ mW) under a dc field (+2 V): (a) the steady state pattern and (b, c) the transformation of the pattern upon fast shift of the NLC cell leftward or rightward, respectively. The polarisation of the light beam was close to the polarisation (vertical) of the ordinary wave.

low-molar-mass compounds (to the best of our knowledge, the highest negative and positive nonlinearities are $\eta_\alpha = -0.05$ cm, calculated from the data of Janossy and Kosa (*30*), and $\eta_\alpha = 0.8$ cm, calculated from the results of Kosa *et al.* (*31*)).

4. Conclusion

We have studied in detail the light-induced director reorientation in NLCs doped with dendrimers of the fifth (G5) and first (G1) generations containing light-absorbing azobenzene fragments or with an azo dye (D) identical to the fragments.

It was found that the orientational optical nonlinearity induced by the dendrimer G5 is always negative, whereas the sign of those induced by G1 and D depends on the interaction geometry of the director and light field (light beam propagation direction and polarisation) and external ac voltage. All the specific features of the light-induced Freedericksz transition were observed in the planar NLC doped with G5.

The results suggest that composite liquid crystalline systems containing macromolecules are promising for increasing the efficiency of the optical orientation.

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