Optica Applicata, Vol. XXIV, No. 4, 1994

Nonlinear liquid-crystalline optical cells

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The paper discusses the nonlinear optical phenomena observed in the nematic liquid crystals in relation to the conditions of their investigation. The experimental configurations of the liquid crystal cells are analysed and the basic results of examination of the nonlinear properties of liquid crystals are presented. Some qualitative remarks for the experiments are also given.

1. Introduction

Liquid crystals (lc's) are very attractive materials for studying the nonlinear optical phenomena at least for two reasons:

– first, because they exhibit very large nonlinearity, and

– second, for the interaction between light and lc structure is relatively slow.

These opportunities facilitate the observation of the nonlinear processes and their dynamics (although the latter is rather disadvantageous in practical applications). Huge nonlinear response of lc's to the laser excitation arises from their unusual physical properties, leading to collective phenomena in liquid crystalline media. Liquid crystals are composed of highly anisotropic molecules showing long-range, relatively weak interactions. This results in molecular correlation over macroscopic distances and therefore in an ordered structure of the substance. Since the molecular forces in lc's are markedly weaker compared to the solid crystals, the structure of lc's is easily influenced by various external agents, *i.e.*, electric and magnetic fields, mechanical stresses and temperature.

Among nearly 30,000 lc substances presently known, there are several distinct phases, classified in two main types: nematic (including chiral modification) and smectics. Nematics, the molecules of which are orientationally ordered with a preferred long-axis direction, represented by the unit vector $\boldsymbol{n}$, while positionally not correlated, attract at present most attention because of simpler structure and the ease for theoretical and experimental examination.

2. Nonlinear optical properties of liquid crystals

The liquid crystalline substances besides the usual nonlinearity of intramolecular origin (electronic) show the pronounced orientation and thermal nonlinearity contributions. While the electronic contribution is not exceptionally different from that of ordinary liquids, the orientational mechanism – which appears as a specific feature of materials having collective molecular motion – as well as thermal processes bring the lc's in the class of the strongest nonlinear materials (exceeding the standard CS_2 by several orders of magnitude).

The theoretical analysis of optical reorientation effects in lc's is based on the description of the lc electrooptic phenomena in static fields, which are well recognized [1], [2]. External quasistatic electric field acting on lc induces electric torque and consequently evokes molecular response which manifest themselves in elastic and viscous forces. By fixed director orientation by the surfaces, called "strong anchoring", there arises structural deformation in lc layer. This is accomplished by space variation of the refraction index. Depending on the initial molecular alignment $\boldsymbol{n}_0$ with respect to the $\boldsymbol{E}$ field direction, the deformation appears as a threshold effect (if $\boldsymbol{n}_0$ and $\boldsymbol{E}$ are parallel or perpendicular) or thresholdless, if $\boldsymbol{n}_0$, $\boldsymbol{E}$ are inclined. In theoretical description of the optical nonlinear effects the action of the static field is replaced by the $\boldsymbol{E}$-vector of the light wave [3]. However, the static-field and the optical deformations are not exactly analogous.

3. Optical reorientation of lc structure

It is quite clear that also the optical $\boldsymbol{E}$-field of the travelling light can influence the lc molecules tending to align them along the $\boldsymbol{E}$-vector ($\varepsilon_{\mathrm{opt}\parallel} > \varepsilon_{\mathrm{opt}\perp}$). In opposite, the changes brought about by light in the lc structure affect again the light wave changing its direction and polarization. The mutual interaction of the medium and light gives rise to diffraction and interference effects. Basically the following two distinct types of optical deformations of lc structure can be distinguished:

– the adiabatic or continuous one with the characteristic length L_c greater than the light wavelength λ, $L_c \gg \lambda$, and

– the non-adiabatic deformations if it is comparable with the wavelength, $L_c \approx \lambda$.

The former, optical adiabatic deformations are the analogs of the static-field reorientation of lc layer, for in the usual cases the thickness d of lc cells markedly exceeds the light wavelength λ, while the latter, nonadiabatic ones are purely optical phenomena and have no analogy in the static electrooptical reorientations. They can be produced, for instance, by interference of two beams incident at different angles, or also by two components – ordinary and extraordinary – of the same beam.

4. Basic nonlinear optical experiments

4.1. Experimental conditions

For the sake of interpretation simplicity, most of the theoretical as well as experimental examinations of nonlinear effects in lc's are carried out in the nematic cells with homogeneous structure of the following three types [4]–[7]:

– nematic homeotropic, *i.e.*, if $\boldsymbol{n}_0$ is perpendicular to the lc limiting surfaces (Fig. 1),

– nematic planar, if n_0 is parallel to the limiting surfaces (Fig. 2),

– chiral nematic Grandjean-planar, if the helix axis of the structure is perpendicular to the limiting surfaces of the lc layer (Fig. 3).

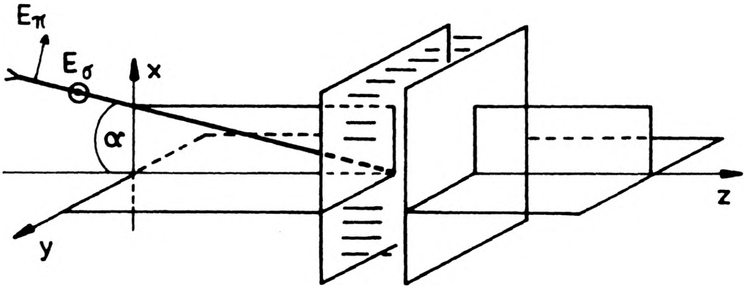

Fig. 1. Configurations of the experimental liquid crystal cells with homeotropic-planar geometry

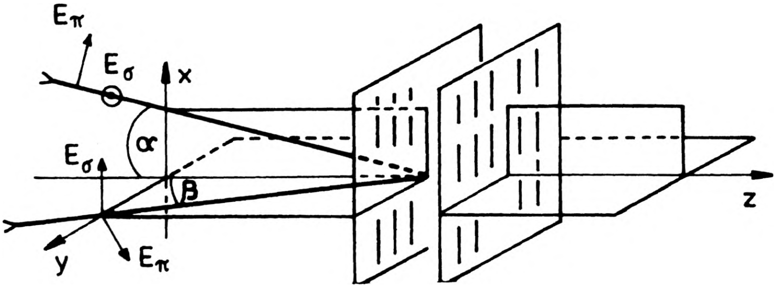

Fig. 2. Nematic-planar configurations of the liquid crystal cells

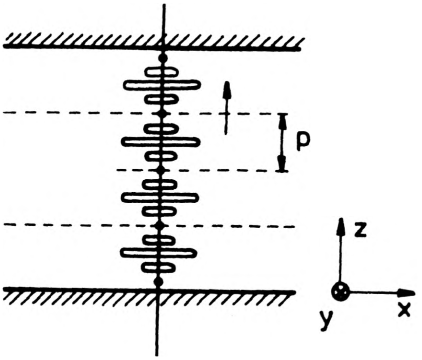

Fig. 3. Chiral nematic-planar-Grandjean geometry of the lc cells

In general, both adiabatic and non-adiabatic deformations take place simultaneously in the lc layer. Their separation is, however, possible in some cases. In practice, only the adiabatic deformation is experimentally observed by usually accessible laser light intensities.

4.2. Theoretical predictions [3], [8], [9]

4.2.1. Homeotropic texture, strong anchoring (Fig. 1)

The two deformations occur; along normal to the lc layer (xz-plane) the adiabatic deformation is possible, while parallel to it (xy-plane) – only the non-adiabatic one.

If the laser beam propagates at *normal incidence* ($\alpha = 0$), both beam polarizations σ and π are equivalent; since in that case $\boldsymbol{E}_{\mathrm{opt}} \perp \boldsymbol{n}_0$, then the reorientation is a threshold type with any minimal threshold value met.

For the beam impinging at an *inclination angle* ($\alpha \neq 0$), the reorientation is thresholdless only for π polarization because $\boldsymbol{E}$ and $\boldsymbol{n}_0$ are neither parallel nor perpendicular; but for σ polarization the effect is a threshold type, where

$$I_{\mathrm{th}}(\alpha) \geqslant I_{\mathrm{th}}(0);$$

moreover the value of $I_{\mathrm{th}}(\alpha)$ increases stepwise at discrete angle values α_m, for small angles: $\alpha_m \sim \lambda/d\ (2m-1/2)^{1/4}$, m is an integer.

The latter case is a pure optical effect having no analogy to any static-field distortions.

4.2.2. Planar cells, strong anchoring (Fig. 2)

For *normal incidence* ($\alpha = \beta = 0$), only a non-adiabatic deformation is possible, namely if the light beam propagates through the cell as an ordinary wave (y-axis polarization). The effect is a threshold-type with I_{th} dependent on the cell thickness d. For all non-adiabatic deformations rise of d increases I_{th}, just in contrast to the adiabatic distortions. In this case (y-polarized beam) the eventual adiabatic rotation of the lc molecules in the xy-plane cannot occur, due to Maguin's theorem [1]. Physically, the optical $\boldsymbol{E}$-vector follows the continuous rotation of the director $\boldsymbol{n}$ (if such appears, *e.g.*, initiated by thermal fluctuations), the both being always exactly orthogonal, so the light cannot produce any torque to cause structural distortions.

Obviously, for extraordinary wave propagating at normal incidence nothing happens ($\varepsilon_{\mathrm{opt}\parallel} > \varepsilon_{\mathrm{opt}\perp}$ – the structure is in the minimum energy state).

If the laser beam is thrown *at a certain angle* α *in xz-plane* ($\alpha \neq 0$, $\beta = 0$), both adiabatic and non-adiabatic deformations take place:

– for σ-polarized beams the adiabatic deformation appears at any light intensity, since the effect is thresholdless,

– for π-component non-adiabatic deformation can occur, provided that a threshold is reached, which increases again stepwise with the angle α.

If the beam, however, is *inclined in yz-plane* ($\alpha = 0$, $\beta \neq 0$), only non-adiabatic distortions are possible for π-polarization above some threshold rising in steps with the angle β. Clearly, for σ-polarized beam in that case nothing can happen.

4.2.3. Chiral nematic cells (cholesterics)

The chiral nematic lc's possess unique optical properties due to their specific periodic structure. The optical specificity arises from the fact that the period p is of the order of visible wavelength λ. It results in Bragg reflection in the visible region, large optical activity and the selective reflection of light. The parameters of the effects depend upon the structure periodicity and the cell geometry [2].

In detail only the simplest geometry was analyzed theoretically and experimentally for the cells with planar-Grandjean texture (Fig. 3) of the pitch $p \approx \lambda$, and by normal incidence. Theoretical description of the chiral nematics is complex. Since the light can deform the structural periodicity due to orientational and thermal mechanisms, changes in all optical effects observed here are to be expected.

5. Experimental results

The fundamental results obtained experimentally [3], [5] due to giant (orientational) and/or thermal nonlinearity for the both: nematics (n) and chiral nematics (cn) are shortly listed below.

1. Self-focusing and self-diffraction, both are caused by spatial variations of refracting index, produced by the transverse intensity profile [6] of the laser beam (n).
2. Bistable switching in the Fabry—Perot lc cells (n).
3. Generation of additional orders of the Bragg reflection (cn).
4. Variations of the optical activity (however, a high optical power is needed: 10%-change requires the beam intensity of 10^6 W/cm^2), (cn).
5. Changes of the light elipticity in the selective reflection region (cn),
6. Changes of the light elipticity dispersion with light frequency in the selective reflection region (cn).

6. Summary

1. The liquid crystals exhibit a huge specific nonlinearity — the orientational optical nonlinearity (giant nonlinearity).
2. The interest is mainly focused on the nematic liquid crystals (*the strongest nonlinearity and the simplest structure*), some on cholesterics, and a little smectics.
3. The nonlinear optical effects start already at the light intensity of about 100 W/cm^2 which corresponds to $E_{\mathrm{opt}} \approx 120$ V/cm, while the threshold for analogous static distortions is of the order of $E_{\mathrm{stat}} \approx 500$ V/cm.
4. Further lowering of the optical threshold is possible by application of a static assistant field up to 0.1 W/cm^2).
5. Generally, both adiabatic and nonadiabatic deformations are present. They differ in the efficiency and the inducting light intensities. The adiabatic deformation is easy to observe, while the nonadiabatic ones are weakly detected experimentally (*needed threshold* ≈ 1 GW/cm^2).
6. The optical orientational nonlinearity (OON) is accompanied by thermal nonlinearity (TON). Both have comparable efficiencies but differ in dynamics: OON is slower — *up to several seconds*, TON — *under microseconds.*

7. The orientational nonlinearity responds also to the nanosecond laser pulses superposing partial deformations [9], provided that the repetition frequency is higher than the lc relaxation process, *i.e.*, $f_{\mathrm{pulse}} > 1/\tau_{\mathrm{def}}$, but requires more power. The pulse laser OON is markedly smaller than the cw laser OON.

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