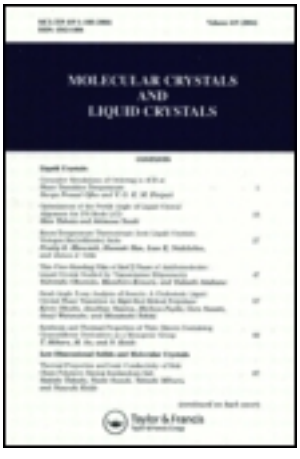

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

Orientation Nonlinearity of Absorbing Liquid Crystals

Anatoliy A. Kovalev [a], Vladimir N. Sadovsky [a] & Natalia A. Usova [a]

[a] Institute of Electronics, BSSR Academy of Sciences, Minsk, USSR

Version of record first published: 22 Sep 2006.

To cite this article: Anatoliy A. Kovalev , Vladimir N. Sadovsky & Natalia A. Usova (1990): Orientation Nonlinearity of Absorbing Liquid Crystals, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 191:1, 357-361

To link to this article: http://dx.doi.org/10.1080/00268949008038618

Mol. Cryst. Liq. Cryst. 1990, Vol. 191, pp. 357–361
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

ORIENTATION NONLINEARITY OF ABSORBING LIQUID CRYSTALS

ANATOLIY A. KOVALEV, VLADIMIR N. SADOVSKY, NATALIA A. USOVA
Institute of Electronics, BSSR Academy of Sciences, Minsk, USSR

Abstract Some mechanisms of the cubic nonlinearity of LCs are discussed. They are due to director rotation by the temperature gradient or the concentration gradient of the photoisomer forming as a result of radiation absorption.

INTRODUCTION

Common thermal optical nonlinearity of absorbing liquid crystals (LCs) is connected with a strong temperature dependence of the refractive indices $n_{\parallel}$ and $n_{\perp}$ for fixed optical axis . Orientation nonlinearities which have been studied intensively recently occur through rotation of the director $\mathbf{n}$ by the field of light waves [1].

This work reports some mechanisms of the cubic nonlinearity in the absorbing LCs based on idea considered below [2].

Let a LC be acted upon by a nonuniform light field. Absorption of light may result in modification of a certain scalar parameter ψ of the LC (below, we shall consider modulation of temperature T or the concentration N of the photoisomer). $\nabla\psi$ is the thermodynamic force conjugate to the flux J_{ψ} (thermal flux J_T or diffusion flux of the photoisomer particles J_N). In centrosymmetrical LCs this force contributes to the dissipative part of the angular momentum flux tensor (gyrothermal effect [3]) and to the dissipative part of the momentum flux tensor if there is no symmetry centre (Lehmann effect). The divergence of the angular momentum flux and the antisymmetrical part of the momentum flux can be considered as a force acting upon the LC director and resulting in

local rotation of the optical axis. The same effect occurs owing to the dependence of the LC elastic moduli on the parameter ψ.

Let us consider the interaction of two plane waves

$$\mathbf{E}=\mathbf{e}_1 \cdot a_1 \cdot \exp(i(\omega_1 \cdot t - \mathbf{k}_1 \mathbf{r})) + \mathbf{e}_2 \cdot a_2 \cdot \exp(i(\omega_2 \cdot t - \mathbf{k}_2 \mathbf{r})) + c.c \quad (1)$$

Temperature distribution is found from the heat equation and takes the form :

$$\delta T = Q_T \cdot a_1 \cdot a_2^* \cdot \exp(i(\Omega t - \mathbf{q}\mathbf{r})) + c.c.,$$

$$Q_T = \frac{(\mathbf{e}_1 \cdot \hat{\beta}_T \cdot \mathbf{e}_2)}{i \cdot \rho \cdot C_P \cdot \Omega + æ_{ij} \cdot q_i \cdot q_j}, \quad (2)$$

where $\hat{\beta}_T$ is the tensor of the absorption coefficients, æ is the heat conduction tensor, $\Omega = \omega_1 - \omega_2$, $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$.

When considering photoisomerization mechanisms of nonlinearity we shall bear in mind trans-cis isomerization in the molecules having azo-group as a possible process

$$R\diagup N{=}N\diagup R \quad <------> \quad R\diagdown N{=}N\diagup R .$$

Cis-isomer has a V-like form and has no mesophases. Therefore, formation of a relatively small amount of the cis-isomer under the action of radiation brings about significant changes of the liquid cristal properties [4]. Concentration of the isomer will be found from the diffusion equation which is solved as follows :

$$\delta N = Q_N \cdot a_1 \cdot a_2^* \cdot \exp(i \cdot (\Omega t - \mathbf{q}\mathbf{r})) + c.c.,$$

$$Q_N = \frac{(\mathbf{e}_1 \cdot \hat{\beta}_N \cdot \mathbf{e}_2)}{i \cdot \Omega + D_{ij} \cdot q_i \cdot q_j + \tau_0^{-1}}, \quad (3)$$

here $\hat{\beta}_N$ is the tensor describing photogeneration of impurity, D_{ij} is the diffusion tensor, τ_0 is the photoisomer lifetime. In the present paper we shall not consider selfaction effects, hence, only interference term will remain in the expressions for δN and δT.

CHOLESTERIC LIQUID CRYSTALS

Let us consider the mixture of cholesteric LCs at the compensaton point. The force acting upon the director owing to Lehmann effect and owing to the temperature dependence of the helix pitch h takes the form:

$$\mathbf{f} = -G_T \cdot [\mathbf{n}\,\nabla]\,\delta T; \quad G_T = G_L + \pi \cdot k_{22} \cdot \partial h^{-1}/\partial T, \tag{4}$$

The correction to the dielectric permittivity is found from the equation for the director

$$\gamma \frac{\partial n_i}{\partial t} + (\delta_{ij} - n_i \cdot n_j)\left(\frac{\partial F}{\partial n_j} - \frac{\partial}{\partial x_k}\frac{\partial F}{\partial n_{j,k}} + f_i\right) = 0, \tag{5}$$

where γ is the orientation viscosity, F is the free energy, and is written as:

$$\delta\epsilon_{ij} = \epsilon_a (n_i \cdot \delta n_j + n_j \cdot \delta n_i) = \Gamma_{ij} \cdot a_1 \cdot a_2^* \cdot \exp(i(\Omega t - \mathbf{qr})) + \text{c.c.} \tag{6}$$

$$\Gamma_{ij} = i \cdot \epsilon_a \cdot G_T \cdot Q_T \frac{n_i [\mathbf{nq}]_j + n_j [\mathbf{nq}]_j}{i \cdot \gamma \cdot \Omega + (k_{33} - k_{22})(\mathbf{nq})^2 + k_{22} \cdot q^2}. \tag{7}$$

Photoisomerization analog of the equations (4-7) is obtained by substituting N for T . In (6) we deliberately omitted the well-known contribution connected with the dependence $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ on N and T. In experiment it can be made small by choosing suitable geometry. The following equations

$$\frac{\partial a_1}{\partial z} = i \cdot \mu \cdot |a_2|^2 \cdot a_1 - \frac{\beta_1}{2} \cdot a_1, \qquad \frac{\partial a_2}{\partial z} = i \cdot \mu^* \cdot |a_1|^2 \cdot a_2 - \frac{\beta_2}{2} \cdot a_2, \tag{8}$$

where $\mu = \frac{(\mathbf{e}_1 \cdot \Gamma \cdot \mathbf{e}_2)}{2 \cdot c \cdot n}$, describe the energy exchange between the waves 1 and 2 if Im $\mu \neq 0$. The threshold power of the pump wave, i.e. power necessary to compensate the linear absorption β_a, depends only on the absorption anisotropy rather than on its absolute value and is estimated as :

$$P_s \approx \frac{k_{22}}{G_T} \cdot æ \cdot \frac{\epsilon}{\epsilon_a} \cdot k^2 \cdot \alpha^3 \approx 1 \text{ W/cm}^2$$

for $æ = 0.6 \cdot 10^{-4}$ W/cm°C, $\epsilon_a/\epsilon = 0.1$, $G_T/k_{22} = 4 \cdot 10^3$ (°C·cm)$^{-1}$, $\alpha = 0.1 \approx 6°$ - an angle between $\mathbf{k}_1$ and $\mathbf{k}_2$, $k \approx 10^5 \text{cm}^{-1}$. For the photoisomerization analog, G_N/D can be significantly larger than $G_T/æ$.

NEMATIC LCs

It is known that in nematic LCs the Lehmann effect is forbidden and $h^{-1} = 0$ because of centrosymmetricity. However, the gyrothermal effect is allowed, which gives

$$f_i = L \cdot (T_{,i} - n_i \cdot n_k \cdot T_{,k})_{,j} \cdot n_j. \tag{9}$$

The force (9) is proportional to the second-order derivative of temperature, hence, to obtain the energy exchange it is necessary to use cells with inhomogeneous initial orientation of the director. Also, the dependence of k_{aa} on T should be allowed for. Another possibility consists in using a cell with walls with substantially different heat conductions and such interaction geometry for which one of the derivatives is taken over the coordinate ξ normal to the cell plane:

$$f = i \cdot L \cdot q \cdot \frac{\partial T}{\partial \xi} \cdot \exp(i \cdot \mathbf{qr}) + \text{c.c.}. \tag{10}$$

In this case estimation of the threshold power of the pump wave gives 1÷10 W/cm² for the grating pitch of 10÷50 μ.

C SMECTICS

The free energy of the C-smectic LCs is [5]

$$F = F_c + F_1 + F_{c1} + F_x, \tag{11}$$

where F_c, F_1 and F_{c1} are the energies of the distortion of C-director field, deformation of layers, and their cross-contribution, respectively,

$$F_x = D_1 \cdot \frac{\partial \Omega_z}{\partial x} + D_2 \cdot \frac{\partial^2 U}{\partial x \partial y} + D_3 \cdot \frac{\partial \Omega_z}{\partial z} \tag{12}$$

is the contribution connected with the noncentrosymmetricity. Further, we shall assume $D_a = \partial D_a / \partial \psi \; \delta\psi = \tilde{D}_a \cdot \exp(i(\Omega t - \mathbf{qr})) + \text{c.c.}$, $\delta\psi = \delta T$ or δN and is defined by the expressions (2,3). In other words, we shall consider the compensated mixture of the chiral smectics. Variation of the free energy (11) with the dissipative function

$$R = \frac{1}{2} \cdot \left[\gamma_1 \cdot \left(\frac{\partial \Omega_z}{\partial t} \right)^2 + \gamma_2 \cdot \left(\frac{\partial^2 U}{\partial y \partial t} \right)^2 + \gamma_3 \cdot \left(\frac{\partial^2 U}{\partial x \partial t} \right)^2 \right] \tag{13}$$

results in the following expressions for $\tilde{\Omega}_z$ and $\tilde{U}$ ($\{\Omega_z, U\} = \{\tilde{\Omega}_z, \tilde{U}\} \cdot \exp(i(\Omega t - \mathbf{qr})) + \text{c.c.}$):

$$\Omega_z = [i \cdot (q_x \cdot D_1 + q_z \cdot D_3) \cdot a_{22} - q_x \cdot q_y \cdot D_2 \cdot a_{12}] / \Delta,$$
$$U = [q_x \cdot q_y \cdot D_2 \cdot a_{11} - i \cdot (q_x \cdot D_1 + q_z \cdot D_3) \cdot a_{21}] / \Delta, \tag{14}$$

where $a_{11} = i \cdot \Omega \cdot \gamma_1 + B_1 \cdot q_x^2 + B_2 \cdot q_y^2 + B_3 \cdot q_z^2 + B_{13} \cdot q_x \cdot q_z$,

$a_{22} = i \cdot \Omega \cdot (\gamma_2 \cdot q_y^2 + \gamma_3 \cdot q_z^2) + A \cdot q_z^2 \cdot q_y^2 + A_{12}^2 \cdot q_x^4 + A_{21}^2 \cdot q_y^4$,

$a_{12} = -a_{21} = i\cdot(C_1\cdot q_x^2 + C_2\cdot q_y^2)q_y$; $\Delta = a_{11}\cdot a_{22} - a_{12}\cdot a_{21}$.

γ_a are the coefficients of viscosity, A,B,C are the elastic moduli. The correction $\delta\epsilon_{ij}$ is determined by rotation of the director $\delta\mathbf{n} = \delta\mathbf{c}\cdot\sin\theta + \delta\mathbf{e}\cdot\cos\theta$, where $\delta\mathbf{e}$ is the deviation of the normal to the layer from the z-axis, $\delta\mathbf{c} \approx \Omega_z$ is the deviation of c-director, θ is the tilt angle of molecules in the layer. The relations (14) show that the grating of the layers' displacement is in-phase while the grating of the c-director is shifted by $\pi/2$ relative to the intensity grating for $\Omega=0$. However, in the one-axis approximation the expression for the correction $\delta\epsilon_{ij}$ does not contain inphase gratings since $\delta\mathbf{n}$ contains U as a spatial derivative. It should be expected that the nonlinear susceptibility of the smectic LCs will be of the same order as in cholesteric LCs because the elastic moduli of these two classes of LCs are approximately equal. It should be noted that smectic LCs are characterized by a more intricate dependence on geometric factors.

Thus, the noncentrosymmetrical LCs allow an effective energy exchange between waves of milliwatt power for the frequency degenerate interaction. Nonlocal nonlinear susceptibility of the centrosymmetrical LCs turns out to be 2 or 3 orders of magnitude weaker. The mechanisms considered are characterized by a strong dependence of the nonlinear susceptibility on the wave vector of recorded gratings and are analogous, in some respects, to the nonlocal nonlinearity of photorefractive crystals.

REFERENCES

1. B. Ya. Zeldovich, N. V. Tabiryan, Uspekhi Fiz. Nauk, 147, 633 (1985).
2. A. A. Kovalev, V. N. Sadovsky, N. A. Usova, Kvant. Elektron, 14, 997, (1987).
3. V. B. Nemtsov , Physica, 86A, 513 (1977).
4. S. G. Odulov, Yu. A. Reznikov, O. G. Sarbei, et al., Ukr. Fiz. Zhurn., 25, 1922 (1980).
5. P. G. de Gennes, The physics of liquid crystals (Clarendon Press, Oxford, 1974).