



## Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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## PHOTOINDUCED CHANGE OF CHOLESTERIC LC-PITCH

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**Abstract** The effect of reversible shift of selective reflectivity band in induced cholesteric LC under the laser irradiation is observed.

Conformational molecular transition caused by light absorption is a reason of a type of the LC conformation optical nonlinearity (CNL)<sup>1</sup>. In nematics CNL shows itself in the dependence of refractive index on the intensity of light. This dependence is conditioned by changes of the polarizability of own molecules  $\gamma$  and the order parameter  $S$  near them under phototransformation<sup>2</sup>.

In cholesterics change of the molecular conformation leads not only to another value of  $\gamma$  but to changes of twisting power  $\beta$  and so to photostimulated variations of cholesteric pitch  $P$ . This in turn results in the shift of cholesteric selective reflectivity band maximum,  $\lambda_{\max}$ <sup>3</sup>.

Purely this type of nonlinearity could be observed in systems where cholesteric pitch temperature dependence is negligible and laser heating does not affect the effect

Photostimulated shift of selective reflectivity band caused by reversible phototransformation of twisting dopant molecules was detected in induced cholesteric -

teric with the weak dependence of  $P$  on temperature consisted of cyanobiphenyl matrix LC-807 and optically active dopant (OAD) 1-arimiden-mentanon. Phototransformation of OAD was induced at room temperature by HeCd laser beam, its emission ( $\lambda = 0.44 \mu\text{m}$ ) falls at the edge of dopant absorption band and matrix transparency region.

According to <sup>4</sup> under light absorption alike OAD molecules biphenyl fragment turns about C=C bond. Under this condition twisting power of the OAD molecule decreases which causes the increasing of the pitch step. It should be noted that the origin of the OAD photoisomerization is not yet established. For instance, it is not yet clear why despite high activation energy needed to turn the fragment around C=C bond ( $\approx 170 \text{ kJ} \cdot \text{mol}^{-1}$ ) the isomerization process is reversible.

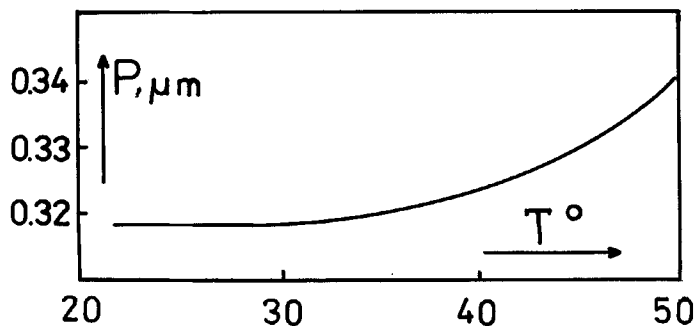


FIGURE 1

Under He-Cd laser intensities  $I \approx 0.3 \text{ W/cm}^2$  medium heating evaluated using the expression  $T \approx \alpha \rho^2 I / \pi \kappa^5$  ( $\alpha \approx 10 \text{ cm}^{-1}$  is system absorption coefficient,  $\kappa \approx 2.5 \cdot 10^{-3} \text{ W/cm} \cdot \text{K}$  is thermoconductivity coefficient,  $\rho \approx 0.05 \text{ cm}$  is the beam radius) is about 0.1 K. This could not be sufficient to change  $P$ , which value practically is independent of temperature at room temperature ra-

nge (fig.1).

Changing of  $P$  was observed as variation of trans - mission and reflectivity of circularly polarized He-Ne laser testing beam directed perpendicular to the cell. As He-Ne laser wave length  $\lambda = 0.63 \mu\text{m}$  falls into the transparency region of the matrix and dopant molecules, radiating of the system does not change  $\beta$ ,  $T$  and  $P$  values.

Experiments were performed with planar sample, the cell thickness  $L$  was  $50 \mu\text{m}$ , steady-state cholesteric pitch  $P \approx 0.32 \mu\text{m}$  (at  $I=0$ ).

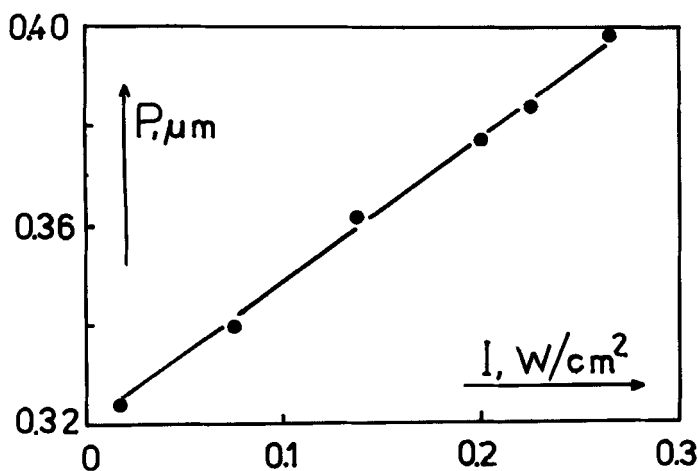


FIGURE 2

Fig.2 shows the dependence of steady-state value of  $P$  versus laser beam intensity  $I$ . It is linear within experimental accuracy, tangent of the tilt angle characterizes system sensitivity and is treated as the effective parameter of cholesteric CNL. Evaluation performed shows that its value is not less than the order of magnitude larger than the thermal non-linearity parameter for ordinary cholesteric without OAD, whose  $P(T)$ -dependence is rather strong.

The kinetics of the increasing of the cholesteric pitch  $P(t)$  is nonexponential and complicated (fig. 3): the effective process starts after a while and this time lowers linearly with intensity increasing. After switching off the excitation relaxation of cholesteric pitch behaves similarly.

In order to interpret the results obtained one needs to solve a problem of director distribution at the cell with planarly orienting surfaces taking into account that steady-state value of  $P_0$  in infinite sample depends on the light intensity  $I$ .

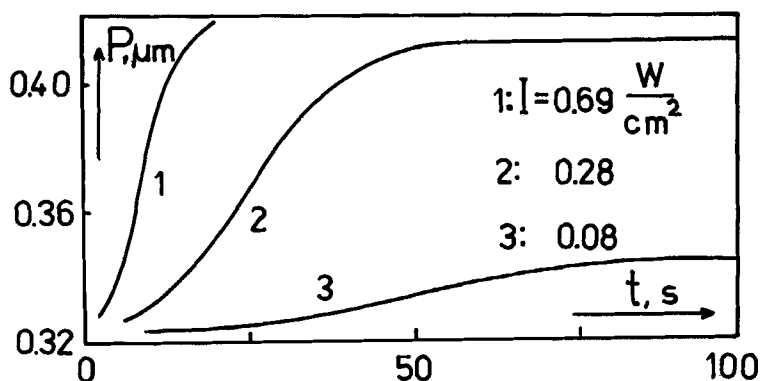


FIGURE 3

At anchoring parameter  $\xi = WL/K \gg 1$  ( $K$  is Frank elastic constant,  $W$  is an anchoring energy of cholesteric with orienting surfaces) the solution gives the following expression for cholesteric pitch wave-vector:

$$Q = \frac{\pi m}{L}(1 + 1/\xi) - (Q_0 + \frac{dQ_0}{dN} \Delta N(I)). \quad (1)$$

As the change of cholesteric pitch  $\Delta P \ll P$ , so we can write following expression for  $P = 2\pi/Q$ :

$$P \approx \frac{2\pi}{\frac{\pi m}{L}(1+1/\xi) - Q_0} \left( 1 - \frac{dQ_0/dN}{\frac{\pi m}{L}(1+1/\xi)} \Delta N(I) \right). \quad (2)$$

In these expressions  $Q_0 = Q$  in infinite sample,  $m = [(1-2/\xi)LQ/\pi]$  is an integer part of the numerical value in brackets,  $N$  is a stereoisomer concentration proportional to intensity  $I$ . It is obvious that value of  $Q$  in the cell differs from that in infinite sample and changes linearly with intensity increasing which is consistent with experimental data. When wave-vector value reaches some critical magnitude at which  $Q_m = Q_{m+1}$ , it abruptly changes at  $\pi(1+1/\xi)/L$  and then is going on linear change versus intensity. The number of such steps is determined by light intensity and the width of a single zone  $Q_{m+1} - Q_m \approx \pi/L$ . At the limits of one zone parameter of nonlinearity is proportional to anchoring parameter  $\xi$ .

Kinetics of transition to a steady-state  $Q$ -wave depends on the initial value of  $Q(t=0)$  and the ratio  $t_i/t_n$ , where  $t_i$  is stereoisomers lifetime,  $t_n$  is a relaxation time of director field. If the light intensity is such that  $Q(t=0)$  and  $Q(t=\infty)$  belong to the same zone and  $t_i \ll t_n$ , the  $Q(t)$ -dependence is determined by  $t_n$ . At the intensities for which abrupt change of  $Q$  takes place, the decreasing of  $Q$  divides into two stages. At the first one, limited by time during which stereoisomers concentration reaches the value corresponding to  $Q_m$ ,  $Q(t)$  is also determined by  $t_n$ . Then  $Q_m$  jumps up to the value of  $Q + \pi/L$  and again  $Q(t)$  depends on the magnitude of  $t_n$ , until  $Q$  become equal  $Q_{m+1}$ . With intensity increasing the time of achieving the critical  $Q$ -values decreases.

Experimental results can be qualitatively described in frames of the suggested model taking into account that the change of  $P \approx 8 \cdot 10^{-2}$  m in our case is much more larger than a single zone width  $P_{m+1} - P_m \approx P_0^2 / 2L \approx 9 \cdot 10^{-4}$  m. As a result dependences  $P(I)$  and  $P(t)$  turn out to be monotonic.

In order to reveal experimentally "zone" structure of the  $P(I)$ -dependence it is convenient to study samples where the number of cholesteric helix steps is comparable to the cell thickness. At this case the zone width is about  $P_0/2$  and also comparable with variation limit of  $\Delta P$ .

In conclusion it should be pointed out that systems discussed are quite perspective for optical data processing, because they need no temperature stabilization and have high sensitivity.

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