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THE ORIENTATIONAL OPTICAL NONLINEARITY OF PLANARLY ALIGNED LONG-PITCH CHOLESTERICS

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ABSTRACT

Optical nonlinearity of self-focusing type is investigated both theoretically and experimentally, which is due to reorientation of the cholesteric director by the light field, the effective nonlinear susceptibility decreasing abruptly with the shortening of the pitch P.

Theoretical APPROXIMATION

Orientational optical nonlinearities due to liquid crystal director reorientation were recently intensely investigated by several authors [1-5]. These nonlinearities became the subject of interest both because of their giant values and of the ability of creating tunable space-inhomogenious director profiles by means of space-modulated light fields. Yet up to now all these researches were concerned with nematic mesophase.

In this letter the results are reported of experiment on laser-field-induced reorientation of long-pitch cholestiric director, leading to third-order optical nonlinearity of self-focusing type, and a simple theoretical model of the effect is presented.

As all our experiments dealt with long-pitch ($\Delta n P \gg \lambda$) cholesterics, Δn being refractive index anisotropy, we made our calculations within the Mauguin's limit. Namely, we assumed the field tobe transverse o- or e-wave, the polarization unit vector of those "rotating" in the plane normal to the wave vector K with spatial period P along Z axis (see fig.1)

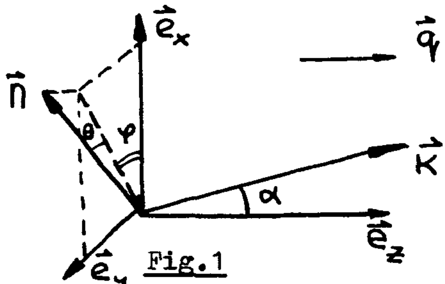

Fig.1

The explicit expression for e-wave unit vector is

$$\vec e_e = \frac{\vec e_x \cos^2\alpha \cos qz + \vec e_y \sin qz - \vec e_z \sin\alpha \cos\alpha \cos qz}{(1 - \sin^2\alpha \cos^2 qz)^{1/2}} \qquad (1)$$

The director unit vector equals $\vec n(z) = \vec e_x \cos qz + \vec e_y \sin qz + \delta\vec n$ $\delta\vec n$ having all the three space components. The linear with respect to $\delta\vec n$ term in the expression for wave electric field-cholesteric interaction free energy equals

$$F_{el} = -\frac{\varepsilon_a |E|^2}{8\pi} (\vec n^\circ \vec e_i)(\delta\vec n \cdot \vec e_i) \qquad (2)$$

$\vec n^\circ$ standing for undisturbed director, ε_a - dielectric constant anisotropy for light frequency, $\vec e_i$ - e- or o-wave unit vector. It readily can be seen that for o-wave expression (2) equals zero, i.e. the nonlinearity is absent. The same is true for the normal incidence of e-wave, according to $\delta\vec n \perp \vec n^\circ$.

In terms of $\delta\varphi$ and $\delta\theta$ angles, where $\delta\varphi = \varphi - qz$ $\delta\theta = \theta$ (see fig.1) (2) can be expressed as follows

$$F_{el} = \frac{B}{2} \sin^2\alpha \sin 2qz\, \delta\varphi + B \sin\alpha \cos\alpha \cos qz\, \delta\theta \qquad (3)$$

B being equal to $\frac{\varepsilon_a |E|^2}{8\pi}$. The expression for the elastic Frank's free energy, assuming uniformity of the director profile along X and Y axes (which is the formal case of infinite plane wave, corresponding in fact to the beam with the focal waist diameter exeeding the pitch, as we'll see later) is presented in [1])

$$F_\rho = \frac{1}{2}\left(\frac{d\theta}{dz}\right)^2\left[K_{11}\cos^2\theta + K_{33}\sin^2\theta\right] + \frac{1}{2}\left(\frac{d\varphi}{dz}\right)^2\cos^2\theta \times$$
$$\times\left[K_{22}\cos^2\theta + K_{33}\sin^2\theta\right] + \frac{1}{2}K_{22}q^2 + K_{22}q\frac{d\varphi}{dz}\cos^2\theta \qquad (4)$$

Leaving in (4) only the terms of order not higher than 2 one can readily obtain:

$$F_\rho = \frac{1}{2}K_{11}\left(\frac{d\delta\theta}{dz}\right)^2 + \frac{1}{2}K_{33}q^2(\delta\theta)^2 + \frac{1}{2}K_{22}\left(\frac{d\delta\varphi}{dz}\right)^2 \qquad (5)$$

To calculate real values for $\delta\varphi(z)$ and $\delta\theta(z)$ it is nesessary to minimize the total free energy by varying $\delta\varphi, \delta\theta$. Doing so we obtain following:

$$\begin{cases} K_{11}\dfrac{d^2\theta}{dz^2} - K_{33}q^2\theta - B\sin\alpha\cos\alpha\cos qz = 0 \\ K_{22}\dfrac{d^2\varphi}{dz^2} + \dfrac{B}{2}\sin^2\alpha\,\sin 2qz = 0 \end{cases} \qquad (6)$$
$$\theta(0) = \theta(L) = \varphi(0) = \varphi(L) = 0$$

Here and after we omit signs "δ" before θ and φ. The boundary conditions correspond to the tightly fixed alignment at the cell walls. It should be noted that both theoretically and experimentally we considered the case when the sample's thickness $L = \frac{n}{2}P$ i.e. cholesterics with nondisturbed helical pitch.

Evidently, equations (6) have strickt solution, the contribution from the general solution of homogenious equation for θ being negligible as it decreases by a factor of 10 at a distance of only $\sim\frac{1}{30}L$ from the boarders of the sample even when $L = P$. The nonlinear contribution into the electric induction vector $\vec{D}$ due to $\delta\vec{n}$ equals

$$\delta D_i = \delta\varepsilon_{ik}E_k = \varepsilon_a(n_i^0\delta n_k + n_k^0\delta n_i)E_k \qquad (7)$$

If we introduce the effective dielectric constant of the medium for e-wave as (8), then

$$\varepsilon_{eff} = \varepsilon_0 + \frac{\varepsilon_2}{2} |E|^2 \tag{8}$$

$$\varepsilon_2 = \frac{2\delta\varepsilon_{eff}}{|E|^2} = \frac{2\delta\vec{D}\cdot\vec{e}_e}{|E|^3} = \cos^2 qz \left[\frac{\varepsilon_a^2 \sin^4\alpha}{32\pi K_{22} q^2}\sin^2 qz + \frac{\varepsilon_a^2 \sin^2\alpha \cos^2\alpha}{8\pi(K_{11}+K_{22})q^2}\right] \tag{9}$$

As we are interested in self-focusing term in nonlinearity we are to omit oscillating terms in (9), in other words, to take the average value of (9) along Z. As a result we obtain the final expression for effective constant of orientational self-focusing nonlinearity of long-pitch ("Mauguin's") cholesteric:

$$\langle\varepsilon_2\rangle_{eff} = \frac{\varepsilon_a^2}{4\pi q^2}\left[\frac{\sin^2\alpha\cos^2\alpha}{(K_{11}+K_{33})} + \frac{\sin^4\alpha}{16K_{22}}\right] \tag{10}$$

Evidently, for real experimental conditions ($\alpha \lesssim 30^\circ$) the second term in (10) can be neglected within the accuracy of our aproximation. It is useful to to introduce the parameter ς ,which equals the ratio of nonlinear constants of cholesteric and planarly aligned nematic[1] of the same thickness and with the same values for ε_a, K_{ii} ,and α .

$$\varsigma = \frac{\pi^2}{32}\frac{K_{11}}{(K_{11}+K_{33})}\left(\frac{P}{L}\right)^2 \tag{11}$$

EXPERIMENTAL TECHNIQUE

The experimental arrangement was as follows (see fig.2): the radiation of argon-ion laser, operating in the lowest transverse mode (Gaussian one), of the total power of about 200 mW (controlled by the calorimeter "K") was focused by the lens "L_1" with focal length of 25 cm into the cell "C", which was Cano's wedge.

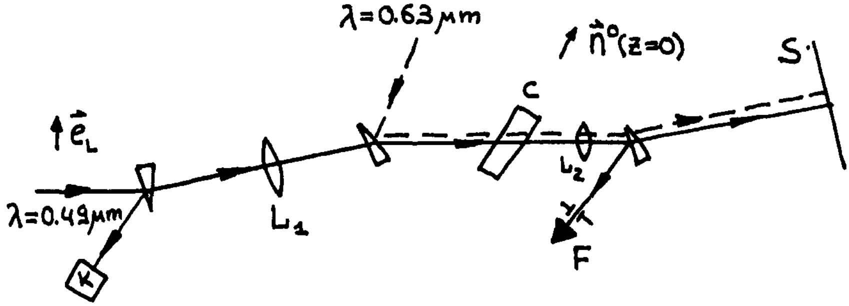

Fig.2

The transverse size of the lowest cross section of the beam (which was placed into the cell) was a = =HWe^{-2}M=100μm, which exceeded significantly the pitches of all cholesteric samples used. Simultaneously the sample was illuminated by a broadened beam of He-Ne laser, which enabled us to obtain by means of lens "L_2" the image of the cell at the screen "S". A part of the argon-laser beam was deflected after passing the sample onto the scanning photodetector "F" with small aperture, which was used to measure the angular divergence of the beam (in the absence of "L_2". The investigated cholesterics were weak solutions (0.1÷1 weight %) of cholesteryl-chloride in nematic 5CB. The concentration of the mixture was ajusted so that the pitch of the samples, measured by Cano - Grandjean method, $P=\frac{2L}{n}$,n being integer and L - standard length of 60μm.

Further, by means of ajusting the focal spot at equal distanses from two Grandjean walls we achieved the case of cholesteric texture of 60μm thickness with nondisturbed pitch.

In our experiment we controlled also the main refractive indices of the sample $n_{\parallel}$ and $n_{\perp}$ by a technique using deflection of the beam transmitted through the Cano's wedge and rotation of its polarisation.

It should be noted that within the accurasy of about 1% both indices were similar to those of pure 5CB. This fact is due to very low concentration of cholesteryl-chloride and enables us to obtain straightly the value of ξ (see(11)).

RESULTS AND DISCUSSIONS

The nonlinearity of self-focusing type was observed, that decreased rapidly with the sortening og the pitch. The sign of the effect (focusing or defocusing) was found out to be positive by a method similar to [2]. For o-wave, in accordance with theoretical consideration above, the effect was absent. The investigation was carried out of the dependences of ξ upon the cholesteric pitch and of the nonlinearity $\langle\varepsilon_2\rangle_{eff}$ upon refractive angle α . Those dependences are plotted in fig.3a),b) correspondently.

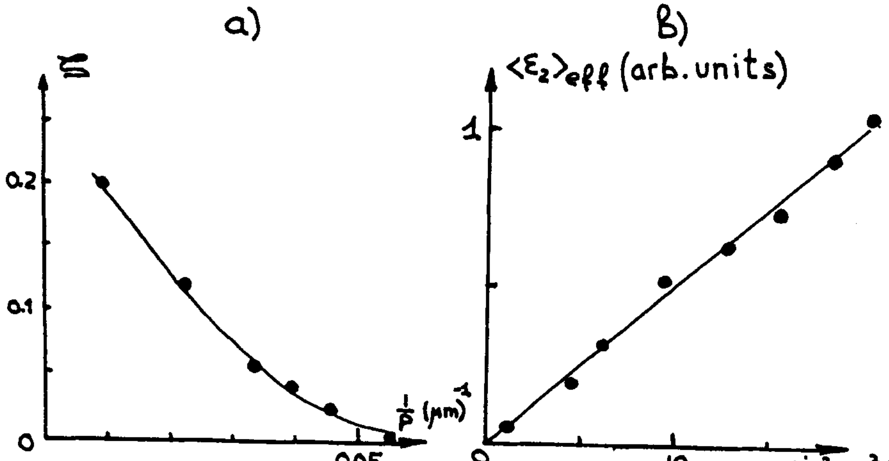

Fig.3

The measurements were carried out via comparison of $\langle\varepsilon_2\rangle_{eff}$ with ε_2 of the probe planarly aligned sample of pure 5CB, placed into the same cross section of the beam at the same incident angle. ξ was measured both from the number of the self-focusing rings (see[3-5]) and from the increase of the angular divergency of the transmitted beam by means of glass lenses, plased into the focal waist instead of the cell (see [2]).

It can readily be seen that the dependence of $\langle\varepsilon_2\rangle_{eff}$ of $\sin^2\alpha\cos^2\alpha$ is linear within the experimental accuracy. As to the dependence of S upon $\frac{1}{P}$, it perfectly well coinsides with the theoretical one if the elastic constants ratio in (11) is assumed to be 0.55.

As we've already sayd, the optical nonlinearity of cholesteric appears to be abruptly decreasing with the shortening of the pitch and even when the pitch equals the cell thickness $\langle\varepsilon_2\rangle_{eff}$ is only 20% of ε_2 of a nematic sample with the same L, ε_a and K_{ii}, i.e. $\langle\varepsilon_2\rangle_{eff} = 0.02\ \frac{cm^3}{erg}$.

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