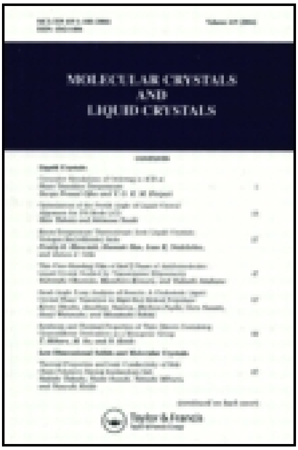

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The Orientational Optical Non - Linearity of Liquid Crystals

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The Orientational Optical Non-Linearity of Liquid Crystals

III. Smectics

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The cubic optical non-linearity of smectic liquid crystals (SLC) conditioned by director reorientation and layer structure deformation under the action of a light field is calculated. The strongest effects for smectic C phases are conditioned by the *c*-director turning without layer structure deformation. For smectic A phases, the director turning is accompanied by layer deformation, and because of the condition of layer incompressibility, the strongest response to the influence of the light field turns out to be non-local and its value depends on the cell thickness.

Estimations of optical non-linearity constants have been made; $\varepsilon_2 \sim 0.2$ cm^3/erg, for the light self-focusing and $\chi_{el} \sim 0.6 \cdot 10^{-6}$ cm^3/erg for the four-wave interaction; these are, respectively, 9 and 5 orders larger than the optical non-linearity for CS_2.

1 INTRODUCTION

Non-linear optical effects in liquid crystals attract much attention—see for example the survey by Arkelian *et al.*[1] The cubic optical non-linearity of oriented nematics (NLC) and cholesterics (CLC) has been calculated in detail in our papers.[2,3] This non-linearity has to be very large. It is determined by the local turning of the director under the action of the light field, i.e. by distortion of the initially homogeneously oriented mesophase.

The cubic optical non-linearity of smectic liquid crystals is calculated in the present paper. The existence of ordered planes brings about some remarkable peculiarities of the SLC response to the orientational influence of external fields.

The self-focusing effect in SLC is considered in detail. The properties of tensor χ (describing the four-wave interaction process in SLC) are investigated. The point is that the possibilities of wave front reversal (WFR)[4,5] by means of the dynamic hologram of four-wave interaction (WFR–FWI) have recently been intensively investigated. The plane reference wave $E_1 \exp\{i\mathbf{k}_1\mathbf{r}\}$

interfering with a signal wave $E_3(\mathbf{r})$ of complicated structure records a hologram in the medium, i.e., causes dielectric susceptibility perturbations $\delta\varepsilon(\mathbf{r}) \sim E_1 E_3^*(\mathbf{r}) \exp\{i\mathbf{k}_1\mathbf{r}\}$. If another plane wave $E_2 \exp\{i\mathbf{k}_2\mathbf{r}\}$ is simultaneously propagating towards the first one, i.e., $\mathbf{k}_2 = -\mathbf{k}_1$, then as a result of the hologram read-out, the wave $E_4(\mathbf{r}) \sim E_1 E_2 E_3^*(\mathbf{r})$ appears. Such a wave has the wave front conjugated relative to the signal wave front. Very promising applications of WFR for radiation self-targeting and for the correction of laser divergence make the WFR problem very attractive. In the present paper, it is shown that SLC non-linearity allows us to realize WFR–FWI at very low levels of the interacting wave powers.

2 THE BASIC EQUATIONS OF MOTION

Static deformations of SLC are characterised by parameters whose number depends on the SLC type. First of all, all SLC types have the parameter $u(\mathbf{r})$ in common, which characterises the displacement of the SLC layers from their equilibrium position. This displacement is counted off in the direction normal to the layers. The magnitude $(\mathbf{m}\nabla u)$ characterises the relative change of inter-layer distance. A non-zero value of $(\mathbf{m}\nabla u)$ contributes to the great elastic energy of a unit volume of medium.

$$F_e = \tfrac{1}{2}B(\mathbf{m}\nabla u)^2. \tag{1}$$

Therefore, a great response of a SLC to the light field will occur only in the case where the resulting distortions satisfy the incompressibility condition $(\mathbf{m}\nabla u) = 0$. It is worthwhile considering separately the cases of smectics C and A.

Smectics C The axes of C-type SLC molecules are oriented along some direction $\mathbf{n}$, making a constant angle ϕ with the normal $\mathbf{m}$ to the layers, so that $(\mathbf{nm}) = \cos\phi = \text{const}$ (see Figure 1). That is why the c-director notion is introduced as a unit vector in the layer planes characterizing the n-director projection on that plane; $\mathbf{c} = [\mathbf{n} - \mathbf{m}(\mathbf{nm})]/\sin\phi$, $|\mathbf{c}| = 1$. That is the reason why for smectic C phases, as is known, (see Ref. 8, page 306), the configuration with strongly plane layers is the only possible one which is not accompanied by great elastic energy. Therefore, the smectic C response to the light field will be considered in terms of the approximation $\mathbf{m} = \mathbf{m}^\circ = \text{const}$, $u = 0$. In this approximation, the unit volume free energy depends on the inhomogeneity of the c-director orientation. If we introduce an angle α between the c-director and some fixed direction in the layer planes, then the

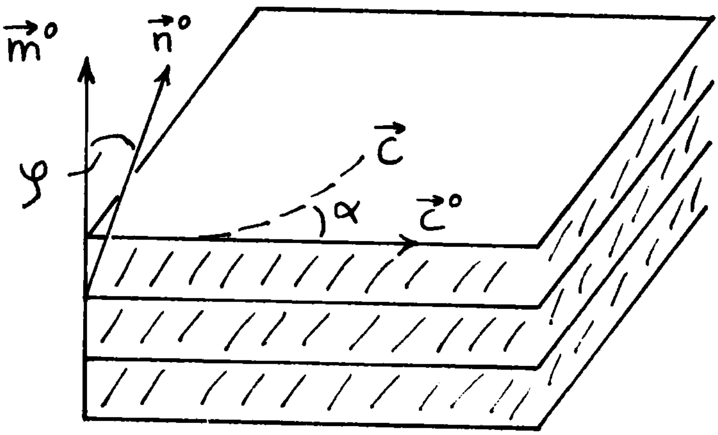

FIGURE 1 The plane, layered structure of a smectic C. The normal to the layers $\mathbf{m}^0$ is constant throughout the volume; the orientation of the molecular axes $\mathbf{n}$ makes an angle ϕ with the normal $\mathbf{m}^0$. The $\mathbf{n}$ vector projection on the layer planes characterises the c-director; the angle between the c-director and a particular undisturbed direction $\mathbf{c}$ is denoted by $\alpha(\mathbf{r})$.

free energy density F_c (erg cm^{-3}) can be written in the form

$$F_c = \tfrac{1}{2}B_1(\mathbf{c}\nabla\alpha)^2 + \tfrac{1}{2}B_2([\mathbf{m}^0 \times \mathbf{c}]\nabla\alpha)^2 + \tfrac{1}{2}B_3(\mathbf{m}^0\nabla\alpha)^2 + B_{13}(\mathbf{c}\nabla\alpha)(\mathbf{m}^0\nabla\alpha). \quad (2)$$

The constants B_1, B_2, B_3, B_{13} have the same dimension and the same order of magnitude as the Frank constants for nematics or cholesterics. The spontaneous light scattering in the C phase caused by the c-director fluctuations, has been experimentally investigated[12] where there are data about the smectic C constants.

Moreover, we assume that homogeneous magnetic field $\mathbf{H}$ is applied to the smectic C sample and the interaction energy will be written in the form $F_H = -(\kappa_a/2(\mathbf{nH})^2$, where κ_a is the magnetic susceptibility anisotropy, $\kappa_a = \kappa_{\parallel} - \kappa_{\perp}$. Since $\mathbf{n} = \mathbf{m}\cos\phi + \mathbf{c}\sin\phi$ and $\phi = \text{const}$, $\mathbf{m}^0 = \text{const}$, then omitting the constant addition, it is possible to take

$$F_H = -\tfrac{1}{2}\kappa(\mathbf{cH})^2 \quad (3)$$

where $\kappa = \kappa_a \sin^2\phi$.

By this we assume that the magnetic field is directed in the plane of the smectic layers. Such an assumption allows us to simplify the form of the equations without changing the physical meaning of the effects under consideration. In the case where $\mathbf{H} \sim \mathbf{n}$, we shall replace in all formulae below κH^2 by $\kappa_a \sin^2\phi(\mathbf{c}^0\mathbf{H})^2 + \kappa_a \sin\phi\cos\phi(\mathbf{mH})(\mathbf{c}^0\mathbf{H})$. The magnetic field $\mathbf{H}$ reinforces the homogeneous orientation of the smectic c in both cases, where $\mathbf{H} \sim \mathbf{n}$ and $\mathbf{H} \sim \mathbf{c}^0$; in the latter case the Fréedericks transition is a ghost transition.[10]

Finally, we shall describe the interaction of the light field

$$\mathbf{E}_{\text{real}}(\mathbf{r}, t) = 0.5[\mathbf{E}(\mathbf{r}, t)\exp\{-i\omega t\} + \mathbf{E}^*(\mathbf{r}, t)\exp\{i\omega t\}]$$

with the medium by the term $F_E(\varepsilon_a/16\pi)(\mathbf{nE})(\mathbf{nE}^*)$, which gives for the C phase

$$F_E = -\rho_1[(\mathbf{cE})(\mathbf{mE}^*) + (\mathbf{cE}^*)(\mathbf{mE})] - \rho_2(\mathbf{cE})(\mathbf{cE}^*) \tag{4}$$

where $\rho_1 = \varepsilon_a \sin\phi\cos\phi/16\pi$, $\rho_2 = \varepsilon_a \sin^2\phi/16\pi$. Thus we assume that with respect to optical properties, the c phase is a one-axis medium with the optic axis directed along the n-director.

The complex amplitude $\mathbf{E}(\mathbf{r}, t)$ for quasi-monochromatic fields is a slow function of t, and this allows us to discuss transient processes.

For describing establishment processes we also introduce the dissipation function R (erg cm^{-3} s^{-1}), such that

$$R = \tfrac{1}{2}\gamma\left(\frac{\partial\alpha(\mathbf{r}, t)}{\partial t}\right)^2 \tag{5}$$

where γ is a viscosity constant with the dimensions g cm^{-1} s^{-1}.

Here we are interested in small distortion of α only, and we are not interested in the influence of hydrodynamic motions on the relaxation time (compare with the analogous problem for nematics;[2] for the isotropic phase of cholesterics, the addition of hydrodynamic effects has been discussed).[9] The variational equations of motion have the form

$$\frac{\delta F}{\delta\alpha} - \frac{\partial}{\partial x_i}\frac{\delta F}{\delta(\partial\alpha/\partial x_i)} = -\frac{\delta R}{\delta\dot{\alpha}} \tag{6}$$

where as F we shall now insert the sum $F_c + F_H + F_E$ from Eqs. (2), (3) and (4). Let the initial homogeneous orientation of the c-vector be supported by the magnetic field so that $\mathbf{c}^0 \sim \mathbf{H}$. By doing this we shall be concerned with the first-order perturbations caused by the intensity of the light field, so that

$$\mathbf{c}(\mathbf{r}, t) = \mathbf{c}^0\cos\alpha(\mathbf{r}, t) + \mathbf{s}^0\sin\alpha(\mathbf{r}, t),$$
$$\mathbf{n}(\mathbf{r}, t) = \mathbf{n}^0 + \mathbf{s}^0\alpha(\mathbf{r}, t)\sin\phi,$$
$$\mathbf{s}^0 = [\mathbf{m}^0 \times \mathbf{c}^0]. \tag{7}$$

Therefore, we consider the angle α from the direction $\mathbf{c}^0$. As a result, we obtain from Eqs. (2)–(7)

$$(B_1 c_i c_k + B_2 s_i s_k + B_3 m_i m_k + 2B_{13} c_i m_k)\frac{\partial^2 \alpha}{\partial x_i \partial x_k} - \kappa H^2 \alpha - \gamma \frac{\partial \alpha}{\partial t}$$
$$= A_{ik} E_i E_k^*,$$

$$A_{ik} = s_i(\rho_1 m_k + \rho_2 c_k) + s_k(\rho_1 m_i + \rho_2 c_i)$$
$$= \sin\phi \frac{\varepsilon_a}{16\pi}(s_k n_i + s_i n_k). \qquad (8)$$

Here and later we omit the index "0" at $\mathbf{c}^0$, $\mathbf{m}^0$, $\mathbf{s}^0$.

Smectics A For the A type of SLC, the molecules are oriented perpendicular to the layers, and therefore, the directions **m** and **n** coincide. There is no degree of freedom corresponding to the *c*-director for a smectic A phase. The director vector $\mathbf{n}(\mathbf{r}) \equiv \mathbf{m}(\mathbf{r})$ is connected with the displacement parameter by the relation

$$\mathbf{n}(\mathbf{r}) = \mathbf{n}^0 - [\nabla u - \mathbf{n}^0(\mathbf{n}^0 \nabla u)]. \qquad (9)$$

We assume here that deviations from the undisturbed structure of flat layers are small, $|\mathbf{n}(\mathbf{r}) - \mathbf{n}^0| \ll 1$. In this approximation, the vector $\delta\mathbf{n}(\mathbf{r}) = \mathbf{n}(\mathbf{r}) - \mathbf{n}^0$ is perpendicular to $\mathbf{n}^0$. The smectic A interaction energy with the static magnetic field and with the light electric field has the same form as for a smectic *c*, if it is written using the vector $\mathbf{n}(\mathbf{r})$. Differences arise only when we take into consideration the concrete expression (9) for $\mathbf{n}(\mathbf{r})$. Taking into account the free energy of the deformed state of a smectic A we have

$$F = \tfrac{1}{2}B(\mathbf{n}\nabla u)^2 + \tfrac{1}{2}k_{11}[\nabla^2 u - (\mathbf{n}\nabla)(\mathbf{n}\nabla u)]^2$$
$$+ \tfrac{1}{2}\kappa_a H^2[(\nabla u)^2 - (\mathbf{n}\nabla u)^2]$$
$$+ \frac{\varepsilon_a}{16\pi}[\nabla u - \mathbf{n}(\mathbf{n}\nabla u)][(\mathbf{n}\mathbf{E})\mathbf{E}^* + (\mathbf{n}\mathbf{E}^*)\mathbf{E}]. \qquad (10)$$

Here and below we omit the index "0" at $\mathbf{n}^0$, assuming $\mathbf{n} \equiv \mathbf{n}^0 = \text{const}$. Besides, we assume that the external magnetic field **H** is directed along **n**. We write the dissipation function in the form

$$R = \tfrac{1}{2}\gamma\left(\frac{\partial n(\mathbf{r}, t)}{\partial t}\right)^2 \approx \tfrac{1}{2}\gamma\{\nabla \dot{u})^2 - (\mathbf{n}\nabla \dot{u})^2\}. \qquad (11)$$

The variational equations of motion

$$\frac{\delta F}{\delta u} - \frac{\partial}{\partial x_i}\frac{\delta F}{\delta(\partial u/\partial x_i)} + \frac{\partial^2}{\partial x_i \partial x_k}\frac{\delta F}{\delta(\partial^2 u/\partial x_i \partial x_k)} = -\frac{\delta R}{\delta \dot{u}} + \frac{\partial}{\partial x_i}\frac{\delta R}{\delta(\partial \dot{u}/\partial x_i)} \qquad (12)$$

for F and R derive from Eqs. (10) and (11) take the form

$$\frac{\partial}{\partial x_i}\left[Bn_i n_j \frac{\partial u}{\partial x_j} - K_{11}(\delta_{ij} - n_i n_j)\left(\frac{\partial^2}{\partial x_j \partial x_m} - n_l n_m \frac{\partial^2}{\partial x_j \partial x_l}\right)\frac{\partial u}{\partial x_m}\right.$$
$$\left. + \kappa_a H^2\left(\frac{\partial u}{\partial x_i} - n_i n_j \frac{\partial u}{\partial x_j}\right) + \gamma \frac{\partial \dot{u}}{\partial x_i} - \gamma n_i n_j \frac{\partial \dot{u}}{\partial x_j} + D_{ilm} E_l E_m^*\right] = 0, \quad (13)$$

$$D_{ilm} = \frac{\varepsilon_a}{16\pi}(\delta_{im} n_l + \delta_{il} n_m - 2 n_i n_l n_m).$$

Below we shall be interested in the case where the size of smectic A sample is sufficiently small such that the incompressibility condition $(\mathbf{n}\nabla u) = 0$ can be taken to be satisfied for the whole volume. In this way, the term $\sim B$ in Eq. (13) can be neglected.

3 SMECTICS-C; NON-LINEARITY

3a Light self-focusing

In considering the self-focusing problem, we assume that the bilinear combination $E_i^*(\mathbf{r})E_k(\mathbf{r})$ composed of the $\mathbf{E}(\mathbf{r}) = \mathbf{E}\exp\{i\mathbf{k}\mathbf{r}\}$ components of the light field is slowly dependent on the coordinate $\mathbf{r}$. It is possible in this case to neglect in Eq. (8) the terms $\sim B_1, B_2, B_3, B_{13}$, so that

$$\frac{\partial \alpha}{\partial t} + \frac{1}{\tau}[\alpha - \alpha_{stat}(\mathbf{r})] = 0, \; \tau = \frac{\gamma}{\kappa H^2},$$
$$\alpha_{stat}(\mathbf{r}) = \frac{\varepsilon_a \sin\phi |E(\mathbf{r})|^2}{8\pi\kappa H^2}(\mathbf{se})(\mathbf{ne}). \quad (14)$$

We have here assumed that the field $\mathbf{E}(\mathbf{r}) = \mathbf{e}E(\mathbf{r})\exp\{\mathbf{ikr}\}$ describes a certain kind of wave (0 or e) in the one-axis medium with the linear polarization unit vector $\mathbf{e}$.

The addition to the effective dielectric susceptibility for this wave can be written in the form

$$\delta\varepsilon_{ef} = \delta\varepsilon_{ik} e_i e_k = \varepsilon_a(n_i \delta n_k + n_k \delta n_i) e_i e_k$$
$$\equiv \varepsilon_2 |E(\mathbf{r})|^2 = \frac{\varepsilon_a^2}{4\pi\kappa_a H^2}(\mathbf{se})^2(\mathbf{ne})^2|E(\mathbf{r})|^2. \quad (15)$$

Let us note that the multipliers $\sin\phi$ from Eqs. (14) and (7) and the multiplier $\sin^2\phi$ from the relation $\kappa = \kappa_a \sin^2\phi$ were reduced. Therefore, $\kappa_a = \kappa_{\|}$

$-\kappa_\perp$ appears in Eq. (15), and the whole response from Eq. (15) turns out to be independent of the angle ϕ characterizing the C type of smectic.

Let us discuss now the effective non-linear constant ε_2 from Eq. (15) depending on the wave type and propagation direction $\mathbf{p} = \mathbf{k}/k$. The ordinary wave "0" in the one-axis medium has the polarization vector $\mathbf{e}_0$ normal to the optic axis $\mathbf{n}$, i.e. $(\mathbf{n}\mathbf{e}_0) = 0$. Therefore, from Eq. (15) it follows that for the 0-type wave, the self-focusing non-linearity is zero in the approximation under consideration, and non-zero effects exist only for the extraordinary wave. We would note that an analogous situation also arises for nematics.[2] Thus, for the appearance of strong self-focusing in a NLC it was necessary to direct the extraordinary wave at the sufficiently oblique angle β (i.e. $\beta \neq 0$ and $\beta \neq \pi/2$) to the director $\mathbf{n}$. This statement holds true for smectics C. The effective nonlinear constant ε_2, however, depends not only on $\cos\beta = (\mathbf{np})$, but also on $\mathbf{k} = \mathbf{p}k$, the vector orientation relative to the vector $\mathbf{s} = [\mathbf{m} \times \mathbf{c}] \sim [\mathbf{m} \times \mathbf{n}]$, where $\mathbf{m}$ is the normal to the layers. Really, for extraordinary wave $\mathbf{e}_e = [\mathbf{n} - \mathbf{p}(\mathbf{np})]/|\mathbf{n} - \mathbf{p}(\mathbf{np})|$, and from Eq. (15) we obtain

$$\begin{aligned}\varepsilon_2 &= \frac{\varepsilon_a^2}{4\pi\kappa_a H^2}(\mathbf{np})^2(\mathbf{sp})^2 \\ &= \frac{\varepsilon_a^2}{16\pi\kappa_a H^2}\sin^2 2\beta\cos^2\psi.\end{aligned} \tag{16}$$

A notation ψ for the azimuthal angle which the vector $\mathbf{s}$ makes with the projection of the $\mathbf{k}$ vector on the plane normal to $\mathbf{n}$ is such that $(\mathbf{sp}) = \cos\psi \sin\psi$ (see Figure 2).

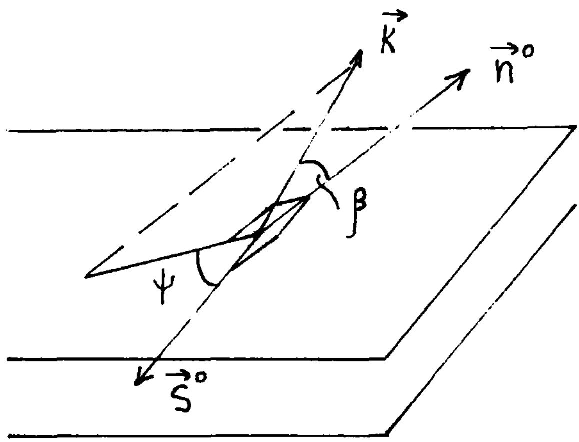

FIGURE 2 The notations of the angles β and ψ in the self-focusing problem for a smectic C.

The additional phase shift ϕ on the length L due to the non-linearity $\Delta\varepsilon = \varepsilon_2 |E|^2$ equals

$$\phi = \varepsilon_2 |E|^2 \frac{\omega}{2cn_e} L = \eta \frac{cn_e |E|^2 L}{8\pi}. \tag{17}$$

If the power density $P = cn_e |E|^2/8\pi$ is expressed in watts per cm^2, then the constant η has the dimension rad cm $watt^{-1}$.

Let us now make some numerical calculations. Let $\varepsilon_a = 1$ and $\kappa_a \sim 10^{-7}$. When the value of the magnetic field $H = 3 \cdot 10^3$ gauss, it can be expected that the medium will be oriented well enough. At $\psi = 0$, $\beta = \pi/4$, we have from Eqs. (16), (17) $\varepsilon_2 = 0.2\ cm^3/erg$, and for the wavelength $\lambda = 0.5\ \mu m$, in a vacuum, $\eta = 3.2$ rad cm $watt^{-1}$ (or $\eta = 3.2 \cdot 10^6$ rad cm $Mwatt^{-1}$). Taking $\gamma \sim 0.1$ poise, we obtain for the establishment time $\tau \sim 0.1$ s. For a cell thickness of the order of $L = 10^{-2}$ cm, a power density $P = 20\ watt/cm^2$, the phase poise ϕ comes to 0.6 rad. The critical power of self-focusing $P_c = \lambda^2 cn_e/32\pi^2\varepsilon_2 = \lambda/4\eta n_e = 2.10^{-6}$ watt. Thus, self-focusing in a smectic C may be easily carried out. However, it is necessary to choose the correct geometry for the experiment, so that $\sin^2 \beta \neq 0$, $\cos \psi \neq 0$. Besides, for too long times of observation, heating of the medium can distort or disguise the effects predicted here. Estimations analogous to those carried out in Ref. 2 show that during the non-linearity establishment time $\tau \sim 0.1$ s, and for the above mentioned value of the power, one can neglect thermal effects. Let us note that the estimation we have obtained for the optical non-linear constant turned out to be over 9 orders of magnitude larger than the non-linearity for CS_2.

3b Four-wave non-linearity

The importance of wave front reversal has been noted in the introduction. In this connection we shall consider here the four-wave non-linearity of a SLC. Let two plane waves fall on the smectic C sample

$$\mathbf{E}_{\text{real}}(\mathbf{r}, t) = \tfrac{1}{2}\{\mathbf{e}_1 E_1 \exp\{-i\omega_1 t + i\mathbf{k}_1\mathbf{r}\} + \mathbf{e}_3 E_3 \exp\{-i\omega_3 t + i\mathbf{k}_3\mathbf{r}\} + \text{c.c.}\}. \tag{18}$$

To obtain the smectic C response, it is necessary to insert the field in the form of Eq. (18) into Eq. (8). In this way we shall be interested in only that term in the response which is proportional to the product $E_1 E_3^*$. By calculating α and expressing $\delta\varepsilon$ through it, we obtain

$$\delta\varepsilon_{ik}(\mathbf{r}, t) = 4\pi\chi_{iklm}(\Omega, \mathbf{q})(\mathbf{e}_1 E_1)_l(\mathbf{e}_3 E_3)_m^* \exp\{-i\Omega t + i\mathbf{q}\mathbf{r}\},$$

$$\chi_{iklm}(\Omega, \mathbf{q}) = \frac{\varepsilon_a^2}{64\pi^2} \frac{\sin^2 \phi}{\Gamma} (n_i s_k + n_k s_i)(n_l s_m + n_m s_l),$$

$$\Gamma = -i\gamma\Omega + B_1(\mathbf{cq})^2 + B_2(\mathbf{sq})^2 + B_3(\mathbf{mq})^2 + 2\mathrm{B}_{13}(\mathbf{cq})(\mathbf{mq}) + \kappa H^2,$$

$$\Omega = \omega_1 - \omega_3, \mathbf{q} = \mathbf{k}_1 - \mathbf{k}_3. \tag{19}$$

Having it in mind to consider the case for WFR–FWI when the frequencies of all four waves coincide, we shall assume below $\Omega = 0$ in Eq. (19). We need the case $\Omega \neq 0$ to estimate the establishment time of the process. Scattering of the opposite reference wave $\mathbf{e}_2 E_2 \exp\{i\mathbf{k}_2\mathbf{r} - i\omega t\}$ with $\mathbf{k}_2 = -\mathbf{k}_1$ on the distortions (19) gives the additional term in the induction $\delta D_4 \sim E_1 E_2 E_3^*$ corresponding to the third wave WFR. It is also necessary to add to this term the contribution from the process whereby the waves E_2 and E_3^* record distortions of $\delta\varepsilon(\mathbf{r})$ in the medium, and the wave E_1 reads them out. As a result, we obtain for the E_4 wave amplitude an equation

$$\frac{dE_4}{dl} = i\frac{4\pi\omega^2}{k_3 c^2}\chi_{ef} E_1 E_2 E_3^*, \tag{20a}$$

$$\chi_{ef} = \tfrac{1}{2}\{\chi_{iklm}(0, \mathbf{k}_1 - \mathbf{k}_3) + \chi_{ilkm}(0, \mathbf{k}_2 - \mathbf{k}_3)\}e_{4i}^* e_{1k} e_{2l} e_{3m}^*.$$

Since the waves E_1 and E_2 are reciprocally conjugated, they belong to the same type, either ordinary or extraordinary. Just the same statement applies to the pair of waves E_3 and E_4. Because of this, we shall assume $\mathbf{e}_1 = \mathbf{e}_2 = \mathbf{e}_2^*$, $\mathbf{e}_3 = \mathbf{e}_4 = \mathbf{e}_4^*$, and then

$$\chi_{ef} = \frac{\varepsilon_a^2 \sin^2\phi}{128\pi^2}\{(\mathbf{n}\mathbf{e}_1)(\mathbf{s}\mathbf{e}_3) + (\mathbf{n}\mathbf{e}_3)(\mathbf{s}\mathbf{e}_1)\}^2 \cdot \{\Gamma^{-1}(0, \mathbf{k}_1 - \mathbf{k}_3) + \Gamma^{-1}(0, \mathbf{k}_2 - \mathbf{k}_3)\}. \tag{20b}$$

Let us consider the particular case in which the signal wave is of the ordinary type and propagates perpendicular to the axis $\mathbf{m}$ and the direction $\mathbf{s}$, i.e. $(\mathbf{m}\mathbf{k}_3) = (\mathbf{s}\mathbf{k}_3) = 0$, and then $(\mathbf{s}\mathbf{e}_3) = 1$. Let the reference wave propagate approximately along this direction, $\mathbf{k}_1/k_1 \approx \mathbf{k}_3/k_3$, and be of the extraordinary type. Then $\mathbf{k}_1 - \mathbf{k}_3 = \mathbf{v}_3(\omega/c)(n_e - n_0)$, $\mathbf{k}_2 - \mathbf{k}_3 = -\mathbf{v}_3(\omega/c) \cdot (n_e + n_0)$, and moreover $\mathbf{v}_3$ coincides here with the c-director $\mathbf{c}$. In a typical situation when $H \sim 10^4$ gauss, one can neglect the term $\sim H^2$ in Eq. (19) for $\Gamma(\mathbf{q})$. Then the main addition gives the term $\sim \Gamma^{-1}(0, \mathbf{k}_1 - \mathbf{k}_3)$, and from Eq. (20b) we obtain

$$R = \frac{|E_4|^2}{|E_3|^2} = |\tilde{\mu} E_1 E_2 L|^2, \tilde{\mu} = \frac{\lambda_{vac} n_0 \sin^2\phi}{16\pi^2 B_1}\left(\frac{n_e + n_0}{2n_0}\right). \tag{20c}$$

The Eq. (20c) has been obtained on the assumption that the reflection coefficient by the intensity R is less than one unit.

Equation (20c) can be rewritten in the form $R = \mu^2 L^2 P_1 P_2$, where P_1, P_2 are the power densities of the reference waves and we have introduced a new constant $\mu = \tilde{\mu} 8\pi / cn_e$. If at the same time we express the magnitudes P_1, P_2 in watts per cm^2, then the constant μ will have the dimension cm $watt^{-1}$. Taking for the refractive indices of the ordinary and extraordinary waves the values $n_0 = 1.5$, $n_e = 1.8$, and $\varepsilon_q = 1$, the wavelength in a vacuum as $\lambda = 0.5$ μm, the tilt angle of the molecules in the smectic C as $\phi \sim \pi/4$ and $B_1 \sim 10^{-6}$ dyn, we get $\mu \sim 0.3 \cdot 10^{-3}$ cm/watt. Corresponding to this, the value of the constant χ_{ef} from Eq. (20b) is of the order $0.6 \cdot 10^{-6}$ cm^3/erg, which is 5 orders greater than the non-linearity for CS_2. The power density $P_1 \sim P_2 \sim P$, which is necessary to obtain the reflection coefficient $R \sim 1$ at the interaction length $L \sim 0.1$ cm comes to $P \sim 3.10^4$ watt/cm^2. The establishment time estimated as $\tau \sim \gamma / B_1 q^2$, for $\gamma \sim 0.1$ poise turns out to be of $\tau \sim 0.7 \cdot 10^{-4}$ s.

4 SMECTICS A; NON-LINEARITY. LIGHT SELF-FOCUSING

4a Writing and solution of the equation for the director

We consider the self-focusing non-linearity for a smectic A in a particular example represented in Figure 3. We shall assume that the initial homogeneous state of the smectic A with plane layers is supported by the external magnetic field **H**, so that $\mathbf{n} \sim \mathbf{H}$. The n-director reorientation for the smectic A causes, without fail, layer deformation. A strong response on the light field occurs only in the case where this deformation does not bring about compression, i.e. if $(\mathbf{n}\nabla u(\mathbf{r})) = 0$. This means that layer displacement $u(\mathbf{r})$ and layer tilting $\delta\mathbf{n} = -\nabla u + \mathbf{n}^0(n^0\nabla u)$ must also differ from zero at the boundaries of the cell.[10]

The problem of boundary conditions for SLC has not been sufficiently studied.[11] We will assume therefore that the SLC is practically non-fixed at the cell walls, i.e. that there are no constraints put on $u(\mathbf{r})$ or on $\nabla \mathbf{u}(\mathbf{r})$.

Let us choose the coordinate axes x, y in the boundary plane, and z along the normal to the cell boundary. Let us introduce, besides this, the coordinates ξ, η, ζ with the relations

$$\xi = y, \eta = z\cos\beta + x\sin\beta, \zeta = -z\sin\beta + x\cos\beta \tag{21}$$

where β is the tilt angle of the molecular planes relative to the z-axis (see Figure 3). The axis ζ, through this, turns out to be directed along the $\mathbf{n}$, and the incompressibility equation takes the form $\partial u(\xi, \eta, \zeta)/\partial\zeta = 0$, wherefrom $u(\mathbf{r}) = u(\xi, \eta)$ is independent of the coordinate ζ.

We shall call the director line or N-line any line directed along the axis ζ.

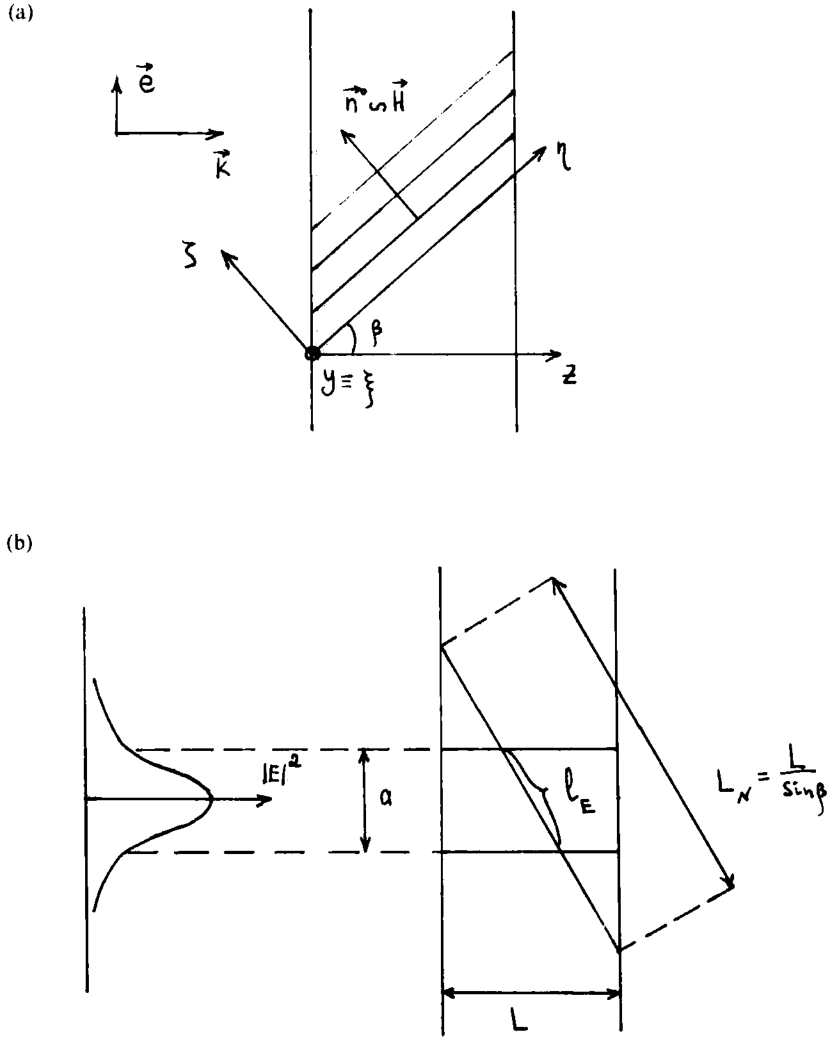

FIGURE 3 (a) the model of a smectic A cell with the optic axis orientation along ζ and with the smectic layers along (η, ξ). Here $y \equiv \xi$ is perpendicular to the plane of the figure. (b) the orientational influence of a light field with transverse size a on the SLC is located only at a length $l_E = a/\cos\beta$, but it disturbs the smectic structure to a length $L_N = L/\sin\beta$.

We find from the incompressibility condition that the displacement $u(\mathbf{r})$ is identical for all points of the N-line. Moreover, the magnitude of the director tilt turns out to be dependent only on ξ and η, i.e. the tilting is the same for all points on the given N-line. From this it follows that the response of smectic A to light fields is non-local in character. Even if the orienting force $\sim EE^*$ is located just on a small part of the given N-line, the response $\delta\varepsilon_{ik}(\mathbf{r}) \sim n_i \delta n_k + n_k \delta n_i$ will be the same along the whole extent of that N-line. Moreover, since the action of the magnetic field, tending to return the director to the initial orientation $\mathbf{n}$, spreads over whole volume of the SLC, the response $\delta\varepsilon_{ik}$ for a given $|E|^2$ turns out to be proportional to the ratio of the part of the N-line covered by the light field l_E, to $L_N = L/\sin\beta$, which is the full N-line length in a cell of thickness L. We shall give below the quantitative grounds, for this statement.

It is convenient, using the independence of $u(\mathbf{r})$ of the coordinate ζ, to integrate Eq. (13) over ζ with fixed ξ, η, i.e. along the whole N-line.

Besides this, for the self-focusing problem all perturbations can be assumed to be fairly smooth. Then we can neglect the terms $\sim k_{11}$, i.e. the Frank energy in Eq. (13). We obtain, as a result

$$\frac{\partial^2 u}{\partial \xi^2} + \frac{\partial^2 u}{\partial \eta^2} = -\left(\frac{\partial D_\xi}{\partial \xi} + \frac{\partial D_\eta}{\partial \eta}\right), \tag{22}$$

$$D_i(\xi, \eta) = \frac{\varepsilon_q}{16\pi\kappa_a H^2} \int_{\zeta_1}^{\zeta_2} d\zeta \{(\mathbf{nE}(\mathbf{r}))E_i^*(\mathbf{r}) + (\mathbf{nE}^*(\mathbf{r}))E_i(\mathbf{r}) - 2n_i |(\mathbf{nE})|^2\}$$

(for simplicity we restrict ourselves by the stationary case, $\dot{u} = 0$). The upper and lower bounds of integration ζ_1 and ζ_2 define the ends of the N-line in the cell and are equal to

$$\zeta_1(\eta) = -L_N + \eta \operatorname{cotan}\beta, \quad \zeta_2(\eta) = \eta \operatorname{cotan}\beta.$$

Since the light field is located over a limited region of space, Eq. (22) for $u(\xi, \eta)$ has to be solved with the condition $u(\xi, \eta) \to 0$ when $\sqrt{\xi^2 + \eta^2} \to \infty$. Taking account of $\delta\mathbf{n} = -\nabla u$, Eq. (22) is brought to the form

$$\frac{\partial}{\partial \xi}(\delta n_\xi - D_\xi) + \frac{\partial}{\partial \eta}(\delta n_\eta - D_\eta) = 0. \tag{23}$$

The solution of Eqs. (22) and (23) has, in the general case, a complicated enough form, especially since the vector $\mathbf{D}$ is given by the integral from the intensity distribution $|E(\mathbf{r})|^2$ with variable bounds of the integral.

Let us note that in the case where $(\operatorname{rot}\mathbf{D})_\xi = \partial D_\xi/\partial_\eta - \partial D_\eta/\partial\xi = 0$, the vector $\delta\mathbf{n}(\xi, \eta)$ equal to $\delta\mathbf{n}(\xi, \eta) = \mathbf{D}(\xi, \eta)$ will be the solution of Eq. (23). We shall use this circumstance for obtaining relatively rapidly the response $\delta\varepsilon_{ik}(\mathbf{r})$ and for making numerical estimations on this basis.

4b Discussion of self-focusing effects

Let us consider a ribbon-like beam falling on the SLC cell. Let the field $\mathbf{E}(r) = \mathbf{e}_x E(x) \exp\{ikz\}$ correspond to the extraordinary wave (the polarization $\mathbf{e}_x$), limitlessly extended in the $y \equiv \xi$ direction and falling on the medium normally to the boundary. For the intensity $I(x) = |E(x)|^2$ transverse distribution width we introduce a notation a.

The distortion picture of the smectic A is essentially dependent on the relation between $l_E = a/\cos\beta$ and $L_N = L/\sin\beta$, the whole length of the N-line in the cell with thickness L (see Figure 4).

The case $a \gg L \operatorname{cotan}\beta$, when the field intensity is practically constant along the whole N-line, corresponds to the greatest magnitude of the constant ε_2. The covering length l_E coincides with the N-line length, and

$$D_\eta = \frac{\varepsilon_a \sin^2\beta}{16\pi\kappa_a H^2}|E(x)|^2, \tag{24}$$

$$D_\xi = 0, (\text{rot}\,\mathbf{D})_\zeta = 0.$$

Then $\delta n_\eta = D_\eta$, and the non-linear addition to the light beam phase $\phi(x)$ equals

$$\phi(x) = \varepsilon_2|E(x)|^2\frac{\omega}{2cn_e}L = \eta P(x)L,$$

$$\varepsilon_2 = \frac{\varepsilon_a^3\sin^2 2\beta}{16\pi\kappa_a H^2}. \tag{25}$$

Thus, when $a \gg L \operatorname{cotan}\beta$ the transverse non-locality of the response cm^3/erg, $\eta = 3.2$ cm/watt, and the phase shift at the beam centre comes to $a = 0.1$ cm $\gg L \operatorname{cotan}\beta = L, \varepsilon_a = 1, \kappa_a H^2 = 9.10^{-1}$ erg/cm^3. Then $\varepsilon_2 = 0.2$ cm^3/erg, $\eta = 3.2$ cm/watt, and the phase shift at the beam centre comes to ~ 6 rad when the beam power density is $P = 200$ watt cm^2. If we also take the beam size in the other coordinate y to be of the order 0.1 cm, the whole power $W \approx Pa^2$ makes up 2 watt. Thus, the possible observation of external self-focusing in a smectic A may be realistic enough. The physical difference between the coordinates x and y in our problem is connected with the strong astigmatism of the light self-focusing in a smectic A.

The distortion picture for a smectic A turns out to be highly interesting in another case, where $a \ll L \operatorname{cotan}\beta$, although the non-linearity must be less in this case. It is convenient to characterise the light beam by two parameters – by the intensity at the centre $|E(x = 0)|^2$, and by the effective transverse size

$$a_{\text{ef}} = |E(x = 0)|^{-2}\int_{-\infty}^{\infty}|E(x')|^2 \mathrm{d}x'. \tag{26}$$

For $\delta n_\eta(\eta) = D'_\eta(\eta)$ we get

$$\delta n_\eta(\eta) = \begin{cases} \dfrac{\varepsilon_a \sin^2 \phi}{8\pi\kappa_a H^2} \dfrac{a_{ef}}{L} |E(0)|^2, & \text{when } 0 \le \eta \le L \cos \beta \\ 0, & \text{for all other cases} \end{cases} \tag{27}$$

Above we assumed that the origin of the coordinate system is at the beam center at the input boundary.

More strictly, Eq. (27) is not true for small areas $|\Delta\eta| \sim a \sin \beta$ near to the interval bounds. In those areas the behaviour of $D_\eta(\eta)$ depends on the specific form of the $|E(x)|^2$ distribution. The light field phase is changed by the amount

$$\phi(x) = a_{ef} \frac{\omega \sin^3 \beta \cos \beta \varepsilon_a^2 |E(0)|^2}{cn_e 8\pi\kappa_a H^2} \left(1 - \frac{|x| \tan \beta}{L}\right), \tag{28}$$
$$\phi(x) = 0 \qquad \text{when } |x| > L \text{ cotan } \beta$$

This expression illustrates, especially vividly, the non-locality of the smectic A response when $a \ll L$ cotan β. Thus, by the use of a weak light beam probe it is possible to measure the phase shift in those areas where the strong wave is absent. In accordance with Eq. (28) this phase depends linearly on x and, thus, the SLC layer serves as a prism relative to the testing beam. Also, this equation is inexact in areas $|\Delta x| \sim a$ near to $x = \pm L$ cotan β, $x = 0$.

As for the external self-focusing, it is necessary, for its description of the strong light beam, to calculate more accurately the integrals for **D** and ϕ. It is possible to obtain the explicit expression for $d^2\phi/dx^2$ for beams with arbitrary profile $|E(x)|^2$

$$\frac{d^2\phi}{dx^2} = -\frac{\omega}{cn} \frac{\varepsilon_a^2}{8\pi\kappa_a H^2} \frac{r^2}{L} \sin^4 \beta$$
$$\cdot \{2|E(x)|^2 - |E(x - L \text{ cotan } \beta)|^2 - |E(x + L \text{ cotan } \beta)|^2\}. \tag{29}$$

This equation is true for the arbitrary relation between the beam size a and parameter L cotan β.

When $a \gg$ cotan β, it coincides with the second derivative of ϕ from Eq. (25). Conversely, when $a \ll L$ cotan β and $|x| \lesssim a$, i.e. inside the beam, we can neglect terms $|E(x \pm L \text{ cotan } \beta)|^2$. Then Eq. (29) demonstrates the peculiarity of the smectic A response. The value of the intensity $|E(x)|^2$ at the given point x of the transverse section defines not the phase raid ϕ, but its second derivative $d^2\phi/dx^2$. Remember that the paraxial focal distance of external self-focusing f is defined by this derivative

$$\frac{1}{f} = \frac{\lambda}{2\pi}\frac{d^2\phi}{dx^2}$$

where λ is wavelength in air.

The phase shift $\phi(x)$ in the case of a Gaussian profile $|E(x)|^2 = |E(0)|^2 \exp\{-x^2/a^2\}$ can be written explicitly for an arbitrary relation between a and L cotan β.

$$\phi(x') = \frac{\omega}{cn}\frac{\varepsilon_a^2|E(0)|^2}{8\pi\kappa_a H^2}\frac{a^2}{L}\sin^4\beta$$
$$\times \{e^{-x'^2}[e^{-L'^2} - 1] + (L' + x')H(L' + x') + (L' - x')H(L' - x')$$
$$- 2x'H(x')\} = \frac{\omega\varepsilon_a^2|E(0)|^2 a^2 \sin^4\beta}{cn8\pi\kappa_a H^2 L}\phi_1(x')$$

Here $H(x')$ is the error integral

$$H(x') = \int_0^{x'} e^{-t^2}dt,\ x' = \frac{x}{a},\ L' = \frac{L}{a}\text{ cotan }\beta.$$

The curve of $\phi_1(x')$ when $a = 0, 2\ L$ cotan β is given in Figure 4.

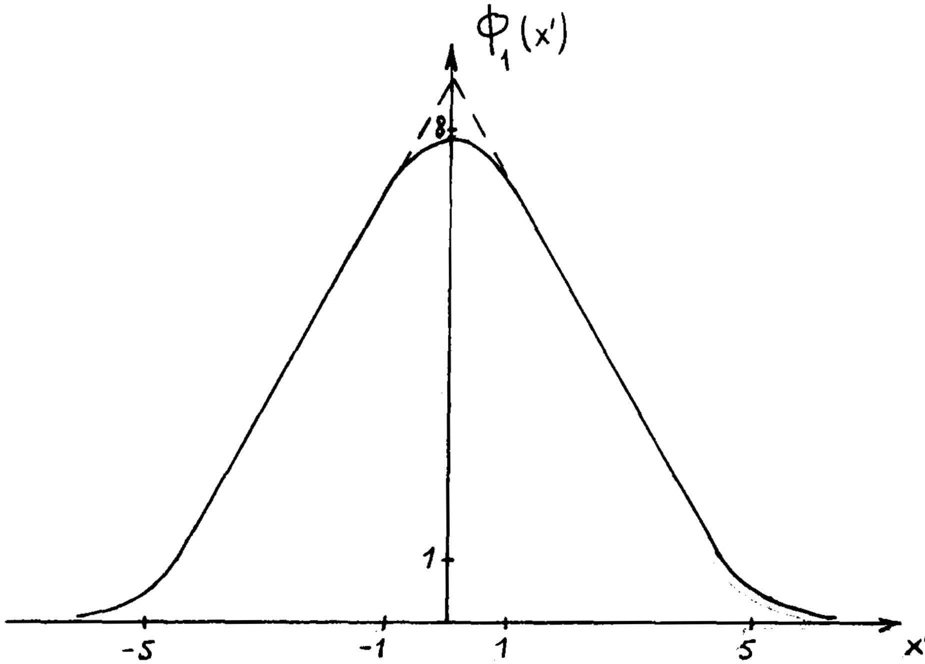

FIGURE 4 The function $\phi_1(x)$ which characterizes the non-linear phase in the beam with transverse size $\Delta x' \sim 1$, when $L(a \tan\beta = 5$.

Numerical estimations for the given power density of the beam lead to a smaller value of the non-linear phase shift $\phi(0)$ than for the case $a \gg L$ cotan β. However, also in this case, the beam power required to register the effect turns out to be fairly realistic. As before, essential astigmatism must occur for self-focusing of two-dimensional beams. The direction of deflection of the testing beam on the coordinates x and y must also be different.

5 CONCLUSION

Very strong effects of cubic optical non-linearity in SLC are predicted and calculated. The experimental geometries for which such a non-linearity must be observed are noted. The discovery and the investigation of these effects are very interesting for possible applications in non-linear optics (self-action, wave front reversal, etc.), and for studying SLC properties. The latter seems to be especially valuable, since many important properties of SLC are still imperfectly understood at present, and the use of non-linear optical methods opens up new possibilities.

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References

1. S. M. Arakelian, G. A. Lyakhov, and Yu. S. Tchilingarian, *UFN*, Vol. 131, No. 1, p. 3 (1980).
2. N. V. Tabiryan and B. Ya. Zel'dovich, *Mol. Cryst. Liq. Cryst.*, in press.
3. N. V. Tabiryan and B. Ya. Zel'dovich, *Mol. Cryst. Liq. Cryst.*, to be published.
4. B. Ya. Zel'dovich, N. F. Pilipetski, V. V. Ragulski, and V. V. Shkunov, *Kvant. Electron.*, **5**, 1800 (1978).
5. A. Yariv, *IEEE J. Quant. Electronics*, **QE-14**, 650 (1978).
6. E. V. Ivankin, B. I. Stepanov, and A. S. Rubanov, *DAN SSSR*, **196**, 567 (1971).
7. R. W. Hellwarth, *J. Opt. Soc. Am.*, **67**, 1 (1977).
8. L. M. Blinov, "Electro- and Magneto-optics of Liquid Crystals," "Nauka," Moscow (1978).
9. B. Ya. Zel'dovich and N. V. Tabiryan, Preprint FIAN N 201 (1978).
10. A. Rapini, *J. Phys. (Paris)*, **33**, 237 (1972).
11. V. I. Khataevitch and A. K. Zeinally, *Pisma ZhTF*, **5**, 651 (1979).
12. Y. Galerne, J. L. Martinand, G. Durand, and H. Veyssie, *Phys. Rev. Lett.*, **29**, 562 (1972).