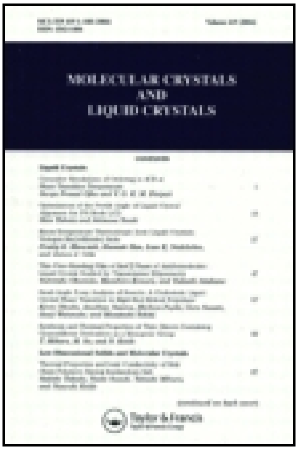

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A. V. Sukhov [a]

[a] Institute for Problems in Mechanics pn Vernadskogo, 101, Moscow, 117526, USSR

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Stimulated Orientational Backscattering and Attendant Phenomena in Cholesteric Liquid Crystal

A. V. SUKHOV

Institute for Problems in Mechanics
pn Vernadskogo, 101, Moscow 11752 6, USSR

Interaction of counterpropagating light waves in cholesteric liquid crystal (CLC) is observed and investigated experimentally, which is due to orientational gratings' excitation in CLC and leads to stimulated backscattering of pump waves.

This process becomes observable by relatively low intensities in case of pump wave frequency close to the selective reflection band of CLC. Self-focusing and nonlinear polarization rotation of pump radiation are studied, which are associated with the stimulated backscattering.

INTRODUCTION

Orientational optical nonlinearities of nematic liquid crystals[1] were investigated in detail recently, including so called "grating" nonlinearities,[2,3] which are due to 3-D orientational gratings of sufficiently small (about 2,5 ÷ 3,5 micrometers) spatial period.

Such gratings are excited by interference pattern of copropagating o- and e-waves in planarly aligned sample and enables one to observe nonlinear interactions (including energy transfer) between two waves, intersecting by rather great angles ($\sim 30 \div 40°$). Nevertheless the abrupt dependence of the effective cubic nonlinear susceptibility upon the wave number of the grating (namely, inverse square dependence) leads to a drastic decrease of the susceptibility by further increase of the intersection angle. In particular, utilization of orientational nonlinearities for energy transfer of counterpropagating waves, including stimulated backscattering (BS) appears to be practically impossible, as the radiation intensities necessary for such energy transfer sufficiently exceed the optical damage threshold of nematic samples available up to date.

Still there is an opportunity of utilization of orientational nonlinearities of liquid crystals for counterpropagating waves' interactions.[4] This opportunity is to use CLC, the helical pitch of which is adjusted so that the laser wavelength lies closely to the selective reflection band of the sample.

In such case a grating of relatively great spatialperiod is required for counter-propagating waves' interaction, namely

$$\Lambda_1 \sim \pi/(\bar{\kappa} - q_0) >> \pi/\bar{\kappa}$$

where $\bar{\kappa} = 2\pi\bar{n}/\lambda$, stands for free space wavelength, q_0 – helix wave number of CLC ($\bar{\kappa} \sim q_0$), $\bar{n}$ – average refraction index. On the contrary, the co-propagating waves' interaction requires a grating with very small period,

$$\Lambda_2 \sim \pi/q_0 \sim \lambda/2 << \Lambda_1$$

which is practically impossible by the reasons mentioned above. So the case of waves' interaction in CLC with selective reflection band near the wavelength of the radiation used—appears to be just "inverse" to the case of interaction in NLC. Namely, the orientational interaction of counterpropagating waves possesses large value of effective cubic susceptibility and is obtainable by relatively low, at any rate non-damaging, intensities. On the other hand, interaction of co-propagating waves, including besides the energy transfer such phenomena, as self-focusing and nonlinear optical activity (NOA), require sufficiently high intensities and cannot thus be observed until the damage threshold of the sample.

STIMULATED BACKSCATTERING

Let's consider the experimental geometry[4] presented in Figure 1. The CLC sample correspond to planar (Grandjean) texture, the local orientational unit vector being

$$\vec{n}(z) = (\sin q_0 z;\ \cos q_0 z;\ 0) + \theta(\cos q_0 z;\ -\sin q_0 z;\ 0)$$

the z-axis being directed along the helix axis, $q_0 = 2\pi/p$—helical "wave number," p—the helical pitch, and θ is a small angle of twist-reorientation induced in our case by the torque exserted by two counterpropagating circularly-polarized light waves, one of which is the pumpwave

$$\vec{E}_{P\pm} = (\vec{e}_x \pm i\vec{e}_y)\, E_{P\pm} \exp(ik_\pm z - i\omega t)$$

and the other is a weak wave (in typical experiment originated by spontaneous scattering of the pump one) the frequency of which is slightly shifted down (so-called Stokes wave)

$$\vec{E}_{S\pm} = (\vec{e}_x \mp i\vec{e}_y)\, E_{S\pm} \exp(-i\kappa_\pm z - i\omega t + i\Omega t)$$

Here subscripts $\pm$ denotes right- or left-circularly polarization of the correspondent wave. Wave numbers $\kappa_\pm$ are separated from selective reflection band edge not less than by $\Delta n q_0$, i.e. $|\kappa_\pm - q_0| \gtrsim \Delta n \kappa_\pm$, $\Delta n = n_\parallel - n_\perp$ is the refraction indice anisotropy of CLC used, so the approximation of circular polarization eigentypes is still valid and one can neglect the linear Bragg's reflection.[5] These waves, interfering in the medium, give rise to the director reorientation of travelling-wave type, the value of which in steady-state regime being following (see Reference 4).

$$\theta(z,t) = -\frac{i\epsilon_a E_P^* E_S}{8\pi}\frac{1}{i\eta\Omega + K_{22}Q_{P,S}^2}\exp(iQ_{P,S}z + i\Omega t) \quad (1)$$

Here $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$ – stands for the dielectric tensor anisotropy of CLC, η – orientational viscosity, K_{22} – Frank's constant, $Q_{P,S}$ – wave number of orientational grating. Subscripts P,S correspond to the polarization types of interacting waves, each of them may be "+" or "–".

Scattering of the pump wave $\bar{E}_P$ by such grating leads to the spatial gain of $\bar{E}_S$ wave, i.e. to the SB of the pump wave (see Reference 1). Steady-state gain coefficient G, optimal frequency shift Ω and correspondent relaxation time of the grating can be easily obtained using traditional theory of stimulated scatterings:

$$G\left(\frac{\text{cm}}{\text{W}}\right) = \frac{10^7\omega^2\epsilon_a^2}{4C^3\kappa_S K_{22}Q_{P,S}^2 n_P};\ \tau = \Omega^{-1} = \eta/K_{22}Q_{P,S}^2 \quad (2)$$

Here c is the light velocity. $Q_{P,S}$ therefore can obtain four different values, corresponding to possible four combination of the "values" of P,S subscripts. They are following:

$$Q_{+,+} = 2(q_0 - \kappa_+);\ Q_{\pm} = (2q_0 - \kappa_+ + \kappa_-)$$
$$= Q_{-,+};\ Q_{-,-} = 2(q_0 + k_-) \quad (3)$$

It's obvious, that the gratings $Q_{+,-}$, $Q_{-,+}$ and $Q_{-,-}$ have spatialperiods of about or even less than the pump wavelength (remember that $q_0 \sim \kappa_\pm$). The value of G is thus extremely low for these gratings and we can omit them in the further consideration, taking into account only the $Q_{+,+}$ grating, i.e. the interaction between right-polarized pump- and Stokes waves. It should be noted here, that we've considered the CLC with "left-twisted" helix only, but the "right-twisted" helix case is quite analogous, only one should substitute $Q_{-,-}$ instead of $Q_{+,+}$. That corresponds to the preferable interaction between left-polarized waves.

So, within our approximation $Q_{+,+}$ can be reduced even to zero by appropriate choice of wave length. Still the opportunity of $Q_{+,+}$ decrease is in real limited by the requirement of conservation of the circulareigenpolarizations regime. The latter demands the wavelength being separated from the selective reflection band edge at least by about the bandwidth. To evaluate the maximum of achievable value for G, we use $\lambda = n_\parallel P + \Delta nP$, just separated by one bandwidth from "red" edge of the reflection band. Simple calculations using (2, 3) give following result:

$$G_{\max} \approx \frac{10^7(n_\parallel + n_\perp)^2\lambda}{32\, n_+^2\ \pi K_{22}C};\ \Omega = \tau^{-1} = \frac{16\pi^2 K_{22}}{2\lambda^2}\Delta n^2 \quad (4)$$

It should be noted that the latter expressions are totally identical to the spatial gain coefficient and frequency shift for the case of forward orientational stimulated scattering in NLC with material constants identical to those of CLC used (see Reference 1). In particular, maximal value of G for the given CLC does not depend upon it's refractive indice anisotropy, and the relaxation time increases drastically and becomes about several milliseconds, which is the typical value for orientational gratings in NLC.

Such great value of relaxation time implies transient regime of SB if excited by a ~1-ms pulse of free-running ruby laser, which was our experimental case. The temporal and spatial evolution of transient Stokes signal, according to the traditional stimulated scattering theory, is following[2]:

$$|E_S|^2 \sim |E_P|^2(t) \exp(I(z,t))$$

where the gain $I(z,t)$ can be expressed as follows:

$$I(t)|_{z=L} = 2^{3/2} (G\tau^{-1}L \int_0^t S(t')\, dt')^{1/2} = 2\left(\frac{10^7\pi\epsilon_a^2 L}{n_+ c\lambda} \int_0^t S(t')dt'\right)^{1/2} \quad (5)$$

Here $S(t)$ [W/cm^2] stands for the current value of Pointing vector z-component of the pump beam, n_+ the refractive index for the right-polarized wave ($n_+ \approx n^-$), L—the sample thickness.

The experimental observation of SB described above was carried out just in the transient regime. The CLC used were mixtures of NLC 5CB with small (about or less than mole 15 per cent) portion of cholesterylnonanoate (CN). The choice of nematocholesteric mixture as the CLC used is due to the requirement of sufficiently high value of dielectric tenser anisotropy $\epsilon_a \approx 0{,}3 \div 0{,}5$, not pertinent to the traditional cholesteric substances. Unfortunately, the temperature dependence of the helical pitch of such mixtures appears to be rather weak, so variations of the helical pith were achieved by varying both temperature and concentration of CN. The free-running pulse of a commercial single transverse mode ruby laser was used as a pump beam, its duration being of about 0,8 ms, energy-several hundreds om mJ. The signal wave was originated within the body of the sample as a result of pump beam spontaneous scattering by thermal fluctuations of director orientation.

Experimental setup is presented in Figure 1. The pump beam formed a 35-μm in diameter illuminated region within the body of the sample, its intensity being uniform across the beam due to the lense L_1, which formed the image of the aperture D_1 on the entrance surface of the sample. Energy of the pump pulse was managed by halfwave plate and Glan prism, controlling simultaneously the pump polarization. Following parameters were controlled during the experiment: total pulse energy by a commercial calorimeter C, temporal evolution of the pump exposition U (i.e. the energy radiated by the laser from the very beginning of the pulse until the current moment t)—by means of integrating photodetector IPD; and also the temporal envelopes of the pump and Stokes pulses ($W_{P,S}$ respectively)—by means of photodetectors $PD_{1,2}$. Time resolution of the latter was maintained about 10 μs, thus eliminating oscillations of about 1 μs, typical for the free-running regime,

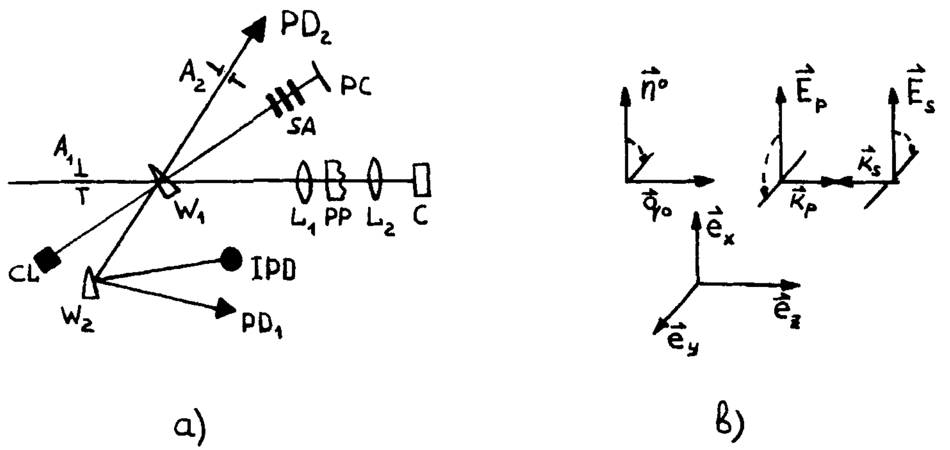

FIGURE 1 a) Experimental setup: $A_{1,2}$ = apertures, CL = calibrating calorimeter, $W_{1,2}$ = fused quartz wedges, PP = phaze plate (inserted only during the OPC-experiments), PD = photodetectors, $L_{1,2}$ = lenses, IPD = integrating photodetector, SA = stepped attenuator, C = CLC cell, PC = photocamera; b) interaction geometry.

since we were interested only in rather slow W_S/W_P ratio changes, originated by the grating evolution.

Besides the energetic characteristics, the angular distribution of the SB wave was registrated by means of multystep attenuator MA and photocamera; and its polarization as well, using the compensator including quarterwave plate and Glan prism.

Properties of the CLC used are presented in Figure 2 and correspondent figure caption. Reflection spectra were measured in depolarized light using commercial spectrophotometer, and the pitch P was calculated using those spectra and the known values of $n_{\parallel,\perp}$ for given mixture. The latter were measured independently, using standard total internal reflection technique. Namely, the TIR angle was measured for the interface between the planarly aligned CLC and a glass prism, the s-polarization of the light, incident through the prism, corresponding to o- or

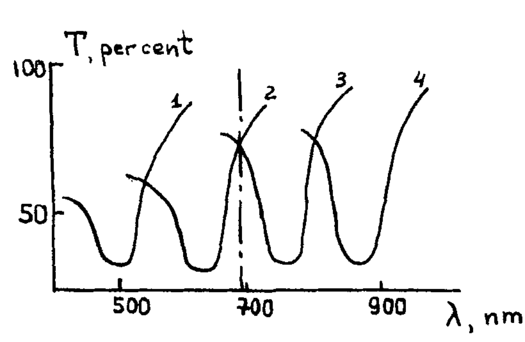

FIGURE 2 Transmission spectra of CLC used (refraction index anisotropy Δn, helical pitch P and mole concentration of CN X are varied): 1) Δn = 0,1701, $\bar{n}P$ = 500 nm, X = 39,9%; 2) Δn = 0,1707, $\bar{n}P$ = 635 nm, X = 32,1%; 3) Δn = 0,171, $\bar{n}P$ = 750 nm, X = 25,4%; 4) Δn = 0,1715, $\bar{n}P$ = 865 nm, X = 18,7%; dashed-dotted line corresponds to laser wave length.

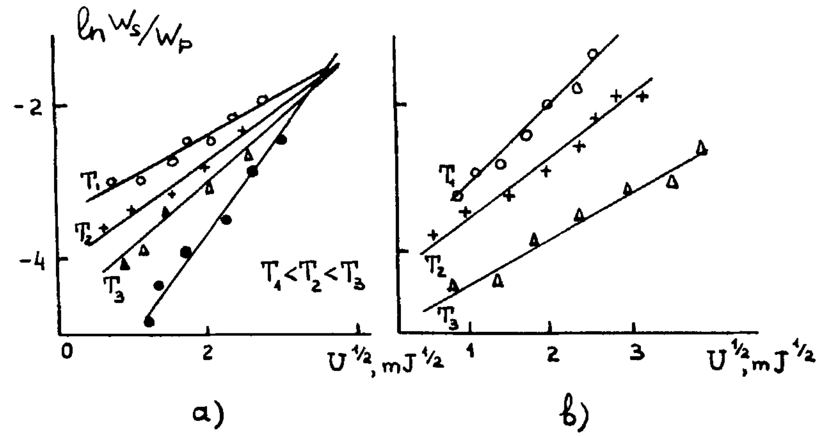

FIGURE 3 SB energetic efficiency dependences of the exposition a) for mixture 2 (Figure 2), b) for mixture 3 (Figure 2).

e-wave for the CLC layer contacting the prism. Qualitatively, the experimental results were following.

The SB signal was absent for mixtures 1 and 4 (Figure 2), selective reflection bands of which are rather far from the pump wavelength—until the optical damage of the samples. For mixtures 2 and 3 (Figure 2) SB signal arises in the case of right-polarized pump beam and vanishes if the latter is left-palarized. The SB signal itself appears to be also right-polarized.

Measurements of energetic dependences, carried out by various temperatures, revealed linear dependence of the gain $I \equiv \ln W_S/W_P - \ln\alpha$ upon $U^{1/2}$ (α is the "spontaneous noise level" within the registration aperture), thus confirming the theoretical prediction (see Figure 3).

The coefficient of this dependence (denoted as ξ), as well as the spontaneous noise level, are sufficiently dependent of the sample temperature. The $\xi(T)$ dependence for two mixtures, presented in Figure 4 is perhaps due to the viscosity

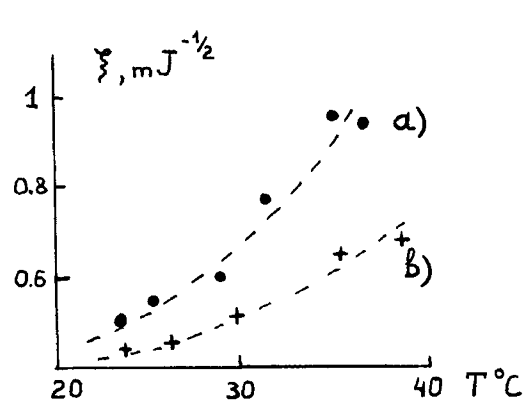

FIGURE 4 Temperature dependences of gain coefficient ξ a) mixture 2, b) mixture 3 (Figure 2).

(η) temperature dependence. It should be noted here, that the experimental values for $\eta(T)$ calculated from $\xi(T)$ using expression (5), vary in the range from 1,5 to 1 puas with the increase of T, which correspond by the order of magnitude to the previously measured values η of 5CB (see, e.g., Reference 3). As to the temperature dependence of spontaneous noise level, it can readily be seen from Figure 3, that it is quite different for the two mixtures. Namely, the noise level increases with growing T for mixture 3 and decreases for mixture 2 (see Figure 5). This fact can be simply explained as follows.

It is well known, that the selective reflection band drifts with the increase of T into the "blue" side, i.e. becomes corresponding to shorter wavelengths, than at low temperatures. So for mixture 2 the reflection peak shift from the pump wavelength increases with the T growth, thus decreasing the spontaneous scattering cross section. On the contrary, for mixture 3 the reflection peak shift decreases with T growth, thus increasing the scattering cross section. Unfortunately one needs very accurate experimental measurements of $P(T)$ dependence to confirm the latter speculations quantitatively, and we've not succeeded in obtaining good enough textures for such accurate measurements of P from the reflection spectra.

As to the SB signal angular distribution, the measurements give a speckle pattern, non-reproducing in different pulses, with typical angular divergence of about 0,4 rad, which one could expect for the transient stimulated scattering, the interaction geometry of which (a cylinder of 35 μm diameter and equal height) allows non-colinear stimulated scattering (see Reference 2).

So one can see that energetic efficiency, polarizational and angular properties of the observed backscattering signal are in good agreement with theoretical approximation delivered above. Orientational SB in CLC requires relatively low (of about 10 ÷ 100 kW/cm^2) pump intensities, and the requirements to its' coherence length are also rather "weak": the coherence length of double cell thickness fits well, that is of about 100 μm. Thus the orientational SB appears to be very promising for aims of optical phase conjugation (OPC) of millisecondrange pulses, say free-

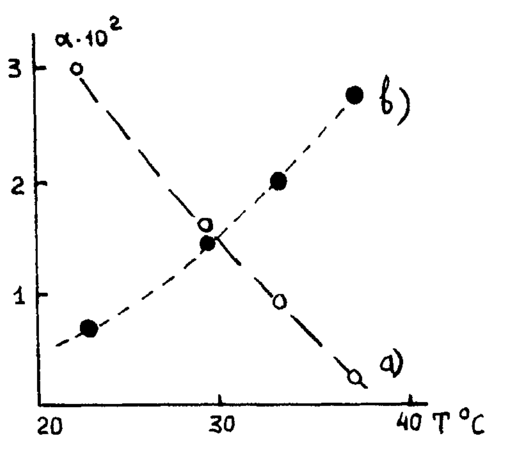

FIGURE 5 Spontaneous "noise level" α a) for mixture 2, b) for mixture 3 (Figure 2).

running pulses of solid-state lasers. We've made attempts to observe such OPC. It is well known (see, e.g., Reference 6), that "discriminational" OPC by SB requires sufficient nonuniformity of the pump radiation within the interaction region. Namely, the Fresnel length of the pump radiation should obey the following relation:

$$l_F \sim \lambda \bar{n}/\vartheta^2 \leq L/I \tag{6}$$

ϑ being the angular divergence (FWHM) of the speckle pump beam, and I—total SB gain. One can simply evaluate, that for $L = 70$ μm (which is the maximum experimental value for high-enough quality texture thickness one can obtain by slab-treating techniques) the angular divergence of $\vartheta \gtrsim 0{,}5$ rad is required to satisfy

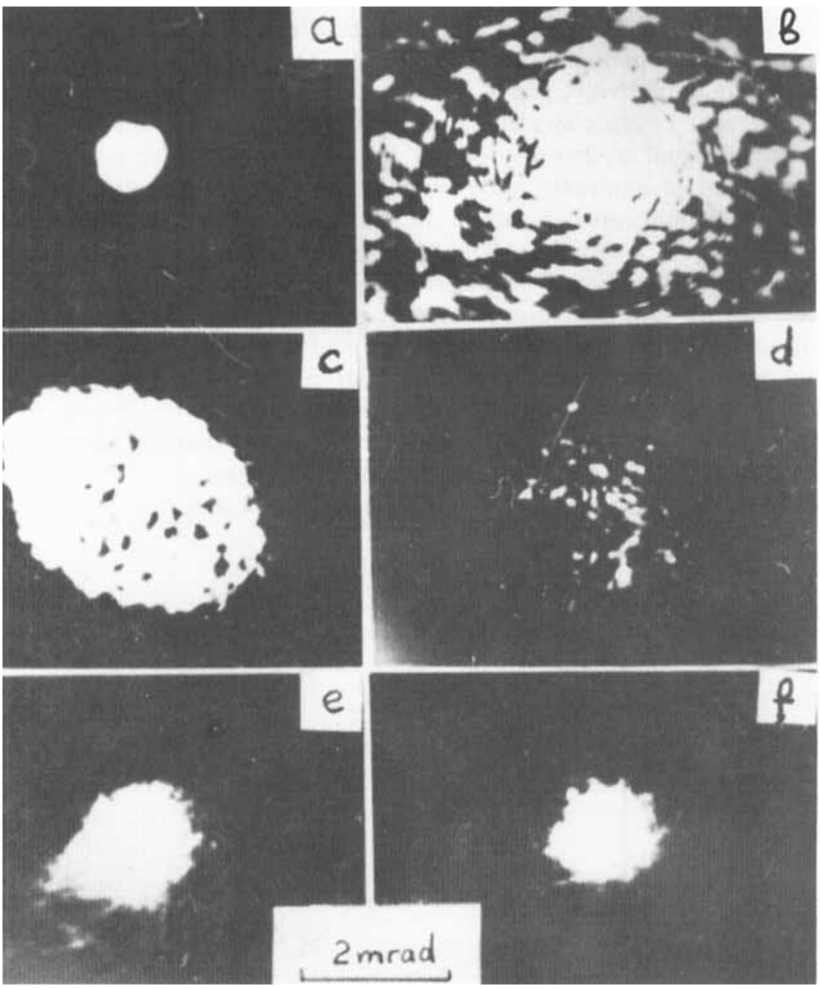

FIGURE 6 Angular distributions of the beams: a) pump; b) pump after the PP; c) linearly reflected by the sample slides after the PP; d) SB (regular pump); e,f) SB (speckle pump).

(6). In our experiment, a phase plate (PP) was inserted into the pump beam, enhancing its' divergence by a factor of 5. Afterwards the radiation was focused into the cell using a short-focus lense L_2. Only partial correction of the angular distribution by SB was observed.

In Figure 6 (e,f) angular distributions of SB signals after passing PP are presented. It can readily be seen that those possess sufficient speckle-component and the angular divergence of the central kern is 1,5 ÷ 2 times more than that of the pump beam before the PP (Figure 6a). Nevertheless the angular divergence of the SB signal after the PP is several times less than that of the incident speckle beam (Figure 6b), or linearly reflected from the sample one (Figure 6c) after passing the PP backwards. Such partial correction is, perhaps due to both rather great Fresuel length (one can hardly obtain $\vartheta \sim 1$ rad divergence of the speckle-beam experimentally) and transient nature of SB. For transient SB (see e.g., Reference 6) the gain for non-conjugated backscattering modes is not twice, but only $\sqrt{2}$ times less than the gain for conjugated ones, so the discrimination of the former is rather "weak."

It should be noted at last, that for rather high power efficiencies of SB achieved in the experiment (up to 40 per cent) the pulse compression of about 1,5–2 times was observed (see Figure 7a), which is also a pertinent feature of transient stimulated scattering.

SELF-FOCUSING AND NONLINEAR OPTICAL ACTIVITY (NOA) IN CLC

The self-focusing and NOA in CLC due to orientational nonlinearity were predicted theoretically.[4] In general feature the predictions were following: NOA doesn't occur

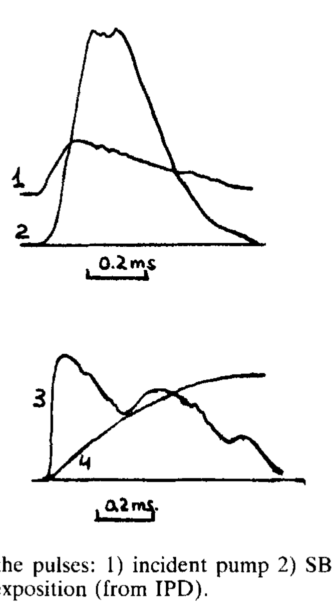

FIGURE 7 Time envelopes of the pulses: 1) incident pump 2) SB signal; 3) transmitted pump (in case of self-focusing); 4) current exposition (from IPD).

for linearly polarized pump radiation. If the latter is ellyptically polarized:

$$\vec{E} = E(\vec{e}_x \cos\nu + \imath e_y \sin\nu)$$

some additional polarization rotation takes place, the "rotation velosity" being

$$\frac{d\varphi}{dz} = A|E|^2 \sin^2\nu;\ A = \frac{\omega\epsilon_a^2}{256\pi cn^- K_{22} q_0^2} \tag{7}$$

The relaxation time of the effect $\pi \sim \eta/K_{22}(2q_0)^2$ and is easily evaluated to be $\tau \leq 10^{-4}$ s, thus leading to steady-state rotation by millisecond-duration pump pulse. As to intensities required, they are of about 5 MW/cm^2, which exceeds the damage threshold of the samples used more than twice (and one should note here, that the mixtures of 5CB and cholesterylnonanoate possess very high damage threshold, e.g. 3,5 times higher than that of MBBA).

Nevertheless, we examined the polarization of transmitted pump beam and obtained an observable rotation by pulse energies, higher than those used by SB-investigation, but still less than the damage threshold. The rotation revealed sufficiently transient behaviour, namely an ambiguous rotation angle dependence of the intensity and strictly linear one of the exposition (see Figure 8).

Rather high values of rotation angle of about 40 degrees were achieved in 35-μm thick sample. Also we should note that the rotation angle didn't depend sufficiently upon the polarization of incident beam, i.e. the rotation occurred for linear polarization as well as for ellyptical one. All these reasons lead to the conclusion that NOA observed in our experiment is originated by laser heating of the medium (thermal relaxation time for 35-μm thick layer is estimated to be several milli-

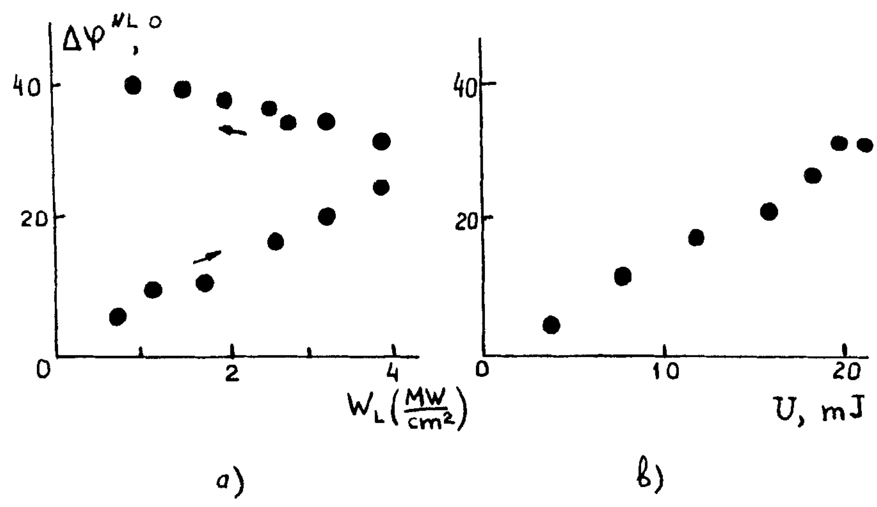

FIGURE 8 Nonlinear polarization rotation angle $\Delta\vartheta^{NL}$ dependences of current values of: a) pump intensity $W_L(t)$; b) exposition $U(t)$.

seconds, thus leading to the transient regime). The comparison of our results with those of the paper,[7] assuming the thermal mechanism of rotation, leads to quite realistic value of absorption coefficient of the sample $\chi \approx 3.10^{-4}$ cm^{-1}.

The same thermal mechanism is perhaps responsible for the transient self-focusing of the pump beam with Gaussian transverse envelope, which was observed in the same experimental conditions with the NOA. Maxima and minima of the intensity in the center of the transmitted spot (corresponding to growing self-focusing rings in the far-field zone, see Reference 1) appeared by quite distinctly fixed values of exposition in different pulses, thus confirming the transient stage of the phenomenon (see Figure 7). Evaluation of self-focusing nonlinearity constant from the experimental data possess the same value of $\chi \approx 3.10^{-4}$ cm^{-1}, thus confirming once more the thermal mechanism of the self-focusing.

CONCLUSIONS

So we can state the nonlinear interaction of counterpropagating light waves was observed, leading to spatial transient gain of spontaneous scattering noise, so-called stimulated backscattering. It is demonstrated, that the excitation of SB-orientational gratings is most effective in case pump wave-length close to selective reflection band of CLC used. Such enhancement of excitation efficiency enables one to observe SB under non-damaging intensities of the pump beam in commercial CLC samples. The possibility of optical phase self-conjugation via orientational SB is demonstrated qualitatively as well.

On the contrary, the co-propagating waves' interactions in CLC, such as orientational self-focusing and NOA, require very high pump intensities and are not in real observable due to optical damage of the samples.

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