Energy exchange between optical waves counterpropagating in a cholesteric liquid crystal

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(Submitted 26 September 1988)
Zh. Eksp. Teor. Fiz. **95**, 1734–1742 (May 1989)

The interaction between optical waves counterpropagating in a cholesteric liquid crystal, due to the excitation of orientational gratings and resulting in stimulated scattering of the exciting waves, was detected experimentally and investigated. This interaction process occurred at relatively low intensities because of the proximity of the radiation wavelength to a selective reflection band of the cholesteric liquid crystal. An investigation was made of the concomitant phenomena of self-focusing and nonlinear rotation of the polarization of the exciting radiation.

Orientational optical nonlinearities of nematics have been investigated recently quite thoroughly[1] and this includes the so-called grating nonlinearities[2,3] due to orientational gratings with a sufficiently small (≈ 2.5–$3.5\,\mu$m) spatial period. Such gratings are formed as a result of interference of ordinary (o) and extraordinary (e) waves in a planar sample of a nematic and give rise to nonlinear interactions (including energy exchange) between waves propagating at a relatively large (≈ 30–$40°$) angles relative to one another. However, the inversely proportional dependence of the third-order effective orientational susceptibility on the square of the grating wave vector reduces strongly the susceptibility on further increase of this angle. In particular, it is practically impossible to use orientational nonlinearities for energy exchange between counterpropagating waves, including orientational backward simulated scattering (STS), since the radiation intensities required for this purpose are considerably higher than the optical damage thresholds of real nematics.

However, it has been suggested[4] that it should be possible to use orientational nonlinearities of liquid crystals to produce interaction between counterpropagating optical waves. This can be done in the case of cholesteric liquid crystals with the pitch of the helix selected in such a way that the wavelength of the exciting radiation lies close to the selective reflection by a given cholesteric liquid crystal (CLC).

The interaction between counterpropagating waves requires excitation of the orientation grating having a relatively large period

$$\Lambda_1 \propto \pi/(\bar{k}-q) \gg \pi/\bar{k},$$

where $\bar{k} = 2\pi\bar{n}/\lambda$; λ is the wavelength of light in a vacuum; q is the wave number of the CLC helix ($\bar{k} \propto q$); $\bar{n}$ is the average refractive index. In the case of concurrent waves the interaction requires a grating with a period

$$\Lambda_2 \propto \pi/q \sim \lambda/2 \ll \Lambda_1,$$

which is practically impossible for the reasons given above. It therefore follows that in the case of a CLC with a selective reflection band near the frequency of the exciting radiation the situation is directly opposite to that in nematics. It is the orientational interaction between counterpropagating waves that is characterized by a high effective cubic susceptibility and can be realized at relatively low (and at any rate nondestructive) intensities. On the other hand, the interaction between concurrent waves and such phenomena as self-focusing and nonlinear optical activity require high intensities and therefore should not be observed right up to the damage threshold of a sample.

The results of an experimental detection and investigation of these effects in a CLC with a selective reflection band near the frequency of the exciting radiation are reported below.

ORIENTATIONAL BACKWARD STIMULATED SCATTERING

We shall consider a planar sample of a cholesteric with an equilibrium pitch of the helix $P = 2\pi/q_0$, where q_0 is the wave number of the helix which (to be specific) is regarded to be left-handed (Fig. 1). The direction of the director in such a cholesteric medium is

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

where z is the coordinate along the helix axis and θ is a small angle of reorientation of the director relative to the equilibrium direction, excited in this case by optical waves (see below). We shall assume that a high-power pump wave

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

is propagating in such a sample in a direction exactly opposite to that of a weak wave which is frequency-shifted to the Stokes region and can be described by

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

The indices $\pm$ represent the right- or left-handed circular polarization of the wave ($k_\pm$ are assumed to be separated from a selective reflection band of the cholesteric by $|k_\pm - q_0| \gtrsim \Delta n k_\pm$, where $\Delta n = n_\parallel - n_\perp$ is the anisotropy of the refractive index of a material, so that we can also assume conservation of the natural circular polarizations and neglect the Bragg linear reflection[5]). Interference of these waves in a medium (see Ref. 4) results in the excitation of reorientation of the director in the form of a traveling wave, which under steady-state conditions is described by

$$\theta(z, t) = -\frac{i\varepsilon_a E_L^* E_S}{8\pi} \frac{1}{i\eta\Omega + K_{22} Q_{L,S}^2} \exp(iQ_{L,S} z + i\Omega t).$$

Here, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the anisotropy of the permittivity of

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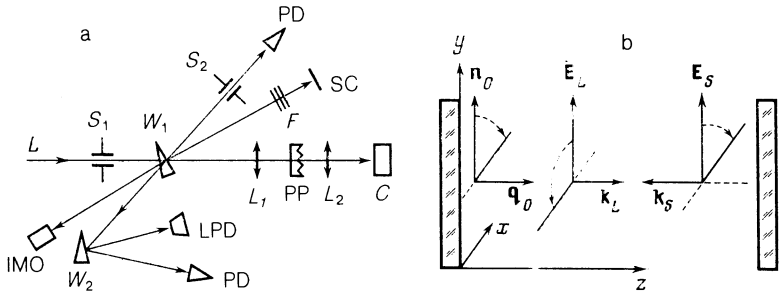

FIG. 1. a) Experimental setup: $S_{1,2}$ are stops; IMO is an energy and power meter (calorimeter); $W_{1,2}$ are fused-quartz wedges; PP is a phase plate; PD is a vacuum photodiode; $L_{1,2}$ are lenses; IPD is an integrating photodiode; F is a step attenuator; C is a cell containing a liquid crystal; SC is a screen. b) Geometry of the interaction showing the orientations of the wave vectors of the pump and signal radiations and of the helix of a cholesteric liquid crystal; y is the direction of the axis of easy orientation at the boundaries of a sample.

the CLC; η is the orientational viscosity; K_{22} is the Frank constant; $Q_{L,S}$ is the wave number of the reorientation grating. The indices L and S refer to the polarizations of the interacting waves and each of them can have the values "+" or "−".

The scattering of the wave $\mathbf{E}_L$ by such a grating is known to result (see, for example, Ref. 1) in an exponential spatial amplification of the wave $\mathbf{E}_S$ i.e., it should result in backward STS of the wave $\mathbf{E}_L$. The steady-state gain G, the optimal frequency shift Ω, and the relaxation time τ of the grating can easily be calculated[1,2] using the traditional STS scattering theory. The results are as follows:

$$G\left(\frac{\text{cm}}{\text{W}}\right)=\frac{10^7\omega^2\varepsilon_a{}^2}{4c^3k_SK_{22}Q_{L,S}^2n_L},\quad \tau=\Omega^{-1}=\frac{\eta}{K_{22}Q_{L,S}^2}, \tag{2}$$

where c is the velocity of light.

We shall now consider the explicit form of $Q_{L,S}$. There are only four following variants, each of which represents a specific combination of the polarizations of the $\mathbf{E}_L$ and $\mathbf{E}_S$ waves:

$$Q_{+,+}=2(q_0-k_+),\ Q_{+,-}=(2q_0-k_++k_-)=Q_{-,+},$$
$$Q_{-,-}=2(q_0+k_-). \tag{3}$$

Hence it is clear that the gratings $Q_{+,-}$, $Q_{-,+}$, and $Q_{-,-}$ have spatial periods of the order of or even less than the wavelength of the exciting radiation λ (we recall that $q_0\sim k_\pm$). Therefore, the gain G for these gratings is extremely low and we can ignore them and concentrate our attention on the grating $Q_{+,+}$, i.e., on the interaction between the pump (L) and signal (S) waves with the right-handed polarization. In the case of a CLC with a right-handed helix the situation is exactly analogous apart from the replacement of $Q_{+,+}$ with $Q_{-,-}$.

The wave number $Q_{+,+}$ can formally be as small as we please if we select a suitable wavelength of light. However, such a possibility of an increase in G is limited by the requirement of conservation of the regime with circular polarizations. This can be done if the wavelength is separated from the selective reflection band by an amount which is at least of the order of the width of this band. In estimates we shall assume that the maximum wavelength is $\lambda=n_\parallel P+\Delta nP$, i.e., that it is separated by the width of the selective reflection band along its long-wavelength edge. After simple transformations, we find that Eq. (2) yields

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

These expressions are completely identical with the expression for G, Ω, and τ in the case of orientational forward STS in a planar nematic sample with the same material constants as the investigated CLC (see Ref. 1). In particular, it is clear from Eq. (4) that the maximum attainable steady-state gain for orientational STS in a selected CLC sample is independent of the anisotropy of the refractive index, and the time needed to form the grating increases to several milliseconds, which corresponds to a change of STS to the time-dependent (transient) regime on excitation by a pulse from a free-running solid-state laser. The gain increment of STS is then described by (see, for example, Ref. 2)

$$I(t)=2^{1/2}\left[G\tau^{-1}L\int_0^t S(t')\,dt'\right]^{1/2}=2\left[\frac{10^7\pi\varepsilon_a{}^2}{n_+{}^2c\eta}\frac{L}{\lambda}\int_0^t S(t')\,dt'\right]^{1/2}. \tag{5}$$

Here, $S(t)$ [W/cm^2] is the instantaneous value of the z component of the Poynting vector of the pump wave, n_+ is the refractive index for light with the right-hand polarization ($n_+\approx\bar{n}$), and L is the length of the cell where the scattering takes place.

Orientational backward STS was realized by us experimentally just in the time-dependent (transient) regime. The investigated CLC were mixtures of the nematic 5CB with cholesteryl pelargonate. This selection was made because of the requirement of a relatively high value of the permittivity anisotropy $\varepsilon_a\approx0.3$–0.5, untypical in the case of traditional cholesterics. The pitch of the helix was varied by altering the composition of the mixture and the temperature of a sample. The pump wave was a pulse from a free-running ruby laser, which was of single-mode nature in respect of the transverse indices, of duration of the order of 800 μs, and of energy up to several hundreds of millijoules. The signal wave appeared inside a sample as a result of spontaneous scattering of the pump pulse by thermal fluctuations of the orientation of the director.

We used the experimental setup shown schematically in Fig. 1. The pump radiation energy was varied by a half-wave plate and a Glan prism, and produced in the sample an illuminated a region with a diameter 35 μm where the transverse distribution of the density was homogeneous. This was achieved by imaging, on the entry face of a cell, of a stop S_1 (which passed the central part of the pump beam), by a lens L_2. Homogeneity of the transverse distribution of the pump intensity made it possible to carry out a more vigorous comparison of the experimental and theoretical results. The total energy of a pump pulse was measured with an IMO calorimeter, the time dependence of the exposure was determined

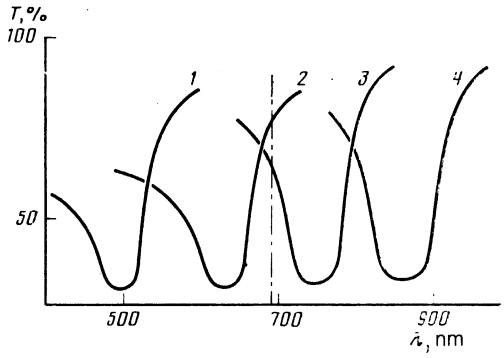

FIG. 2. Dips in the dependences of the transmission coefficient of the samples on the wavelength of light for different parameters of the mixtures: the local anisotropy of the refractive index Δn, the pitch P of the cholesteric helix, and the molar percentage content X of cholesteryl pelargonate in the mixture: 1) $\Delta n = 0.1701$, $P = 500$ nm, $X = 39.93$ %; 2) $\Delta n = 0.1707$, $P = 635$ nm, $X = 32.079$ %; 3) $\Delta n = 0.171$, $P = 750$ nm, $X = 25.406$ %; 4) $\Delta n = 0.1715$, $P = 865$ nm, $X = 18.736$ %. The vertical dash-dot line represents the pump wavelength.

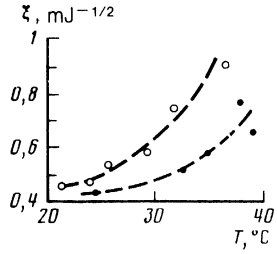

FIG. 4. Influence of temperature on the process of energy exchange [ξ is the coefficient of proportionality in the dependence of $\ln(W_S/W_L)$ on $U^{1/2}$, and T is the absolute temperature]: the open circles represent mixture 2 (Fig. 2) and the black dots represent mixture 3 (Fig. 2).

with an integrating photodetector IPD, and the time envelopes of the pump and backscattering pulses were recorded with a vacuum photodiode PD. Moreover, an investigation was made of the angular spectrum of the backscattering signal by photographing it using a step attenuator F, and the state of the polarization of the signal was determined with a compensator consisting of a $\lambda/4$ plate and a Glan prism.

Our experiments yielded the following results. In the case of mixtures 1 and 4 (Fig. 2), with the selective reflection peak far from the laser (pump) wavelength, the backscattering signal was absent right up to intensities causing damage to a CLC sample. In the case of mixtures 2 and 3 (Fig. 2), when the reflection peaks were located in the immediate vicinity of the laser (pump) wavelength, a backward STS signal was observed for the right-handed circular polarization of the pump radiation (in the case of a mixture with the left-handed helix) and was absent for the pump radiation with the left-handed circular polarization. The backscattering signal itself had the right-handed circular polarization. Measurements carried out at different temperatures T revealed a linear dependence of the gain growth rate $I \equiv \ln(W_S/W_L) - \ln\alpha$ on $U^{1/2}$ ($W_{S,L}$ is the intensity of the signal or pump wave, U is the exposure, and $\ln\alpha$ is the spontaneous noise level), i.e., they confirmed the theoretically predicted exponential amplification of the signal (Fig. 3).

The coefficient of proportionality (denoted by ξ) in the dependence of $\ln(W_S/W_L)$ on $U^{1/2}$ and the spontaneous noise level [$\ln(W_S/W_L)$ when $U = 0$] depended strongly on the temperature of the sample. The dependence $\xi(T)$, plotted in Fig. 4 for two mixtures, was in our opinion due to the temperature dependence of the viscosity η. It should be pointed out that the experimental values of η, converted using the expression for the gain growth rate, ranged within reasonable limits from 1.5 to 1 P on increase in T. The dependence of the spontaneous noise level on T demonstrated, as shown in Fig. 3a for mixture 2, a fall on increase in T, but an increase for mixture 3 (Fig. 3b). As pointed out already, the signal wave was not introduced from outside, but it appeared as a result of spontaneous scattering of the pump wave by thermal fluctuations of the molecule orientation. This was due to the fact that an increase in temperature shifted the selective reflection peak of the CLC toward shorter wavelengths. In the case of mixture 2 it moved away from λ and thus the spontaneous scattering cross section decreased, whereas in the case of mixture 3 the peak approached λ and the scattering cross section increased.

The angular spectrum of the STS signal yielded a speckle pattern (Fig. 5d) which was not repeated from one realization to the next one and had a characteristic divergence 0.4 rad, which was to be expected in the case of time-dependent (transient) STS when the geometry of the interaction region (which was a cylinder with a diameter 35 μm and of the same height) permitted noncollinear STS.

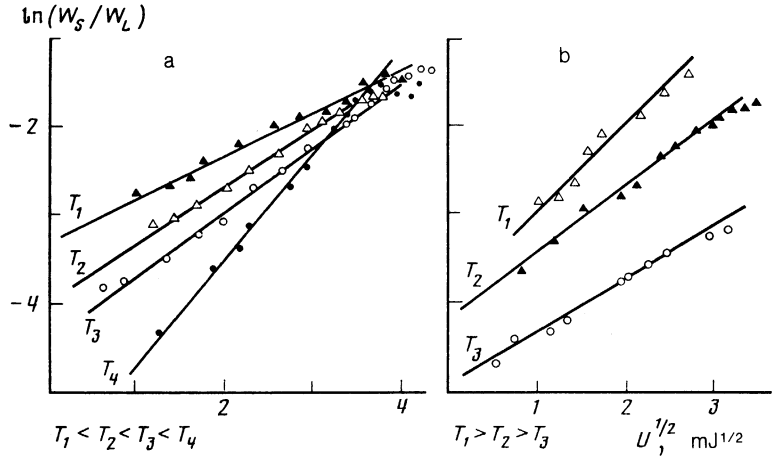

FIG. 3. Dependences of $\ln(W_S/W_L)$ on $U^{1/2}$ demonstrating the dynamics of energy exchange between the pump and signal waves; a) mixture 2 (Fig. 2); b) mixture 3 (Fig. 2).

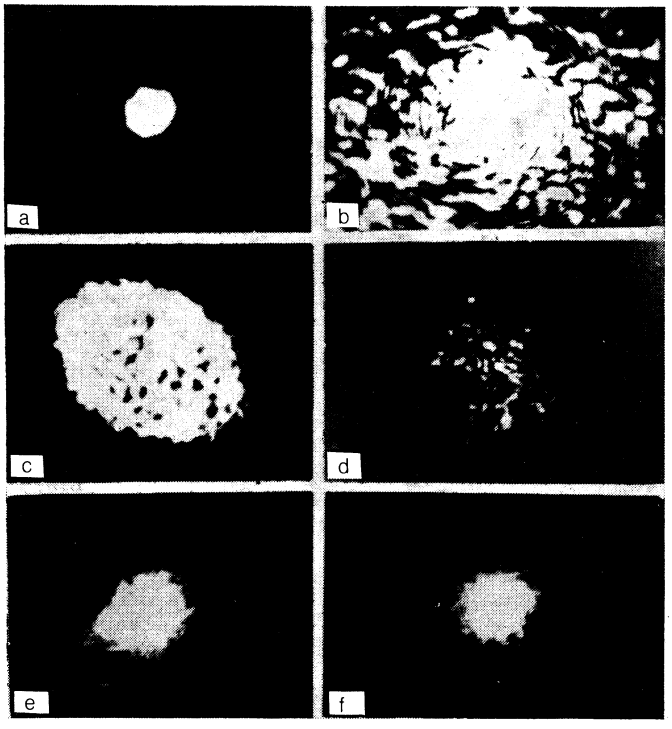

FIG. 5. Angular spectra of the pump waves (a), of the pump waves after transmission through a phase plate (b), linearly backreflected (and transmitted twice by the phase plate) pump waves (c), transient backward stimulated scattering of the signal (d), and backward stimulated scattering signals demonstrating partial phase conjugation after passage through the phase plate (e, f).

We thus found that the energy, polarization, and angular characteristics of the observed backscattering signal agreed well with theory. Orientational backward STS in the investigated CLC required relatively low ($\sim$10–100 kW/cm^2) power densities of the exciting radiation with a low coherence: a coherence length equal to twice the interaction length, i.e., $l_{coh} \approx 100\,\mu$m, was needed. These properties made it attractive to use orientational backward STS for the purpose of phase conjugation of wavefronts of millisecond pulses. We attempted to achieve such phase conjugation. It was known (see, for example, Ref. 6) that to achieve phase conjugation as a result of discrimination during amplification of the backscattering components which were not spatially correlated with the exciting radiation field one would have to ensure that this field is strongly inhomogeneous in the interaction region. Specifically, the Fresnel length of the pump speckle field should obey

$$l_F \propto \lambda\bar{n}/\vartheta^2 \leqslant L/I. \qquad (6)$$

Here, ϑ is the width at half-amplitude of the angular spectrum of the exciting radiation and I is the STS gain growth rate. Estimates indicated that for the interaction length $L \lesssim 70\,\mu$m (for samples of greater thickness a simple treatment of the substrate surfaces produced a satisfactory texture of the CLC) the requirement described by Eq. (6) was satisfied when $\vartheta \gtrsim 0.5$ rad. With this in mind, the initial pump spectrum (Fig. 5a) was distorted by a phase plate P (Fig. 1) so that its divergence increased approximately fivefold (Fig. 5b). The resultant radiation was focused inside the sample by a short-focus lens L_2. This ensured partial correction of the wavefront under backscattering conditions.

Figures 5e and 5f show the angular distributions of the backscattering signals after the second pass through the phase plate. Clearly, their divergence was considerably less than the divergence of the speckle beam incident on the sample or the divergence of the linearly reflected (again after two passes through the phase plate) radiation traveling in the backward direction (Fig. 5c). However, the divergence of the STS radiation (Figs. 5e and 5f) was considerably higher (by a factor of 1.5–2) than the divergence of the exciting radiation and had a strong speckle component, so that there were no grounds for assuming that phase conjugation took place. In our opinion, the partial nature of the wavefront correction was due to the fact that the condition (6) was not satisfied sufficiently rigorously (speckled beams with a divergence $\gtrsim 1$ rad were difficult to create experimentally), as well as to the transient nature of the STS process itself. The point is that (see Ref. 6) in the transient regime the gain growth rate for uncorrelated modes was not half that of the correlated modes, but only $\sqrt{2}$ times smaller.

In comparing the time envelopes of the exciting radiation and STS signal pulses we should also bear in mind that in the case of high efficiencies of conversion into the STS signal (in our experiments the conversion efficiency was up to 40% in respect of the intensity) there was a characteristic time compression of the transient STS signal by a factor of 1.5–2 compared with the exciting radiation pulse (Fig. 6).

SELF-FOCUSING AND NONLINEAR OPTICAL ACTIVITY OF A CHOLESTERIC LIQUID CRYSTAL

It is predicted in Ref. 4 that self-focusing and nonlinear optical activity should occur in cholesterics as a result of the orientational nonlinearity mechanism. These predictions can be summarized briefly as follows: the nonlinear optical activity does not occur in the case of linearly polarized radiation. However, in the case of an elliptic polarization,

$$\mathbf{E}=\mathbf{E}(\mathbf{e}_x \cos\nu + i\mathbf{e}_y \sin\nu),$$

in addition to the linear rotation of the axis of the ellipse of a CLC there is an additional rotation at a rate

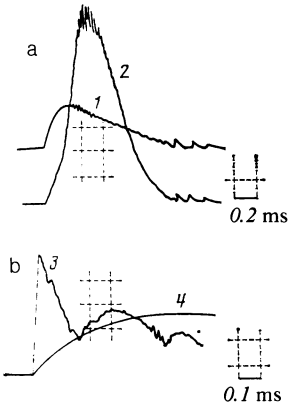

FIG. 6. a) Time envelopes of the pump radiation before reaching a sample (1) and of the transient backward stimulated scattering of the signal (2). b) Time envelopes of the pump radiation after passage through a sample (curve 3 represents peaks and dips demonstrating the dynamics of growth of self-focusing rings) and of the exposure (4).

$$\frac{d\varphi}{dz} = A|E|^2 \sin 2\nu, \quad A = \frac{\omega \varepsilon_a^2}{256\pi c \bar{n} K_{22} q_0^2}. \qquad (7)$$

The time needed to establish this effect is $\tau \sim \eta/K_{22}(2q_0)^2$, which in our case was $\tau \leqslant 10^{-4}$ s, i.e., the effect should be of steady-state nature in the case of excitation by millisecond pulses. The required intensities estimated from Eq. (7) gave $\gtrsim 5.5$ MW/cm^2, which corresponded to energies of $\gtrsim 40$ mJ and these exceeded the damage threshold of the samples. It should be pointed out that our mixtures based on nematic 5CB had a very high (for a liquid crystal) optical damage threshold, which was approximately 3.5 times higher than that of MBBA.

We observed experimentally a strong nonlinear rotation of the plane of polarization of the incident pulse when its energy density exceeded that needed to realize orientational STS. The rotation was very much a time-dependent effect: the dependence of the angle of rotation on the pump intensity was multivalued (loop-shaped) and the dependence on the exposure was strictly linear (Fig. 7). At pump energies of ≈ 10–15 mJ the angle of nonlinear rotation of the plane of polarization reached $\Delta\varphi^{NL} \approx 32°$ (for a sample of thickness $L = 35$ μm). The characteristic parameters of the process, namely, its transient nature, the size of the angle of rotation, and the absence of a significant dependence of this angle on the type of polarization of the incident radiation, all indicate that the rotation was due to a nonorientational mechanism. For pulses of duration $\tau_p \approx 0.8$ ms and a sample of thickness $L = 35$ μm it was found that the thermal processes could be of cumulative transient nature (the time for the removal of heat to the substrate amounted to several milliseconds). A comparison of the quantitative results obtained by us with those reported in Ref. 4 on the basis of the thermal mechanism yielded realistic values of the absorption coefficient $\chi \approx 3 \times 10^{-4}$ cm^{-1}.

The same thermal mechanism was in our opinion responsible for self-focusing of the exciting radiation with a Gaussian transverse intensity distribution, observed under the same experimental conditions. It was also transient: in the case of pulses with different energies the maxima and minima of the intensity fluctuations at the center of the transmitted beam (growing self-focusing rings in the far-field zone—see Ref. 1) appeared for certain fixed values of the exposure. Figure 6b shows the time dependence of the exposure (represented by a continuous curve 4 reaching a plateau at the end of a pulse) and a time envelope of the intensity at the center of the beam transmitted by the CLC (three peaks corresponded to three self-focusing rings). Estimates of the self-focusing nonlinearity constant deduced from the experimental values gave $\chi \approx 3 \times 10^{-4}$ cm^{-1} for the absorption coefficient, which indicated that the observed self-focusing was of thermal nature.

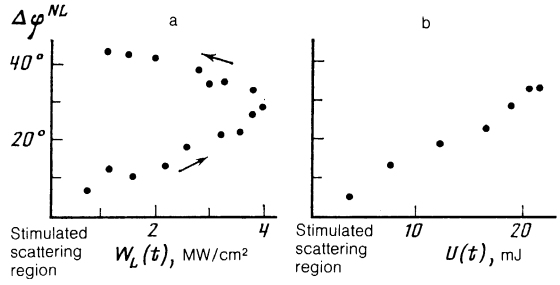

FIG. 7. Dependences of the angle of nonlinear rotation ($\Delta\varphi^{NL}$) of the plane of polarization of the pump radiation on the instantaneous values of: a) pump intensity $W_L(t)$; b) exposure $U(t)$.

CONCLUSIONS

We realized experimentally an orientational nonlinear interaction (stimulated scattering) of counterpropagating optical waves in a cholesteric liquid crystal. It was found that the use of a cholesteric with a selective reflection band near the exciting radiation wavelength made it possible to reduce significantly the energy density of the exciting radiation needed to achieve backward stimulated scattering, so that such scattering occurred at intensities that did not damage real cholesteric samples. A qualitative demonstration was also given of the feasibility of correction of wavefronts by backward stimulated scattering. It would be desirable to carry out an additional investigation in order to achieve phase conjugation of wavefronts of millisecond pulses as a result of such stimulated scattering.

In contrast to the interaction of counterpropagating waves, the interaction of concurrent waves and also self-focusing and non-linear optical activity due to the orientational mechanism of the nonlinearity of cholesteric liquid crystals require very high radiation intensities and their realization is prevented by optical damage to real cholesterics.

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Translated by A. Tybulewicz