Nonstationary optical excitation of bulk short-period orientation lattices in a nematic

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The process of excitation under nonstationary conditions of spatially periodic volume reorientation of the director in a planar nematic sample by the field of interfering *o* and *e* waves is investigated theoretically and experimentally. The self-similarity of the process, i.e., the dependence of all the parameters of the process on a single spatiotemporal variable, is elucidated. The processes of stimulated scattering, four-wave mixing, and nonstationary energy exchange between the waves in the course of the reorientation are investigated in detail. The effect on these processes of the warming up of the medium as a result of absorption is elucidated.

The nematic mesophase's orientational optical nonlinearity, which is being actively investigated at present by many research groups, can, as is well known, give rise to extremely high ($\sim 10^{-1}$–1 cm^3/erg) nonlinear cubic susceptibilities. This has allowed the realization of many traditional nonlinear optical phenomena, such as self-focusing of light,[1] four-wave mixing, and self-diffraction,[2–4] with the aid of low-power cw gas lasers. But along with this obvious advantage, orientational nonlinearity, based on the excitation by light in a nematic sample of orientational deformation with a characteristic inhomogeneity scale of the order of the sample thickness ($\sim 100\ \mu$m) [the so-called giant optical nonlinearity (GON)], possesses a number of important disadvantages.

First, its establishment time is extremely long (~ 1 sec). Second, the degree of stationary reorientation of the director of a nematic and, hence, the modulation of the refractive index of the medium are proportional to the square of the characteristic scale of the inhomogeneity of the spatial orientation distribution. This gives rise to an extremely low (not better than ~ 30 lin/mm) spatial resolution of the medium in an attempt to use the orientational mechanism of nonlinearity in the problems of dynamical holography, since lattices with a large wave number correspond to a much smaller nonlinear susceptibility. Third, the use of GON in dynamical-holography problems is accompanied by unavoidable self-focusing of the reference waves, which worsens the quality of reproduction of the wave fronts. Finally, the GON mechanism makes the recording of three-dimensional holograms, i.e., orientation gratings with the substantial inhomogeneity along the sample thickness, absolutely impossible.

The purpose of the present paper is to demonstrate theoretically and experimentally the fact that we can eliminate these shortcomings of the orientational nonlinearity by a correct choice of the interaction geometry and duration.

BASIC SYSTEM OF EQUATIONS

Let us consider the interaction geometry shown in Fig. 1 for light waves in nematic. Let there propagate in an oriented planar nematic sample of thickness L, in a direction perpendicular to its optical axis, an ordinary wave

$$\mathbf{E}_o=(\mathbf{e}_y\cos\delta+\mathbf{e}_z\sin\delta)E_o(z,t)\exp(i\mathbf{k}_\perp\mathbf{r}-i\omega t)$$

and extraordinary wave

$$\mathbf{E}_e=\mathbf{e}_xE_e(z,t)\exp(i\mathbf{k}_\parallel\mathbf{r}-i\omega t),$$

whose wave vectors $|\mathbf{k}_{\parallel,\perp}|=(2\pi/\lambda)n_{\parallel,\perp}$ make equal small angles δ with the normal to the sample surface. Here $n_{\parallel,\perp}$ are the principal values of the refractive index, and λ is the wavelength in vacuo. The interference of the waves $\mathbf{E}_o$ and $\mathbf{E}_e$ leads to the excitation in the nematic of a spatially periodic orientational deformation $\delta\mathbf{n}=\theta_R\cdot(\mathbf{e}_y\cos\delta+\mathbf{e}_z\sin\delta)$ with wave vector $\mathbf{q}=\mathbf{k}_\parallel-\mathbf{k}_\perp$ [the so-called lattice optical nonlinearity (LON) of the nematics (see Ref. 5)]. Of importance is the fact that, because of the large difference between the phase velocities of the *o*- and *e*-waves in the nematic, $\mathbf{q}$ has a substantial $\mathbf{z}$ component, i.e., the grating described is three-dimensional. The rescattering of the $\mathbf{E}_o$ and $\mathbf{E}_e$ waves by this grating leads in the general case to their volume self-diffraction. The system of equations for the description of the indicated reorientation, a system which consists of the simplified Helmholtz equations for the $\mathbf{E}_{o,e}$ waves and the dynamical equation obtained for the reorientation θ_R within the framework of the Oseen-Frank continuum theory in the single-constant approximation by the method of variation of the free-energy functional of the system, has the form

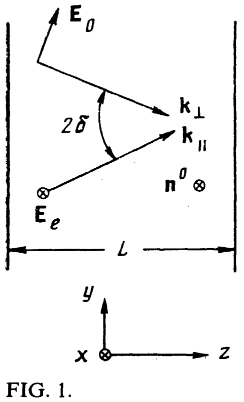

FIG. 1.

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$$\frac{\partial E_e}{\partial z}=\frac{i\varepsilon_a\pi\theta_R}{\lambda n_\parallel \cos\delta}e^{iqr}E_o,$$
$$\frac{\partial E_o}{\partial z}=\frac{i\varepsilon_a\pi\theta_R}{\lambda n_\perp \cos\delta}e^{-iqr}E_e,$$
$$I\frac{\partial^2\theta_R}{\partial t^2}+\eta\frac{\partial\theta_R}{\partial t}+K_1\Delta\theta_R=\frac{\varepsilon_a}{16\pi}(E_oE_e^*e^{iqr}+\text{c.c.}). \quad (1)$$

Here $\varepsilon_a = n_\parallel^2 - n_\perp^2$ is the permittivity anisotropy, K_1 the Frank constant (taken, for simplicity, in the single-constant approximation); $\eta[P]$ the orientational viscosity of the nematic, and I the orientational moment of inertia per unit volume and ranges from 10^{-8} to 10^{-7} g/cm. Let us emphasize that the first two equations of the system (1) do not contain terms corresponding to self-modulation of the phases of the waves, since, in the approximation linear in θ_R, neither of the waves is separately capable of causing reorientation (see Ref. 5), and, thus, their self-focusing is not possible. Let us turn to the third equation of the system. As is well known (see, for example, Ref. 6), the inertial term ($\sim I/\tau_p^2$) of this equation is comparable to the viscous term ($\sim\eta/\tau_p$) only in the case of extremely short characteristic times of the process: $10^{-7}\lesssim\tau_p\lesssim10^{-5}$ (in our case τ_p is the laser pulse duration). Bearing this limitation in mind, we shall henceforth discard the inertial term in the third equation.

The solution to the system obtained (see, for example, Ref. 5) gives for the modulation amplitude θ_R a steady-state value proportional to $|\mathbf{q}|^{-2}$, i.e., essentially dependent, in particular, on the angle 2δ of convergence of the beams. This nonlocality can, as asserted above, lead to appreciable worsening of the quality of reproduction of the holograms of speckle beams with the use of orientational nonlinearity. But under conditions of essentially nonstationary reorientation excitation, when the radiation pulse duration $\tau_p \ll \tau = \eta/K_1q^2$, the third term in the left member of the third equation of the system (1) can be discarded. Thus, the nonlocality of the response of the medium disappears. Below we shall consider this range of durations, which corresponds numerically to $10^{-5}<\tau_p<10^{-3}$ sec for typical K_1, η, and δ.

If now we introduce the complex reorientation amplitude $\theta_R = \theta e^{iq\cdot r}+\text{c.c.}$, and retain in the first two equations of the system (1) only the terms satisfying the Bragg condition, we obtain

$$\frac{\partial E_e}{\partial z}=\frac{i\pi\varepsilon_a}{\lambda n_\parallel\cos\delta}\theta E_o,$$
$$\frac{\partial E_o}{\partial z}=\frac{i\pi\varepsilon_a}{\lambda n\ \cos\delta}\theta^* E_e, \quad (2)$$
$$\frac{\partial\theta}{\partial t}=\frac{\varepsilon_a}{16\pi\eta}E_eE_o^*.$$

Below we assume δ to be small, and replace $\cos\delta$ by unity. Let $A=E_en_\parallel^{-1/2}$, $B=E_on_\perp^{-1/2}$, $q=\varepsilon_a^2/16\lambda\eta n_\parallel n_\perp$, $\sigma=\pi\varepsilon_a\theta/\lambda(n_\parallel n_\perp)^{1/2}$. Then

$$\frac{\partial A}{\partial z}=i\sigma B,\quad \frac{\partial B}{\partial z}=i\sigma^*A,\quad \frac{\partial\sigma}{\partial t}=qAB^*. \quad (3)$$

This system of equations possesses the remarkable property that it can be reduced to a system of ordinary differential equations.[7] Indeed, assuming that A and B at the entrance to the medium differ only by a constant phase factor, possessing the same temporal envelope, i.e., that

$$A(z,t)=\alpha(z,t)E(t),\quad B(z,t)=\beta(z,t)E(t),$$

introducing the new variable

$$y=qz\int_0^t|E(t')|^2dt'$$

and setting $\sigma = yM/z$, we obtain

$$d\alpha/dy=iM\beta\quad d\beta/dy=iM^*\alpha,\quad dM/dy=[\alpha\beta^*-M]/y,$$
$$\alpha(0)=\alpha_0,\quad |\alpha_0|^2+|\beta_0|^2=1,\quad \beta(0)=\beta_0,$$
$$M(0)=\alpha_0\beta_0^*,\quad (dM/dy)_0={}^1/_2i\alpha_0\beta_0^*[|\beta_0|^2-|\alpha_0|^2]. \quad (4)$$

Here the additional condition on dM/dy is required because of the singularity of the third equation of the system at the origin. The system (4) is the basic system of equations for the description of the processes governed by LON under conditions of nonstationary excitation. Following directly from it is the important result that all the characteristics of these processes are determined not by the two variable z and t, but by one self-similar variable, namely, the product of z and the exposure, defined as the density of the radiation energy that has passed through the sample up to the moment of time in question.

FOUR-WAVE MIXING AND WAVE-FRONT INVERSION (PHASE CONJUGATION)

It is natural to begin the investigation of the system (4) with the simplest particular case in which one of the waves (for definiteness, the wave $\mathbf{E}_e$, as was the case in our experimental situation) is of much higher intensity than the other and is undamped ($\alpha\equiv1$) and the exposure is short (i.e., y is small). The expression for $M(y)$ in this approximation follows directly from the boundary conditions

$$M(y)=M_0+\left(\frac{dM}{dy}\right)_0y=\beta_0^*+\frac{i\beta_0^*y}{2}\approx\beta_0^*. \quad (5)$$

If now we direct counter to the $\mathbf{E}_e$ wave another high-power (reference) wave $E_e^-=\alpha_-(y)E(t)n_\parallel^{-1/2}C$, where $\alpha_-(y)=\text{const}$, then its scattering by the described grating will lead to the appearance of a wave $E_o^-=\beta_-(y)E(t)n_\perp^{-1/2}$ with a wave front that is inverted with respect to $\mathbf{E}_o$. If we take account of the fact that, in the Born approximation used by us the diffractive effectiveness of the grating is the same for scattering of the wave $\mathbf{E}_e^-$ into $\mathbf{E}_o^-$ and $\mathbf{E}_e$ into $\mathbf{E}_o$, then we can immediately obtain the answer for the amplitude $|\beta_-|^2$ at the exit from the medium: $|\beta_-|^2=|\alpha_-|^2|\beta-\beta_0|^2$. In the approximation being used $d\beta/dy=iM^*=i\beta_0$, so that $\beta(y)=\beta_0+i\beta_0y$. The final expression for the coefficient of reflection under conditions of four-wave mixing (FWM) in the Born approximation is as follows:

$$R^{NL}=\frac{W_o^-(y)}{W_o(y)}=\frac{|\beta_-|^2}{|\beta_0|^2}=|\alpha_-|^2y^2. \quad (6)$$

Here W_o and W_o^- are the power densities of the corresponding waves. Thus, the running coefficient of reflection depends only on the ratio of the intensities of the reference waves and the self-similar variable y, and does not depend on the angle 2δ of convergence of the recording waves, which allows us, in particular, to realize the inversion of the wave front. Such FWM, accompanied by wave-front inversion, has been experimentally observed.[8] In the experiment a planar cell with a 5CB-nematic sample of thickness $L = 140$ μm was used. The source of radiation was a single-mode—with respect to the transverse index—rubidium laser ($\lambda = 0.6943$ μm) operating in the free-running regime (pulse duration $\tau_p \approx 1$ msec), with energy in a pulse ~ 150 mJ and beam divergence $\theta_0 = \mathrm{FWHM} = 6\times10^{-4}$ rad. The pulse length ensured the fulfillment of the nonstationarity condition $\tau_p < \tau$ for reorientation gratings with spatial period $\Lambda \geqslant 1.4$ μm, which corresponded to an $\mathbf{E}_e$- and $\mathbf{E}_o$-beam convergence angle $2\delta_0 \lesssim 0.5$ rad (in air). The laser radiation was split up by polarizing devices into an e-polarized beam $\mathbf{E}_e$ and an o-polarized beam $\mathbf{E}_o$, whose power was smaller than that of the $\mathbf{E}_e$ by a factor of ten. These two beams intersected in the sample, which was located at the focal constriction of a telescope formed by lenses with focal length $f = 25$ cm. The telescope was used to increase the power density of the reference waves $\mathbf{E}_e$ and $\mathbf{E}_e^-$ in the sample, constricting them down to a diameter a (FWHM) $\approx 150\,\mu$m. The second reference wave was produced by reflection, from a mirror, of the wave $\mathbf{E}_e$ after it has passed through the telescope. The ratio of the intensities of the reference waves was $|\alpha_-|^2 = 0.5$. The following parameters were measured: W_o, W_o^-,

$$Q[\mathrm{mJ}] \sim \int_0^t |E(t')|^2 dt'$$

(with the aid of an integrating network), the total energy $Q_0^-(\infty)$ of the reversed wave, and the angular divergences of the $\mathbf{E}_o^-$ and $\mathbf{E}_o$ waves (by photometrically scanning the beam photographs at the focus of an $f_1 = 100$ cm lens with the use of a step attenuator).

The following results were obtained: Increase of the pulse energy Q, produces a signal $\mathbf{E}_o^-$ that depends nonlinearly on Q. The temporal envelopes W_o and W_o^- differ essentially in shape, the difference being characteristic of nonstationary FWM. The dependence of $R^{NL}(t)$ on the square of the running value of the pulse energy, $Q^2(t)$, has been measured for different angles $2\delta_0$ of convergence of the beams (Fig. 2a). It can be seen that, irrespective of the total pulse energy and the angle $2\delta_0$, all the results fall on one straight line, in accordance with (5). Running values $R^{NL} \approx 30\%$ have been achieved. Notice that, in the steady-state case, R^{NL} for $2\delta_0 = 0.38$ rad should have been smaller by a factor of 25 than the corresponding value for $\delta_0 = 0$. The solid circles in Fig. 2a correspond to the value $\delta_0 = 0$; the open circles, to $\delta_0 = 0.19$; and the triangles, to $\delta_0 = 0.015$.

The proportionality coefficient of the indicated dependence turned out to be four times smaller than the theoretical value, which was computed using a beam dimension $a = 150\,\mu$m. This discrepancy can be explained by the uncertainty in the overlap of the interacting beams, the error in the determination of the dimension a, and a number of other causes of a technical nature. The dependence of the energy coefficient of reflection, R_Q, on $Q^2(\infty)$ (Fig. 2b) also turned out to be linear, which agrees with theory subject to the additional condition that the shapes of the temporal envelopes for different pulses are similar, a condition which was fulfilled in our experiment. Values $R_Q \approx 10\%$ have been attained.

The locality of the nonstationary orientational nonlinearity allowed us to realize the inversion of the wave front of an $\mathbf{E}_o$ signal with a finite angle spectrum. For this purpose, we introduced into the $\mathbf{E}_o$ beam a phase plate that increased the initial beam divergence $\theta_0 = 6\times10^{-4}$ rad (Fig. 3a) to 3×10^{-3} rad (Fig. 3b), left the energy coefficient of reflection unchanged the divergence of the $\mathbf{E}_o^-$ signal, i.e., the signal that traversed it in the opposite direction, practically back to θ_0, which attests the wave-front inversion (WFI) of the $\mathbf{E}_o$ wave (Fig. 3c). Thus, experiment confirms the assumed self-similar nature of the nonstationary orientation-lattice recording and the locality of the LON under nonstationary conditions.

STIMULATED ORIENTATIONAL FORWARD SCATTERING OF LIGHT

Let us now consider the same situation with an undamped pump of the e-type, but at sufficiently high values of y, assuming β_0 (the noise due to the spontaneous scattering of the e-wave by the thermal fluctuations of the director) to be small. The system (4) reduces to the following system:

$$\frac{d\beta^*}{dy} = -iM, \quad \frac{dM}{dy} = \frac{1}{y}[\beta^* - M],$$
$$\beta(0) = \beta_0, \quad M(0) = \beta_0^*, \quad \left(\frac{dM}{dy}\right)_0 = -\frac{i\beta_0^*}{2}. \qquad (7)$$

Let us note that the results do not, as before, depend on δ. Differentiating the first equation of the system once more, and eliminating M, we obtain

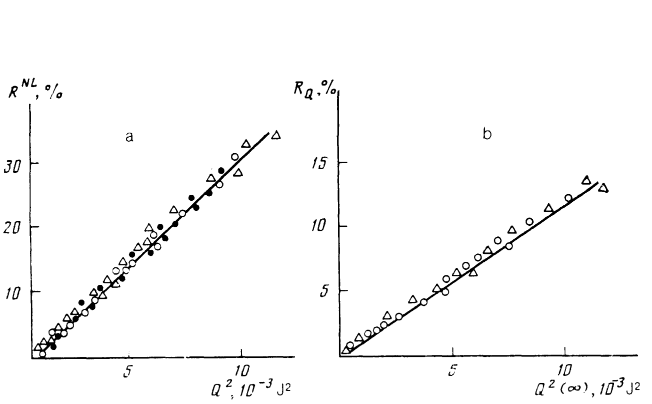

FIG. 2.

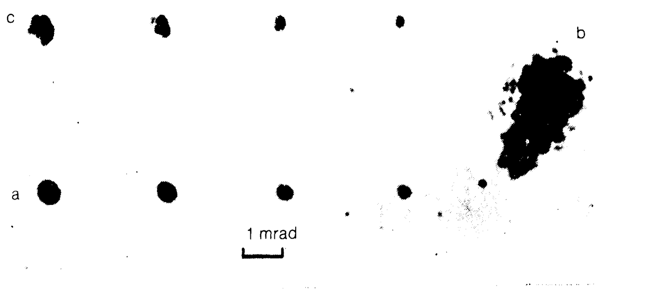

FIG. 3.

$$\frac{d^2\beta^*}{dy^2}+\frac{1}{y}\frac{d\beta^*}{dy}-\frac{i\beta^*}{y}=0. \tag{8}$$

This equation possesses the following solution:

$$\beta^*(y)=\beta_0^* J_0((-4iy)^{1/2}). \tag{9}$$

Here J_0 is a Bessel function of the first kind. The asymptotic form of this solution for large y is as follows:

$$|\beta(y)|^2=|\beta_0|^2\frac{1}{4\pi y^{1/2}}\exp(2^{3/2}y^{1/2}), \tag{10}$$

i.e., we have obtained an exponential spatiotemporal amplification of a weak signal: the so-called nonstationary stimulated orientational forward scattering of light from an e-wave into an o-wave. Below we shall neglect the small pre-exponential factor. Such stimulated scattering (SS) has been experimentally observed.[9] The stimulated forward scattering was observed in a planar 5CB-nematic sample of thickness 70 μm. The *SS* was excited by $\sim$800-μm pulse from a rubidium laser operating in the free-running regime. Estimates of the *SS* establishment time yield $\tau\approx 5$ msec, so that *SS* has an essentially nonstationary character.

Figure 4 shows the experimental setup for the observation of the *SS*. Rubidium-laser (L) radiation, whose energy was varied by filters *LF* within the range from 30 to 300 mJ, passed through an Iceland-spar wedge S_1, which separates from it a polarization component e_L corresponding to an e-type polarization for the liquid crystal (LC), and then through a diaphragm D_1. Next the radiation was focused by a lens L_1 ($f=5$ cm) on the cell C with the *LC*. The lens L_2 formed together with L_1 and C a telescopic system. The greater part of the transmitted radiation was focused on a Glan prism G, which separated from it a polarization component e_0, which corresponded to the *SS* signal, and was detected by a photocell F_2. Part of the radiation that passed through the cell C was reflected from a glass wedge, and passed through an Iceland-spar (S_2) wedge, which split it up into $\mathbf{E}_e$ and $\mathbf{E}_o$ components, and was photographed on a film with the use of a double-optical step attenuator *SA*. This allowed us to define the angular divergence of the $\mathbf{E}_o$ and $\mathbf{E}_e$ waves as the ratio of the dimension of the spot on the film to the focal length of L_2. For the monitoring of the energy and shape of laser pulse, we used a standard (IMO-2) calorimeter K and a photocell F_1. From the latter we measured the $W_o(t)$ signal, and determined the running value of $Q(t)$ (with the aid of an integrating network).

It should be noted that, in the experiment, we used the single-mode—with respect to the transverse index—as well as the multimode, regime of the laser. All the energy measurements were carried out in the multimode regime, since in the single-mode regime the pulse has a microsecond spiked structure, and exact quantitative measurements are extremely difficult to carry out. But in the multimode regime, by choosing the appropriate resonator geometry, we obtained a pulse with an almost smooth temporal envelope. The results of the measurements of the time dependence of the *SS* signal are shown in Fig. 5 in the form of a plot of $\ln(W_o/W_e)$ against the running value of $Q^{1/2}(t)$. The obvious linearity of this dependence indicates a good functional correspondence of the results to the formula (10). For the purpose of carrying out a quantitative comparison of the coefficient of proportionality of the indicated dependence with the theoretical value, we measured the transverse dimension of the beam in the crystal: $a(\text{FWHM})\approx 90\,\mu$m. We found the experimental value of the coefficient of proportionality to be 1.2 times smaller than the theoretical value computed from the expression

$$\frac{400\varepsilon_a}{a}\left(\frac{L}{3cn_\parallel n_\perp\lambda\eta}\right)^{1/2}\ [\text{mJ}^{-1/2}],$$

which also indicates good agreement between theory and experiment within the limits of the experimental error. The experimentally measured spontaneous-noise level $v\approx 5\times 10^{-3}$. The theoretical estimate of this quantity from the well-known formulas for the cross section for spontaneous scattering in the nematics (see Ref. 10) with allowance for the angular aperture $\theta_{\text{rec}}\approx 1.5\times 10^{-1}$ rad of the recording system yields (in the single-constant approximation)

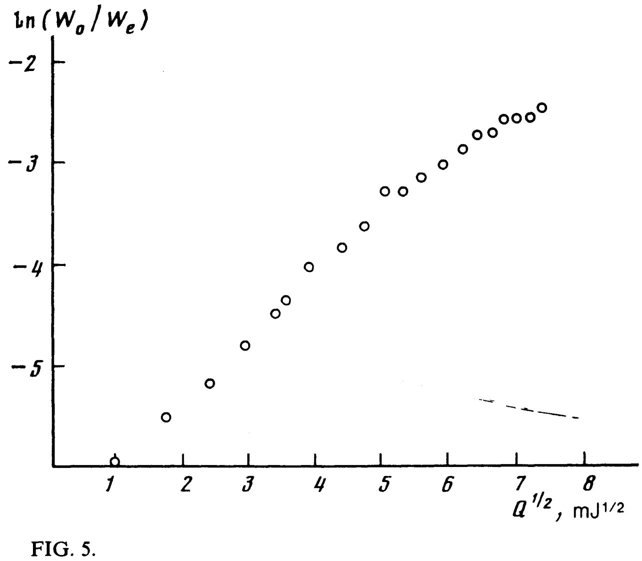

FIG. 5.

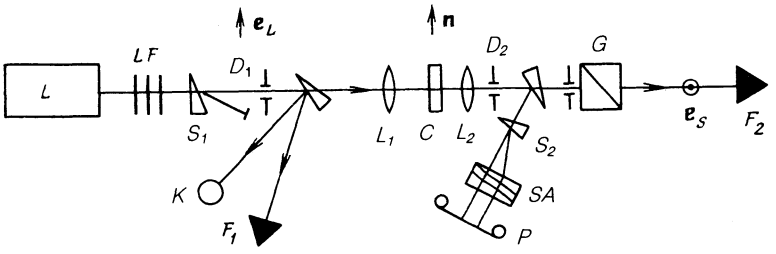

FIG. 4.

$$\nu = \frac{\pi\Delta n^2(n_\parallel + n_\perp)^2 k_B TL}{8\lambda^2 K_1} \int_0^{(\theta_{rec}/\Delta n)^2} \frac{d\xi}{1+\xi}$$

$$\approx 0.19\theta^2_{rec} \approx 4.2\cdot 10^{-3}. \qquad (11)$$

Here k_B is the Boltzmann constant and T is the absolute temperature ($\sim$300 K). The good agreement between the theoretical results and the experimental data can also be seen from (11).

Let us now turn to the measurements of the angular spectra of the *SS* signals and the pump. It should be noted that the longitudinal and transverse dimensions of the region of interaction in our experiment are comparable, so that not only collinear *SS*, but also *SS* at quite appreciable angles ($\sim$0.5 rad) is geometrically admissible. On the other hand, we see that the gain factor does not depend on the angle of convergence of the $\mathbf{E}_o$ and $\mathbf{E}_e$ waves. Thus, there is every reason to believe that the angular spectrum of the *SS* substantially exceeds the angular divergence of the pump. To verify this experimentally, we used single-mode radiation with FWHM divergence (after L_1) $\approx 5\times 10^{-2}$ rad. In this case the experimentally measured width of the angular spectrum of the *SS* signal exceeded the angular aperture of the recording system ($\sim$0.2 rad), and the angular spectrum had a strongly pronounced speckle structure that was not reproducible in the various realizations, which was to be expected in the case of nonstationary *SS* (the pulse time was much shorter than the time characterizing the irregularity of the phase of the spontaneous fluctuations).

Thus, we have experimentally detected nonstationary *SS* at an extremely small value of the interaction length, $L = 70$ μm, and a fairly moderate energy density in the pump radiation $100 \lesssim Q(\infty) \lesssim 300$ J/cm^2. In accordance with theory, the *SS* in question turned out to be dependent on only the self-similar variable y.

NONSTATIONARY ENERGY EXCHANGE BETWEEN WAVES WITH ARBITRARY INTENSITY RATIO

Finally, let us turn to the general case of the system (4), i.e., to the case in which the relation between α_0 and β_0 is arbitrary and y is arbitrary. But first let us make two comments. First, in view of the complexity of the solution to the system (4) in the general case, we limit ourselves to the consideration of the case of collinear interaction of two plane waves. Second, the attainment of high y values through the LON mechanism requires fairly high intensities (in the range from $\sim$100 to 500 kW/cm^2), so that the heating of the medium by the radiation is entirely probable, which, in view of the large $\partial n_{\parallel,\perp}/\partial T$ of the nematics [$\partial n/\partial T \approx (3-10)\times 10^{-4}$ deg^{-1}], will lead to the thermal modulation of the phases of the waves. The latter can have a considerable effect on the nonstationary energy exchange.

Fortunately, for characteristic radiation pulse lengths $\tau_p \approx 1$ msec and sample thicknessses $L \approx 100$ μm, the time characterizing the drain of heat into the substrates is $\sim$10 msec. Therefore, the heating of the medium during the action of a pulse is also of a cumulative nature, and does not destroy the self-similar nature of the system. Let the medium possess a small, polarization-independent (say, impurity) absorption coefficient $\varkappa$ cm^{-1}. It will subsequently become clear that it is precisely the case of small $\varkappa$ ($\sim 10^{-2}$ cm^{-1}) that is of interest to us. Then the system (4) with allowance for the thermal modulation of the phase assumes the form

$$\frac{d\alpha}{dy} = iM\beta + i\gamma_\parallel\alpha, \quad \frac{d\beta}{dy} = iM^*\alpha + i\gamma_\perp\beta, \quad \frac{dM}{dy} = \frac{1}{y}[\alpha\beta^* - M],$$

$$\alpha(0) = \alpha_0, \quad \beta(0) = \beta_0, \quad M(0) = \alpha_0\beta_0^*,$$

$$\left(\frac{dM}{dy}\right)_0 = \frac{i\alpha_0\beta_0^*}{2}[|\beta_0|^2 - |\alpha_0|^2 + \gamma_\parallel - \gamma_\perp]. \qquad (12)$$

Here

$$\gamma_{\parallel,\perp} = \frac{4n_\parallel n_\perp \varkappa c\eta}{\rho c_p \varepsilon_a^2}\left(\frac{\partial n_{\parallel,\perp}}{\partial T}\right)_p,$$

where c is the velocity of light, ρ is the density of the nematic, and c_p is the specific heat at constant pressure. Numerical estimates show[11] that $\gamma_\parallel < 0, \gamma_\perp > 0$, and $|\gamma_{\parallel,\perp}| \sim 1$ even for $\varkappa$ lying in the range from $\sim 10^{-3}$ to 10^{-2} cm^{-1}. Hence it is clear that such small absorption coefficients can have an appreciable effect on the energy exchange. Taking account of the smallness of $\varkappa$, we eliminated from the system (12) the terms describing the attenuation of the waves as a result of absorption, and this system possesses the integral $|\alpha|^2 + |\beta|^2 \equiv 1$. The system (12) describes the evolution of six real quantities. It, however, turns out that we can, by setting $\vartheta = |\beta|^2 - |\alpha|^2$ and $(C + iD)\alpha\beta^* = M$, eliminate from it the phases α and β (which are unimportant to us), obtaining as a result a system for the real quantities, ϑ, C, and D:

$$\frac{d\vartheta}{dy} = (1-\vartheta^2)D, \quad \frac{dC}{dy} = 2CD\vartheta + \frac{1-C}{y} + \gamma_a D,$$

$$\frac{dD}{dy} = \vartheta(D^2 - C^2) - \frac{D}{y} - \gamma_a C,$$

$$\vartheta(0) = \vartheta_0, \quad D(0) = 0, \; C(0) = 1, \quad \gamma_a = \gamma_\parallel - \gamma_\perp < 0. \qquad (13)$$

For $|\gamma_a| \ll 1$, this system possesses the equilibrium solution $D \equiv 0$, $C \equiv 1$, $\vartheta = -\gamma_a$, i.e., the energy exchange does not occur when $\vartheta_0 = -\gamma_a$. Let us investigate the stability of this equilibrium. For this purpose let us prescribe small deviations from it: $\vartheta = w - \gamma_a$, $D = O(w)$, $C = 1 + O(w^2)$. We easily obtain them from the linearized system (13)

$$\frac{d^2w}{dy^2} + \frac{1}{y}\frac{dw}{dy} + (1-\gamma_a^2)w = 0. \qquad (14)$$

This is a Bessel equation of zeroth order. Its solution is

$$w(y) = w_0 J_0[y(1-\gamma_a^2)^{1/2}]. \qquad (15)$$

Thus, we have obtained two qualitative results. First, the presence of absorption leads to directed transfer of energy from the *e*-wave into the *o*-wave through the LON mechanism (the equilibrium ratio of the intensities is different from unity). Second, as y increases, the normalized wave-intensity difference ϑ tends to its equilibrium value not monotonically, but in an oscillatory manner. This is also corroborated by the results of the numerical solution of (13) for the ϑ_0 values that differ greatly for $-\gamma_a$, for which values (14) is not valid.

Figure 6 shows a diagram of the experimental setup used to investigate the above-described nonstationary energy-exchange process. The exciting radiation was a $\sim$800-μsec single-mode—with respect to the transverse index—pulse from a rubidium laser operating in the free generation

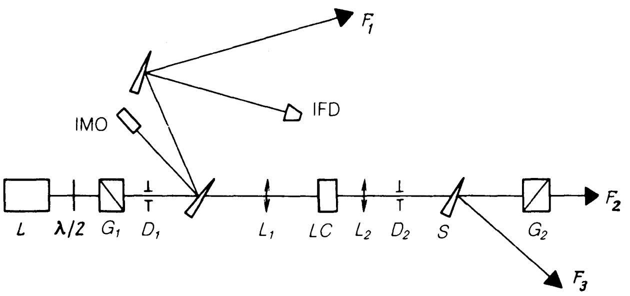

FIG. 6.

regime. A half-wave ($\lambda/2$) plate and a Glan prism G_1 were used to separate the required linear polarization from the incident beam (i.e., the required ϑ_0), and also to control the pulse energy. Since $\vartheta = \vartheta(y)$, to raise the accuracy of the quantitative measurements, a step which is necessary, in particular, for the observation of the small-amplitde ϑ oscillations about $\vartheta_{\rm equil} = -\gamma_a$, we must use a beam of constant intensity over its cross section. With this aim in view, a 5CB-nematic sample of thickness 70 μm was placed in the plane of a 22-fold diminished image of a diaphragm D_1 of diameter 1 mm, which separated out the beam's central part, over whose cross section the intensity was homogeneous. The exciting-pulse energy (in the sample) was varied from 0 to 30 mJ. After passing through the sample, the beam was split up by an Iceland-spar wedge S into its $\mathbf{E}_o$ and $\mathbf{E}_e$ components, the difference I_Δ between whose intensities was measured by photocells $F_{2,3}$ with the aid of the differential input section of an oscillograph. The temporal envelope I_Σ of the incident pulse and the running value of $Q(t)$ were also recorded.

The qualitative results of the experiment consisted in the following (see Fig. 7). For $\vartheta_0 < 0$ the oscillogram $I_\Delta(t)$ passes through zero (Fig. 7a). For the same pump-pulse energy, the same pulse shape $I_\Sigma(t)$ (Fig. 7c), but $\vartheta'_0 = -\vartheta_0$, $I_\Delta(t)$ does not pass through zero (Fig. 7b). This indicates the occurrence of directed energy transfer from the e-wave into the o-wave (i.e., the existence of $\gamma_a \neq 0$). To verify the self-similarity of the function $\vartheta(y) = I_\Delta / I_\Sigma$, we measured this dependence at a fixed value of ϑ_0 and different values of the total pulse energy, i.e., for different $I_\Sigma(t)$ (Fig. 8). Points of different shapes in the figure correspond to different $I_\Sigma(t)$. It can be seen that all the points fall on a single curve, which confirms the self-similarity of $\vartheta(y)$. It was found that, for $\vartheta_0 = 0.2$, the function $\vartheta(y)$ degenerates into a constant $\vartheta(y) \equiv \vartheta_0$, whence the experimental value $\gamma_a = -0.2$ was taken.

Figure 9 shows plots of the function $\vartheta(y)$ obtained through a numerical solution of the system (13) for $\gamma_a = -0.2$ and different values of ϑ_0, as well as the corresponding experimental points. The fit parameter for the scale along the y axis was the unknown—to us—viscosity η. It was found that the best agreement is attainable with the value $\eta = 1.35$ P, which is quite realistic for the nematics in the vicinity of their transition into the solid phase ($T \sim 20$ °C).

Unfortunately, it is not possible to carry out detailed measurements of the $\vartheta(y)$ oscillation about the equilibrium value. The point here is that the theory predicts oscillations in the region $y \gtrsim 4$, but in this region of y the intensity gain

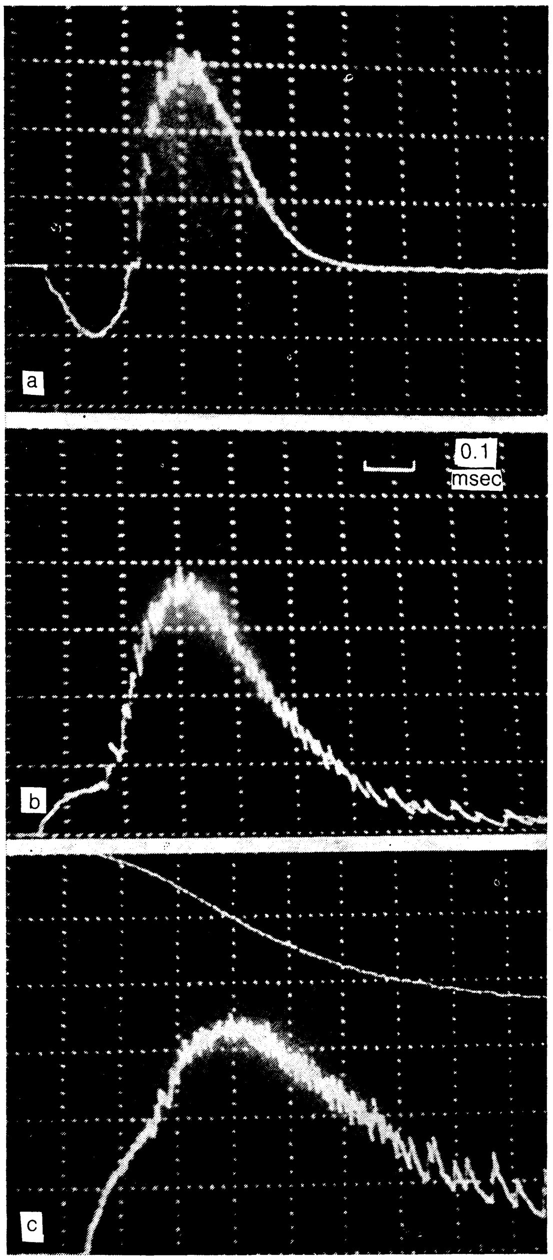

FIG. 7.

due to the SS is equal to $\exp(2^{3/2} y^{1/2}) \gtrsim e^5$ (see the preceding section), and both waves undergo substantial attenuation as a result of the SS, which exhibits the inadequacy of the theoretical model. But the experimentally observed intersection

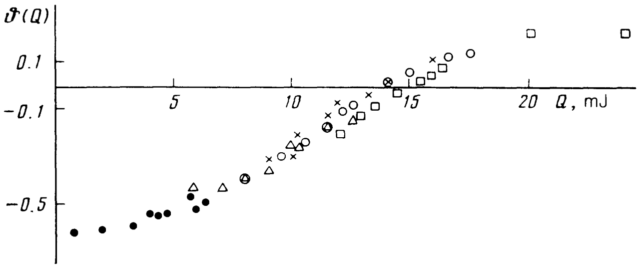

FIG. 8.

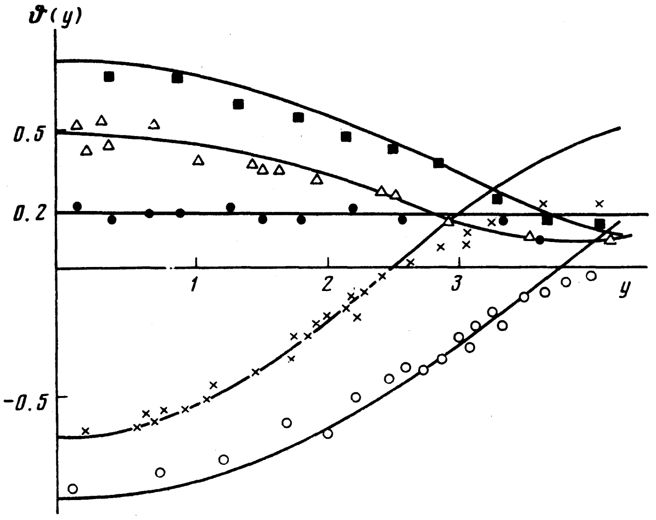

FIG. 9.

of the line $\vartheta = 0.2$ by the $\vartheta(y)$ curves does occur in the model, which indicates the qualitative validity of the theoretical predictions.

Using the value $\gamma_a = -0.2$ and the data in Ref. 11, we obtain the estimate $\varkappa \approx 2\times 10^{-3}$ cm^{-1}. Such insignificant coefficients of impurity absorption in the nematics cannot be measured by any direct method.

DISCUSSION

Thus, in the present investigation we have demonstrated experimentally that, although the orientation gratings in the nematics have long relaxation times, they can be effectively excited by short (~ 1 msec) light pulses. It has been found that the orientational optical nonlinearity of the nematics under nonstationary conditions is, in contrast to the effect under steady-state conditions, of a local nature, and this allows us to effectively apply it in the problems of dynamical holography, especially the LON, which excludes the self-focusing of the reference waves, and allows us to record three-dimensional holograms in a nematic. We have theoretically demonstrated the self-similarity of the process of nonstationary excitation of LON, i.e., the dependence of all the parameters of the process on the single space–time variable y. This self-similarity has been experimentally confirmed.

And finally, it turned out that even extremely weak ($\varkappa \sim 10^{-3}$ cm^{-1}) absorption in the nematics can have an appreciable effect on the energy exchange in the case of nonstationary self-diffraction through LON, giving rise to directed energy transfer from the e-wave into the o-wave. Thus, the indicated self-diffraction is an effective tool for the measurement of such small absorption coefficients in a nematic.

All the foregoing allows us to look forward to the practical applications of the nonstationary LON in the problems of dynamical holography and WFI.

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