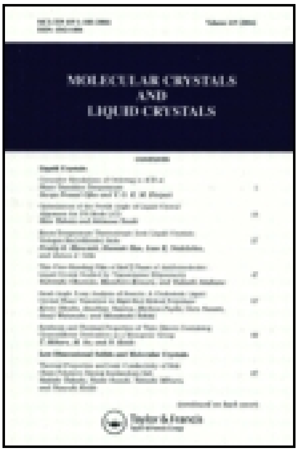

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Wavefront Conjugation in Dye-Activated Nematics in the field of Pulsed Ruby Laser

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WAVEFRONT CONJUGATION IN DYE-ACTIVATED NEMATICS IN THE FIELD OF PULSED RUBY LASER

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Abstract Wavefront conjugation (WFC) of free-running ruby laser by using thermal nonlinearity in dye-activated nematics is demonstrated. Obtained values of the conjugation coefficient R near phase transition to isotropic state are close to 100 %. Temperature dependence of R has been studied theoretically and experimentally.

INTRODUCTION

In our previous works[1,2] we have been the first to study static and dynamic gratings on thermal nonlinearity in oriented nematic liquid crystal layers (NLC) (20-40 μm) activated with dyes in the field of ruby laser radiation of nanosecond and microsecond pulse duration. Effective recording of dynamic gratings (diffraction efficiency in the first order of about 10%[2]) in absorbing NLC's makes it possible to perform WFC of pulsed radiation of a ruby laser. WFC has been initially demonstrated on thermal nonlinearity in NLC by Khoo[3] who used c.w. radiation of an Ar^+ laser.

This paper presents the results of investigations of effective WFC of free-running ruby laser radiation based on four-wave mixing (FWM) in dye-activated nematics.

THEORETICAL ESTIMATES

Let us consider two counterpropagating plane pump waves E_1, E_2 in a planar-oriented nematic layer (see Fig. 1). A weak signal wave E_3 falls on the medium from the left side causing generation of a phase-conjugated wave E_4 that propagates in the opposite direction. Two types of gratings can arise in the medium. The reflecting grating is formed by the signal wave E_3 and the pump wave E_2. The second pump wave is diffracted on that grating to form the wave E_4. Another possibility for the signal wave E_3 to form grating with the pump E_1 corresponds to the transmission FWM geometry.

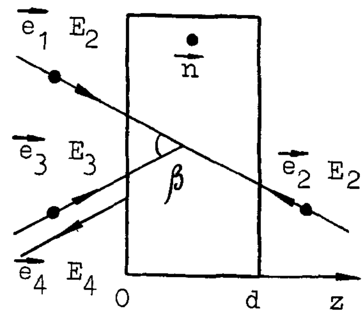

FIGURE 1

Dielectric permittivity of optical uniaxial crystals, where most nematic LC's belong, is described by a tensor $\varepsilon_{ij} = \varepsilon_{\perp}\delta_{ij} + \varepsilon_a n_i n_j$, where $\varepsilon_{\perp}$ is the permittivity of the ordinary wave, $\varepsilon_a = \varepsilon_{\perp} - \varepsilon_{II}$ is the permittivity anisotropy and n is the NLC director. Permittivity changes arising under the action of radiation in NLC's are associated with temperature dependence of the order parameter S as well as with changes in density ρ and local orientation of the director n. According to Sadovskij [4], for small deviations of LC hydrodynamics parameters: temperature δT, density $\delta\rho$ and director δn from their unperturbed values, permittivity changes can be described as follows:

$$\delta\varepsilon_{ij} = \frac{\varepsilon_a(n_in_j - \delta_{ij}/3)}{S}\frac{\partial S}{\partial T}\delta T + \frac{(\varepsilon_\perp - 1)\delta_{ij} + \varepsilon_a n_i n_j}{\rho}\delta\rho + \\ + \varepsilon_a(n_i\delta n_j + n_j\delta n_i) \qquad (1)$$

In deriving Eq. (1), we use only linear terms in expanding $\delta\varepsilon(\delta T, \delta\rho, \delta n)$ taking no account of the local field. In addition, Eq. 1 is applicable to processes where the parameter S has time to relax to the local equilibrium value. The relaxation time for S normally varies between 10^{-8}s far from the phase transition point and 10^{-6}s near the phase transition. The quantities S, $\delta S/\delta T$, ε_a are strongly dependent on temperature, and this dependence is usually measured experimentally. Deviations in δT, $\delta\rho$, δn obey Ericksen-Leslie equations for NLC hydrodynamics[5] which represent a set of coupled equations for hydrodynamic flow velocity v, density ρ, temperature T and director n that is very hard to solve in the general case. As it is known, in the case of NLC, the effect of electric field of a light wave on the medium is mainly assocated with light absorption[2,3] and orientational action of the field on the director[6,9]. For the geometry shown in Fig. 1 $(ne_i) = 1$, where e_i are polarization vectors of the mixing waves. Hence, in this case there is no orientational action of the electric field on the director. Solution to the linearized set of Ericksen-Leslie equations for small deviations in δT, $\delta\rho$, δn and the flow velocity v that arise under thermal action of the interference field of two waves on oriented NLC layer has been found by Sadovskij[4]. According to that solution, the main contribution to $\delta\varepsilon_{ij}$ modulation in Eq.(1) is made by the first term. Therefore, from here on we shall restrict our consideration to changes in the medium temperature as a result of absorption.

Let us conceive the field in the medium as a sum of four waves of the same frequency ω

$$E = 0.5\left(\sum_{j=1}^{4} e_j E_j \exp(-\alpha_j z/2) \exp(i\omega t - ik_j r) + c.c.\right),$$

assuming that $k_1 + k_2 = 0$ and $k_3 + k_4 = 0$. Here, E_j is the amplitudes of the waves and k_j is the wave vectors, $\alpha_i = (e_i \hat{\alpha} e_j) \cos(\beta/2)$ is the absorption coefficient, $\hat{\alpha} = \alpha_{\perp}\delta_{ij} + \alpha_a n_i n_j$ is the absorption tensor. $\alpha_a = \alpha_{\perp} - \alpha_{II}$ is the absorption anisotropy. The coefficients $\alpha_{\perp}$, α_{II} depend on the order parameter and, consequently, on the NLC temperature in the case of LC's intrinsic absorption or addition of dichroic dyes that acquire orientation with the same order parameter S in the nematic matrix. Following Blinov et. al.[7], the dependence of $\alpha_{\perp}$, α_{II} on S can be presented as $\alpha_{\perp} = \alpha_o(I-S)$, $\alpha_{II} = \alpha_o(I + 2S)$, where α_o is the LC absorption coefficient in isotropic state determined by the dye concentration. For simplicity, we shall use only linear approximation in calculating the conjugation coefficient, assuming that $E_{3,4} \ll E_{1,2}$. Then, the grating part of the heat source can be written as

$$\begin{aligned} Q &= (c\sqrt{\varepsilon_{II}}/4\pi)(E_1(0)E_3^*(0)\exp(-\alpha_i z)\exp(-iq_1 r) + \\ &+ E_2(d)E_3^*(0)\exp(-\alpha_i d/2)\exp(-iq_2 r) + c.c.) = \\ &= \tilde{Q}_1 \exp(-iq_1 r) + \tilde{Q}_2 \exp(-iq_2 r) + c.c. \end{aligned} \qquad (2)$$

where c is the light velocity, $q_1 = k_1 - k_3$, $q_2 = k_2 - k_3$ are the grating vectors.

For processes whose characteristic time is larger than sound attenuation time, heating of the matter proceeds at a constant pressure. For the grating heat source with the grating constant $q \gg \alpha_o$, heat conduction along the z-axis can be neglected, and it can be assumed that the temperature distribution in the medium follows the spatial pattern of the heat source $\delta T = \widetilde{\delta T}\ \exp(iqr)$, and in the interference pattern maxima $\widetilde{\delta T}$ is governed by the heat equation

$$\rho_0 C_p \dot{\widetilde{\delta T}} + (\varkappa_\perp + \varkappa_a \cos^2\vartheta) q^2\ \widetilde{\delta T} = \widetilde{Q}, \tag{3}$$

where C_p is the specific heat at a constant pressure, $\varkappa_a = \varkappa_\perp - \varkappa_{II}$, $\varkappa_\perp$, $\varkappa_{II}$ are the thermoconductivity constants, ϑ is the angle between the grating vector q and the director n. Solution of Eq.(3) takes the form:

$$\widetilde{\delta T}(t) = (\rho_o C_p)^{-1} \int_o^t Q(t')\exp((t-t')/\tau_T)\ dt', \tag{4}$$

where $\tau_T = \rho_o C_p/(\varkappa_\perp + \varkappa_a \cos^2\vartheta)q^2$ is the characteristic time of thermal conduction. For free-running laser pulses of width $\tau = 100 - 500\ \mu s$, $\tau > \tau_T$ if the beams convergence angle $\beta > 0.018$ rad., i.e. the heat conduction process can be regarded as stationary and solution of Eq.(4) is approximately equal to $\delta T(t) = (\rho_o C_p)^{-1} \tau_T Q$. For small β 2^o the amplitude for the trasmitting grating with wave vector q_1 sin $(\beta/2)$ is two order of magnitude higher than the amplitude δT for the reflecting gratings with q_2 $\cos(\beta/2)$. Hence, in our case the main contribution to the conjugated wave is made by the transmitting grating.

Nonlinear polarization P^{NL} that occurs as a result of scattering of the E_2 wave at the grating δT and generates the conjugated wave E_4 is

$$P^{NL} = \frac{\varepsilon_a (n(ne_2) - e_2/3)}{S} \frac{\partial S}{\partial T} \delta T \; E_2 . \qquad (5)$$

In the interaction geometry under consideration, the nonlinear polarization vector is directed along the director, and the conjugated wave is extraordinary. The equation for slow amplitude of the wave E_4 takes the form:

$$2 \; \partial E_4/\partial z - \alpha_{II} E_4 = -i4\pi k n_e^{-1} P^{NL}(z), \qquad (6)$$

where $n_e = \sqrt{\varepsilon_{II}}$. Solution of this equation results in the following expression for the conjugation coefficient R:

$$R = \frac{P_4(0)}{P_3(0)} = \left[\frac{\partial S}{\partial T} \frac{\varepsilon_a \lambda}{12 \; S \cos(\beta/2) \varkappa_{\perp} \; n_e^3 \; \sin^2(\beta/2)} \right]^2 * \qquad (7)$$

$$* \left| E_1(0) \right|^2 \left| E_2(0) \right|^2 \exp(-\alpha_{II} d) \; (1 - \exp(-\alpha_{II} d))^2 ,$$

where $P_i = c n_e E_i^2 / 8\pi$ is the power density of the i-th wave.

One can see that the dependence of R on temperature is mainly due to the dependence $\partial S/\partial T(T)$. Uniform heating leads to shifting of the effective temperature, at which conjugation takes place, to the phase transition point and can result in phase transition in the case of high densities.

For $\alpha_{II} d \ll 1$, $R \sim (\alpha_{II} d)^2$. Otherwise, $R \sim \exp(-\alpha_{II} d)$. If the pump wave E_2 is formed by reflection of the pump E_1 after its passage through the cell, then $R \sim \exp(-2\alpha_{II} d)$. Thus, the dependence R on $\alpha_{II} d$ has an optimum.

EXPERIMENTAL RESULTS

A single-mode ruby laser with pulse duration of 0.5 ms and pulse energy of 0.8 J was used as a radiation source. By using a set of mirrors, all three waves E_1, E_2, E_3 were formed from one source. The spot diameter on the specimen was 3 mm. The convergence angle β of the beams E_1 and E_3 was varied from 2^o to 10^o, which corresponded to the spatial period of gratings h = 20 - 4 μm. Intensity of the signal wave E_3 was six times as low as that of E_1. The reference wave E_2 was formed by reflecting the wave E_1 from the mirror. Because of absorption in the layer, the intensities of the waves E_1, E_2 were different and their ration was variable from 0.6 to 0.78. Planar NLC cells 50 μm and 100 μm thick placed into a thermostatic chamber were used. Experiments were conducted on 5CB (transition temperature $T_c = 35^oC$) and a five-component mixture of cyanobiphenyls ($T_c = 56^oC$) with addition of chrome green anthraquinone dye. The absorption coefficient of the dye in LC was varied from 50 cm^{-1} to 200 cm^{-1}.

The coefficient of reflection into conjugated wave R was measured experimentally from the ratio of maximum amplitudes of free-running laser pulse envelopes for the waves E_3 and E_4. We have studied the dependence of the conjugation coefficient on the convergence angle of the waves E_1 and E_3, pump intensities, initial temperature of the layer and absorption coefficient of the medium.

In accordance with Eq. (7), the conjugation coefficient R is a function of convergence angle of the waves E_1 and E_3. Comparison of the efficiencies of the conjugated wave for the angles of 2^o and 10^o shows that with the larger angle R decreases by an order of magnitude. This fact supports our assumption that under conditions of the experiment the heat conduction process can be considered as stationary.

Fig. 2 shows the dependence of R on the product of the pump wave energy densities P_1P_2 for 5CB (d = 50 cm^{-1}, α_o = 100 cm^{-1}, $\Delta T = T_c - T = 0.5^oC$). A linear dependence on the initial part of the curve agrees with Eq. (7). Further increase in the pump energies results in saturation that is connected with the pump depletion and the nematic-isotropic transition at high heating.

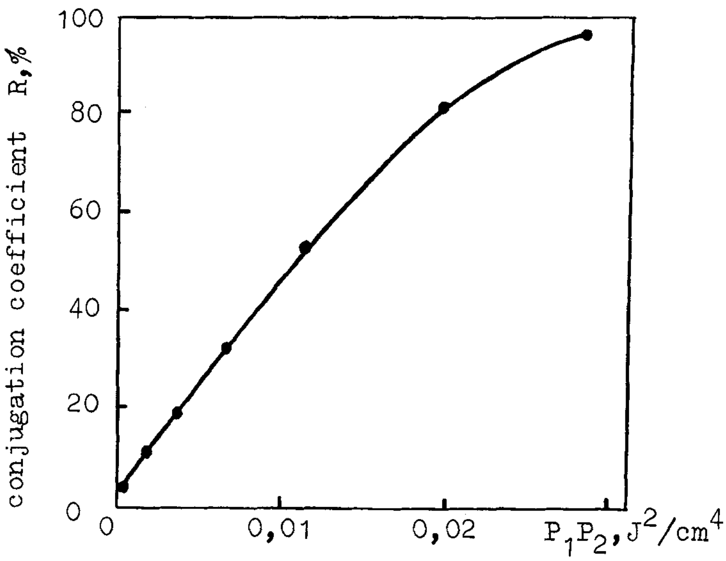

FIGURE 2

Investigation of the temperature dependence of the conjugation coefficient R have shown that in the temperature range $\Delta T = 15^oC$ to 5^oC lying far from the transition point the conjugation efficiency changes insignificantly (1 % to 5 %) (see Fig.3, curve 1 and 2). R sharply increases near the transition point, being as high as about 100% for $\Delta T = 0.3^{\circ}C$. The conjugated signal with $R \approx 13\%$ was also observed in the isotropic phase near the transition point. A similar behaviour of R for an LC phase has been observed by Khoo[3] who irradiated the cell with a c.w. Ar laser at 4 Hz frequency. Moreover, it agrees qualitatively with our formula (7) (curve 3, Fig.3). However, we

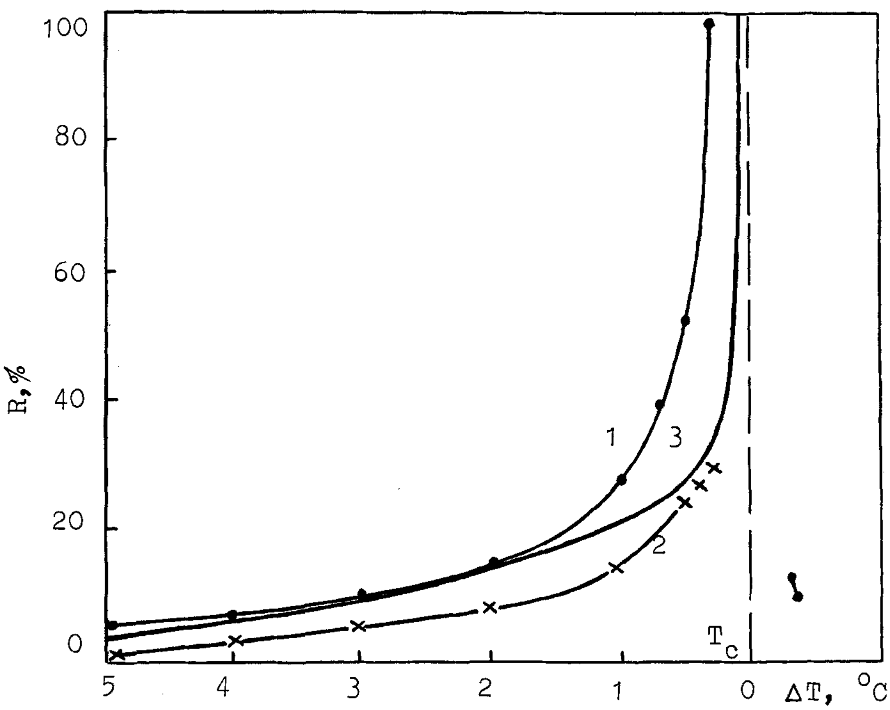

FIGURE 3 Temperature dependence of the conjugation coefficient for 5CB. (1) d = 50 μm, α = 100 cm^{-1}; (2) d = 50 μm, α = 50 cm^{-1}; P_1=0.2 J/cm^2 $\beta = 2^{\circ}$.

believe that initial temperatures of the nematic close to the transition point, laser heating of the LC layer is sufficient for its transition to isotropic liquid. While performing our previous work[8] we observed nematic-to-isotropic transitions stimulated by free-running laser radiation that could be detected by an increase in the layer transparency to a He-Ne laser probe beam. For oriented NLC layers 10-25 μm thick, the energy expenditures needed for transition were 10^{-2}-10^{-1} J/cm^2 while the time of transition was 20-30 μs. Estimates of the heat distribution in the interaction region taken for a 5CB cell with d=50 μm and $\alpha_o \cong 100cm^{-1}$ show that phase transition may occur already for radiation energies $P_1P_2 = 18 \cdot 10^{-3} J^2/cm^4$. As can be seen from Fig. 2, the tendency of the curve $R(P_1P_2)$ towards saturation appears at the same values of P_1P_2.

We have also studied the conjugation efficiency at various absorption coefficients. Experimental measurements indicate that there exists an optimum for this parameter at a given cell thickness. Thus, for the 5CB cell 50 μm thick and $\alpha_o = 100$ cm^{-1}, the curve 1 is characterized by the highest parameter R of the conjugated wave. Decreasing of α_o by a factor of two results in a three-fold decrease in R (curve 2), whereas no WFC is observed with α_o increasing two-fold. For the 100 μm-thick cell, WFC was about ~50% at $\alpha_o = 50$ cm^{-1}, and no conjugated wave was observed for α_o = $100cm^{-1}$. Hence, Eq. (7) gives a good description of the optimum in the R-vs-$\alpha_o d$ dependence. For higher values of $\alpha_o d$ the absence of the conjugated signal and, hence, disagreement with Eq. (7), is connected with the fact that phase transition point is reached sooner due to increasing of the uniform heating.

Thus, we have realized the WFC process based on the thermal nonlinearity in the field of free-running ruby laser radiation and obtained a qualitative agreement between our experimental data and the proposed linear theory of the WFC process in NLC. The results of investigation of WFC under the action of nanosecond laser pulses are expected to be published.

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REFERENCES

1. V. A. Pilipovich, A. A. Kovalev, G. L. Nekrasov, Yu. V. Razvin and S. V. Serak, Dokl Akademii Nauk BSSR, 22,36 (1978).
2. A. A. Kovalev, G. L. Nekrasov, V. A. Pilipovich, Yu. V. Razvin and S. V. Serak, Pisma v Zh.T.F., 5, 159 (1979).
3. I. C. Khoo, Appl. Phys. Lett., 47, 908 (1985).
4. V. N. Sadovskij and N. A. Usova, Akusticheskij Zhurnal, 33, 551 (1978).
5. F. M. Leslie, Arch. for Rat. Mech. and Anal., 28, 265 (1964).
6. B. Ya. Zeldovich and N. V. Tabiryan, U.F.N., 147, 633 (1985).
7. V. G. Rumyantsev, L. M. Blinov and V. A. Kizel, Kristallografiya, 18, 1101 (1973).
8. A. A. Kovalev, G. L. Nekrasov, Yu. V. Razvin and S. V. Serak, Dokl. Akademii Nauk BSSR, 22, 234 (1978).
9. I. Janossy, A. D. Lloyd, Mol. Cryst. Liq. Cryst, 203, 77, (1991)