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Wave Front Conjugation in Bleachable Dye Doped Liquid Crystals

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Wave-front conjugation (WFC) in four-wave interaction (FWI) of monopulse radiation of ruby laser in dye-doped liquid crystals (DDLC) was obtained. The amplification of conjugation wave was observed. The amplitude gratings and the phase gratings on thermal nonlinearity give the contribution in wave front conjugation.

Keywords: laser; wave front conjugation; liquid crystal; bleachable dye; saturation

INTRODUCTION

Wave front conjugation in LC was investigated intensively by many research groups^[1-4]. In paper^[5], we have considered peculiarities of WFC in a nematic LC activated with a linearly absorbing dye in the case of four-wave interaction of the radiation of a free-lasing ruby laser. It was shown that a high energy efficiency of conjugation can be attained on thermal nonlinearity in DDLC layers. The conjugation coefficient $R=P_4/P_3$, where P_4 , P_3 are the powers of conjugated and signal waves, respectively, was equal to 100% in that experiment. In order to study the capabilities of using a LC WFC-mirror to generate high-power light beams with a diffraction quality, it was supposed that investigation of WFC in nonlinearly absorbing LC media at four-wave interaction of nanosecond laser pulses is of great interest.

Experiment

Phase conjugation was studied on a unit schematically shown in Fig.1.a. Pumping was effected with a monopulse ruby laser beam 1 used to form the wave E_1 and, after reflection from a mirror 2, the wave E_2 . After passing through a limiting diaphragm 3 of 3 mm in diameter the pulse energy was 0.2J, its duration was 80 ns and its divergence was about 1 mrad. The signal wave E_3 was formed by mirrors 4,5. The angle between the E_3 direction and the pump

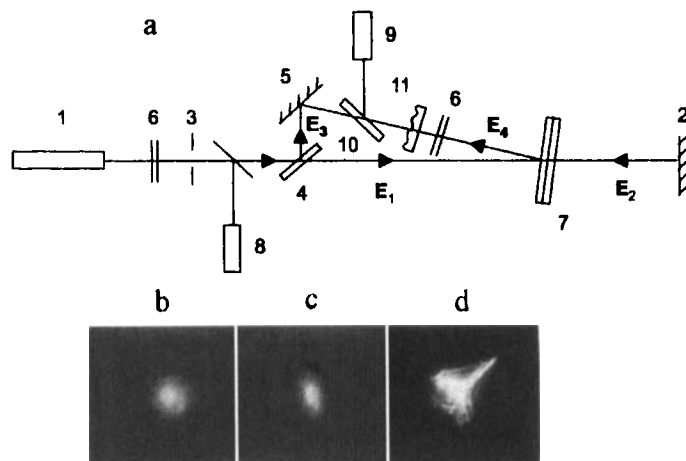


FIGURE 1 The scheme of experiment of WFC at FWI (a) and photos of the illumination distribution in initial conjugated beam (b), distorted conjugated beam (c) and ordinary beam that passed through in homogeneous medium (d).

beam was 10^0 (period of grating $\Lambda = 8 \mu\text{m}$), the signal wave intensity was eight times less than that of the E_1 wave. Energy characteristics of all interacting waves were varied by neutral filters 6.

Solutions of the following bleachable dyes have been studied: VO-phthalocyanine (with *t*-butyl substituent) (VOPhc) and 4,11-di(phenyl-3', 4') bisantene (DPB) in five-component mixture of cyanobiphenils (CB-5) (isotropic phase transition temperature $T_c = 56^\circ\text{C}$). DPB is characterized by linear

absorption oscillator and, consequently, high absorption dichroism in LC ($d=0.7$), its ordering degree is $S_{DBP}=S_{CB-S}=0.63$. VOPhc-dye exhibits low dichroism with $d=0.12$. These LC-solutions were earlier successfully employed for passive Q-switching of ruby laser cavity^[6]. The experiment was carried on with planar-oriented layers whose director L was parallel to the polarization vectors of interacting waves, i.e. e-waves were propagating in the layer. The orientational coating from silicon monooxide was used. LC-cell thickness was $d = 50\mu\text{m}$, and the absorption coefficient for the parallel component $k_{||}$ ($L \parallel E_1, E_2, E_3$) was varied in the range of $80\text{-}120\text{cm}^{-1}$. The LC cell 7 was placed in a thermostatted chamber; temperature gradient in the interaction region was not higher than 0.05°C . Energy efficiency of conjugation was measured by means of photodetectors 8, 9, the signals in channels E_1 and E_3 were equalized in amplitude in the initial state.

Results and discussion

At the moment of laser pulse action, two dynamic holograms (reflection and transmission) are forming in the nonlinear layer as a result of mutual coherence of all three incident waves. Several orders of self-diffraction of the E_1 and E_3 beams on a transmitting grating are easily observable on a screen placed behind the LC cell. The conjugated beam was observed on a screen at a distance of about 1m from the semireflecting mirror 10. Examination of the far-field zone of the conjugated beam showed that irrespective of the presence of the phase-inhomogeneous medium 11 that increases 8-fold the divergence of the signal beam, the conjugated beam did not change its distribution over its cross-section. In this situation, the conjugation coefficient did not decrease. Photographs of the illumination distribution in initial conjugated beam, distorted conjugated beam and ordinary beam passed through inhomogeneous medium are shown in Fig.1 (b,c,d), respectively.

Interaction of the waves E_1 and E_3 results in formation of a grating in the phototropic LC layer due to direct absorption of the exciting radiation energy. Initial change of the complex dielectric permittivity $\Delta\epsilon$ occurs as a result of redistribution of populations of active molecules of the dye. Within the excited state life-time, part of the absorbed energy is thermalized and causes the

disturbance associated with density modulation and change of the order parameter S . Disordering of long LC molecules under heating proceeds very fast, in 10^{-8} s for pulses with power of about 10^7W/cm^2 ^[7]. Investigation of the kinetics of interaction between monopulse ruby laser radiation and bleachable LC media^[7] has demonstrated that the threshold intensity at which thermal processes start to emerge during the pulse action is about $5 \cdot 10^6 \text{W/cm}^2$ ($\Delta T = T_c - T = 30^\circ \text{C}$), which is approximately an order of magnitude lower than its value for isotropic saturating media. At radiation densities over 10^7W/cm^2 , thermal processes start to manifest themselves at the monopulse leading edge. Increasing of the initial temperature of the layer results in the lowering of the corresponding threshold intensity values^[8]. For the DPB in CB-5, these values are equal to 10^6W/cm^2 and $5 \cdot 10^6 \text{W/cm}^2$ for $\Delta T = 2^\circ \text{C}$ whereas near the transition ($\Delta T = 0.5^\circ \text{C}$) bleaching of the layer starts to increase owing to phase transition.

On the other hand the molecular reorientation was revealed in the layers of VOPhc and DPB in CB-5 at the considering geometry $L \parallel E_{1,3}$. The processes that are able to change the layer orientation during pulse action are density change and rotation of excited molecules of dye in optical field^[9]. In pure CB-5 the reorientation at intensities used here was not observed. Reorientation mechanism is connected with dye properties. In work^[9] it was showed that the change of diffraction efficiency is in accordance with dichroism of absorption of dyes. Therefore the principal cause of reorientation of director here is the change of density as a result of thermal microvolume expansion (micro-boiling is observed in isotropic volatile solvents^[10]) and generation of hydrodynamic flow. About it is reported authors^[11]. The time required for the hydrodynamic velocity to reach the steady state $\tau = \rho \Lambda^2 / 2 \pi^2 \eta$, where ρ is the density and η is the flow viscosity (10^{-2} kg/m s) is equal 320ns. This time is longer then the duration of the monopulse.

Let us now turn our attention to the results. Figs.2 (a,b) shows the dependences of the conjugation coefficients R on the monopulse energy density at different initial temperatures of the layer. The variation of the nonlinear absorption coefficients in relative units is shown also. As the excitation energy increases the conjugation coefficient rises nonlinearly. In the range of energy values from 0 to $0.16 \text{J}^2/\text{cm}^4$, R changes insignificantly (up to 1%) both for

VOPhc and DPB in LC. By comparing these results with the results of studying bleaching kinetics^[7,8], it can be stated that in this case the WFC is caused by saturation of the dye absorption. In this range of W values, the absorption coefficient falls off. At high pumping levels, the solution saturates, and the transmission of the BPB and VOPhc-layer in LC increases by 30% and 40%, respectively. Various spectroscopic models of resonance medium have been used to perform theoretical and experimental studies of the WFC of nanosecond laser pulses for saturating isotropic dye solutions^[12,13]. According to the two-level model approximation, as the solution is saturated R should start to fall (see Fig. 2, $W^2 > 0.16 \text{ J}^2/\text{cm}^4$). It was found experimentally^[7] that in

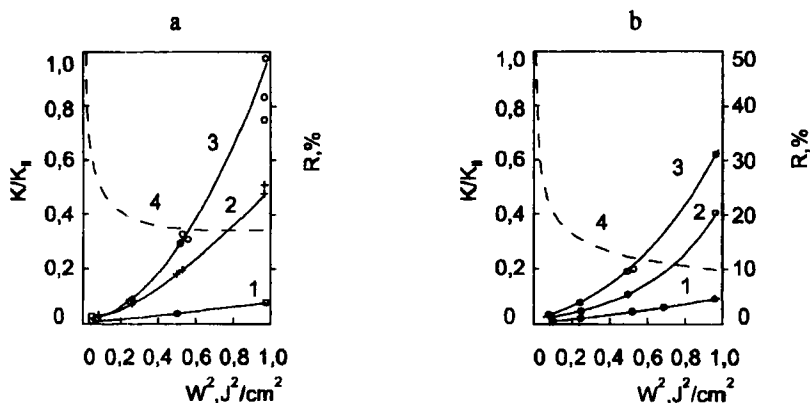


FIGURE 2 Conjugated wave reflection coefficient R versus monopulse energy for: DPB (a) and VOPhc (b) in CB-5 (1-3). Curve 4 corresponds to nonlinear absorption coefficient k . Curves 1,2,3 were obtained at $T=20^\circ\text{C}$, 54°C and 55.5°C , respectively.

the case of VOPhc saturation takes place in the channel s_1-s_2 while in the case of DPB the triplet level τ_1 is involved. As it is seen in Fig.2, for intensities $P > 5 \cdot 10^6 \text{ W}/\text{cm}^2$ ($0.16 \text{ J}^2/\text{cm}^4$) a fast characteristic rise in the energy reflection coefficient is observed for both VOPhc and DPB in LC.

For bleachable LC layers, resonance and thermal changes of the refractive index complement each other, resulting in overall increase of conjugated wave intensity. Changes in the layer order parameter have a considerable effect on R value. As the temperature approaches the phase transition point (Fig.3), where

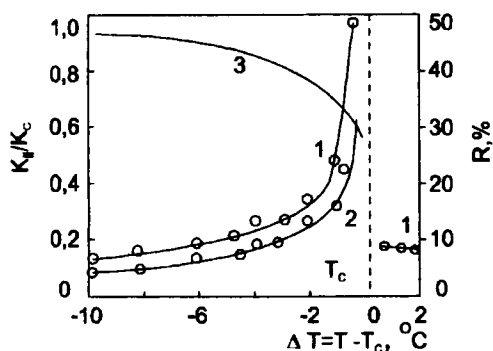


FIGURE 3 Temperature dependences of the reflection coefficient R for DPB (1), VOPhc in CB-5 (2), and absorption coefficient $k_{||}$ (3) for DPB in CB-5. $W=1\text{J}/\text{cm}^2$.

the absorption coefficient of dye near the phase transition depends very much on the temperature, that gives the amplitude grating in addition to the phase grating. Owing to a high absorption dichroism of DPB its transmission can decrease by 30% under heating from 20°C to 56°C . We suppose that the amplitude grating can be responsible for the bigger reflection coefficient for DPB dye in comparison with VOPhc near the phase transition (see Fig. 3).

We have studied the effect of signal wave intensity on the conjugation efficiency. Fig. 4 illustrates the dependence of R on the energy density of the E_3 wave. It is seen that the $W(R)$ dependence is inversely proportional, which testifies to occurrence of energy transfer in the E_4 channel. For the intensity ratio $W_3:W_1=1:90$, estimated amplification of the conjugated signal is as high as 130%.

the refractive index change for extraordinary wave is the largest, R increases significantly. After switching to isotropic state the reflection coefficient decreases. Comparison of $R(W)$ dependences at different temperatures shows that for dichroic molecules DPB at the same initial transmission of the layer efficiency of WFC is higher than for VOPhc. The

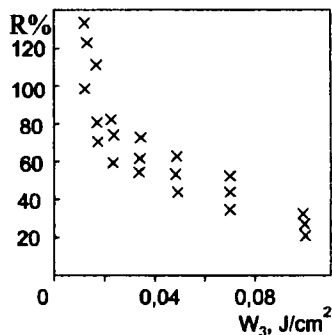


FIGURE 4 The change of conjugation efficiency on the energy of the E_3 wave. $T=20^\circ\text{C}$, $W=1\text{J}/\text{cm}^2$.

CONCLUSION

We obtained WFC at FWI in bleachable DDLC. The gratings on thermal nonlinearity give mean contribution in wave front conjugation. The amplification of conjugation wave was observed. The conjugation coefficient R was as high as 130%.

Acknowledgments

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