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RESONANT OPTICAL NONLINEARITY IN DYE-DOPED MESOGENIC MEDIA AND ABSORPTION SATURATION

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Abstract A detailed study of the transmittance of dye-doped LC-film in dependence on the excitation laser radiation intensity in different LC-phases (nematic and chiral liquid crystals) is presented. It should be underlined that these compounds are based on the same molecular basis (cyanobiphenyls and their derivatives). Transmittance of the investigated systems appeared to be independent on the light intensity in a wide power region. The observed effect could be associated with the dye re-absorption to a higher electron level with the "left" absorption equaling the fundamental one.

INTRODUCTION

It is well-known that fast electronic nonlinearity provides a variety of reach interesting nonlinear effects such as bistability[1], self-diffraction[2], etc. The main favorable features of such a type of nonlinearity are short switching time (few picoseconds)[3] and small writing energies ($<10^{-5}$ J). Sometimes rather similar in an exhibition to the temperature nonlinearity a resonant one needs some more efforts to distinguish it clearly.

EXPERIMENT

Quartz cells of 1.5 and 4.5 μm thickness were used for the investigations of the transmittance intensity dependence

of dye-doped LC. Cells were filled with planarly oriented nematic liquid crystal (NLC) or cholesteric liquid crystal (CLC) both based on the same cyanobiphenyl mixture with the relative chiral agent for the latter compound. They were doped with 1 % of the ketocyanine derivative dye. Absorption spectrum of the dye (1) and selective reflection band of CLC (2) are shown in the Fig.1 (NLC had no absorption in the visible spectral region). One can see that wavelength of the laser radiation falls in the overlapping of the dye absorption spectrum and the CLC Bragg selective reflection band (λ_B = 570 nm).

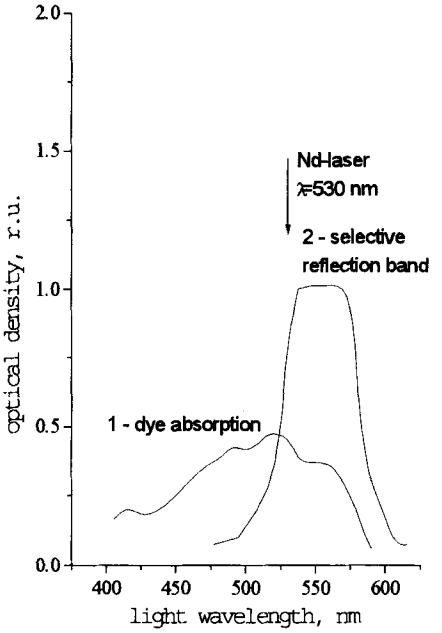


FIGURE 1 Spectral conditions of the experiment.

Experimental setup is sketched in Fig.2. Second harmonic of a Q-switched pulse Nd-laser radiation (λ_{ex} = 530 nm, t_p = 15 ns, w_p = 5 mJ) was used. The laser beam of 1.5 mm diameter attenuated while passing through the

filter set was focused at 0.6 mm spot on the LC-cell. Neutral filters were replaced subsequently from the first FS to the second one for the measuring system worked in the same energy range with varying light intensity from units to tens of thousands of kW/cm^2.

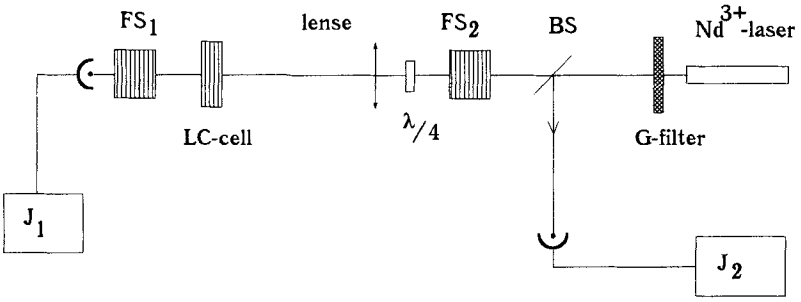


FIGURE 2 Experimental setup: Nd^{3+}-laser, G-filter - green colour filter, BS - beam splitter, FS_i - filter sets, λ/4-plate, lens (f = 20 cm), LC-cell - cell with cell-holder, J_i - microjoulmeters.

J_1 measured the transmitting light intensity while J_2 - 1/10 of the pumpimg energy. As the laser green beam was linearly polarized λ/4-plate determined the circular polarization of the definite sign of circularity in reference to that of the CLC-helix. The accuracy of the measuring devices was about few tens of a per cent, while estimated overall measurement error was less than 1%.

OBTAINED RESULTS

Cells with LC layer of 1.5 and 4.5 μm thickness were examined. The polarization of incident pumping light was circular with the sign of circularity both coincided and opposite to the CLC-helix twisting. The same polarization was used in the treatment of NLC compound to exclude the influence of the dye absorption dichroism. The light

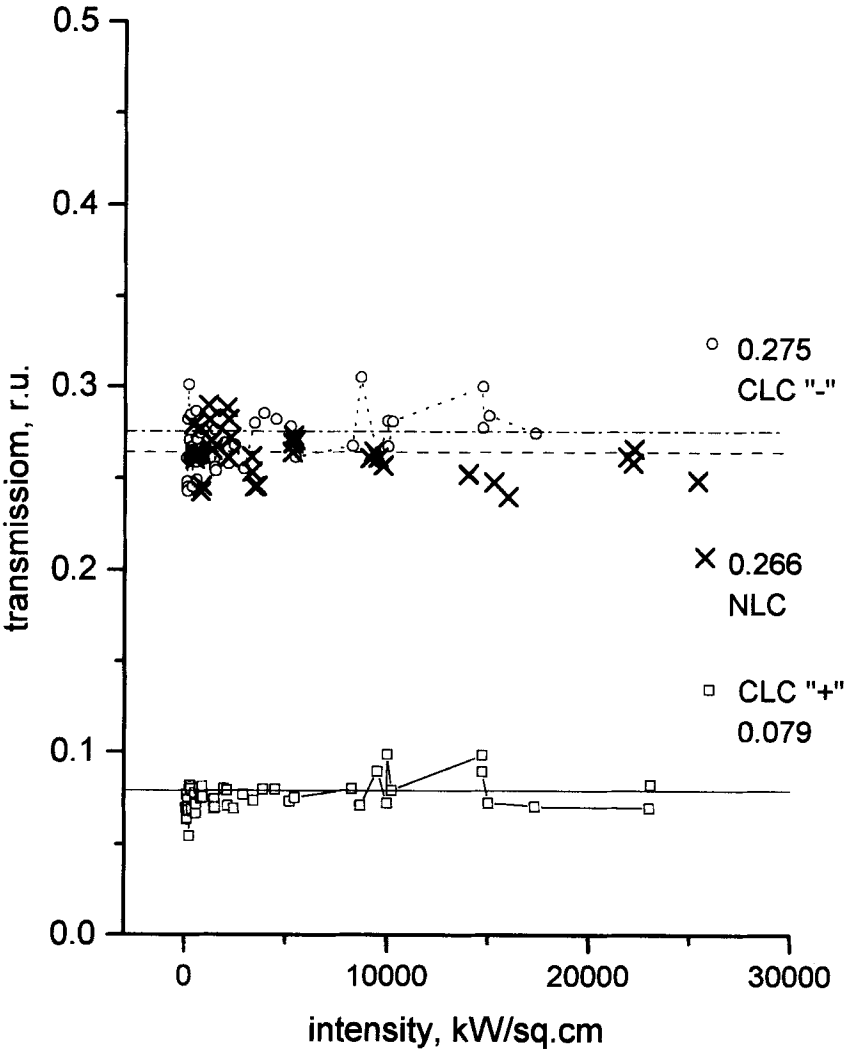


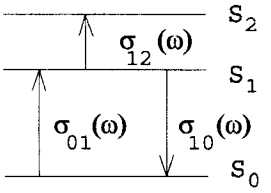
FIGURE 3 Light transmission vs pumping intensity. +/- corresponds to the circular light polarization coincided and opposite in sense to the chiral helix.

transmittance of 1.5 μm cells filled with either CLC or NLC mixtures is shown in the Fig.3. Transmittance of the investigated systems appeared to be independent on the

light intensity in a wide power region. The accuracy of obtained data is within 1 % for NLC and 2 % for CLC. The value of the transmission for the NLC cell almost coincided with that of CLC one with light polarization being opposite to the helix (without Bragg reflection) while selective reflection reduced the transmission for the opposite circular polarization.

CONCLUDING REMARKS

The obtained results are quite unexpected as a rather big content of a strongly absorbing dye often exhibits a saturation of the dopant excitation level under the intense laser pumping. In our experiments the dye absorption appears to be insaturable during the nanosecond laser excitation. It seems that transient processes influence on the absorption in our case.[4] Either singlet or triplet levels might be metastable in the dye molecule. As the quantum output of the phosphorescence is known to be very small in this system we ought to exclude triplet level transitions. So, we may assume that light absorption followed the sceme



including higher singlet level. $\sigma_{12}(\omega)-\sigma_{10}(\omega)\neq 0$ is the "left" absorption, were $\sigma_{ij}(\omega)$ - the probability of the light absorption on the frequency ω in the unit time. The constant in a wide intensity range absorption coefficient testifies that for our system the following condition is

valid: $\sigma_{12}(\omega) - \sigma_{20}(\omega) = \sigma_{01}(\omega)$, i.e., the "left" absorption equals the fundamental one.[4]

As concerning the bistability and self-diffraction phenomena observed in these systems they are both caused by the refractive index change due to a large number of dopant molecules excited to either the first or the second electron levels. Such a resonant optical nonlinearity is very similar to that caused by the usual dye absorption saturation and leads to the fast characteristic times of nonlinear effects too.

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