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Optical phase-conjugation in erioglaucine dye-doped thin film

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Abstract. Optical phase-conjugation (OPC) has been demonstrated in erioglaucine (acid blue 9) dye-doped gelatin films via continuous-wave degenerate four-wave mixing (DFWM) using a low-power He–Ne laser at 633 nm. DFWM and holographic processes are found to contribute to the observed phase-conjugate signal. A maximum phase-conjugate beam reflectivity of about 0.24% has been observed in these dye-doped gelatin films.

Keywords. Optical phase-conjugation; laser-induced gratings; dye-doped thin films; four-wave mixing.

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1. Introduction

Optical phase-conjugation (OPC) has potential applications in image transmission, optical image processing, optical filtering, and laser resonators [1–3]. Generation of PC wave through degenerate four-wave mixing (DFWM) has been investigated extensively in the past two decades in films or bulk media. OPC defines usually a special relationship between two coherent optical beams propagating in opposite directions with reversed wavefront and identical transverse amplitude distributions. The unique feature of a pair of phase-conjugate beams is that the aberration influence imposed on the forward (signal) beam passed through an inhomogeneous or disturbing medium can be automatically removed from the backward (phase-conjugated) beam passed through the same disturbing medium. Several technical approaches are there to efficiently produce the backward phase-conjugate beam. The first one is based on the degenerate four-wave mixing process, the second one is based on various backward stimulated scattering processes such as Brillouin, Raman, Raleigh-wing or Kerr and the third one is based on one-photon or multi-photon pumped backward stimulated emission (lasing) process. Among these three techniques, the backward DFWM geometry plays an important role in generating

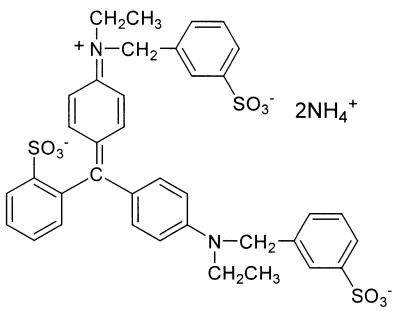

Figure 1. The general structure and formula of erioglaucine dye (molecular formula: $C_{37}H_{42}N_4O_9S_3$).

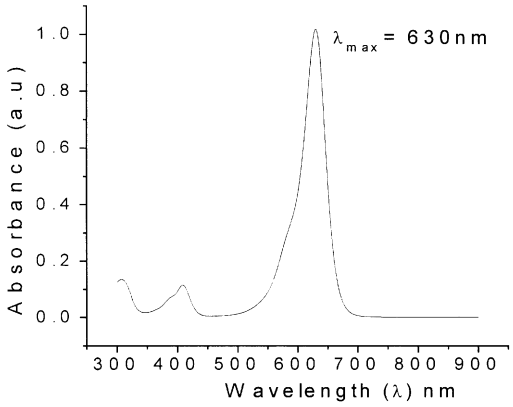

Figure 2. The UV–visible absorption spectra of erioglaucine dye.

phase-conjugate beam, since the pure electronic nonlinearity is assisted by an induced holographic grating in the process. OPC has been reported in many organic or inorganic materials using pulsed or continuous-wave (cw) lasers [4–18]. Glasses and other solid matrices doped organic dyes emerged as promising materials for OPC because of their large third-order nonlinearity $\chi^{(3)}$. In these materials, the phase-conjugate wave can be generated at low light intensities provided by the continuous-wave lasers. Moreover, these materials can be easily prepared in the laboratories.

In this paper, we present PC wave generation in triphenylmethane [19] dye-doped gelatin films using low-power continuous-wave laser excitation. The nonlinear medium (NM) used in this work is gelatin films doped with erioglaucine dye as the photosensitive chromophore.

2. Experimental

2.1 *Materials*

The organic dye erioglaucine (acid blue 9 – C.I.42090) belongs to the triphenylmethane groups. All the dyes of this series are derived from the hydrocarbon, triphenylmethane, and the tertiary alcohol, triphenylcarbinol, both of which are colorless. The chromophore of this class is the quinonoid group, which may appear as C=Ar=NH or C=Ar=O, where Ar = aromatic nucleus (color index 1975). The chemical structure and molecular formula of erioglaucine dye are shown in figure 1. The UV–visible absorption spectrum of erioglaucine dye was studied using UV-2401 PC spectrophotometer and it exhibits the peak absorption at 630 nm as in figure 2. The gelatin films are drawn from 10E75 Agfa Gaevert photographic plates by removing silver halide emulsions using sodium thiosulphate solution. The thicknesses of the films obtained were of the order of 10 microns. These plates were soaked in aqueous solutions of erioglaucine with appropriate dye concentrations for 2 min

and dried at room temperature. These films were used for this study without any further process. The optical density (OD) of the dye-doped film chosen for this work was approximately 1.

2.2 *Methods*

The schematic diagram of the phase conjugation experiment is shown in figure 3. A He–Ne laser (Coherent, 31-2140-000 – 35 mW) beam at 633 nm was divided into three beams, two counter-propagating pump beams E_1 and E_2 namely forward-pump and backward-pump beams respectively and a probe beam E_3 to form the DFWM configuration. The spot size of each of these three unfocussed beams at the nonlinear medium was 1.25 mm in diameter. The constant power ratio of the probe beam (E_3), forward-pump beam (E_1) and backward-pump beam (E_2) used in this work was $\sim 1:10:10$. The angle between the probe beam and the forward-pump beam was 7°. The sample was exposed simultaneously to all these three beams. The optical path lengths of all the three beams were made equal, so that they were coherent at the sample. The phase-conjugate wave retraces the path in the opposite direction to that of the probe beam E_3 and was detected with the help of a photodetector (Field Master^{TM} GS – Coherent Inc.). The experimental set-up was mounted on a vibration isolation table (Melles Griot-Metric version) to avoid the destruction of the laser-induced gratings formed in the erioglaucine dye-doped gelatin film due to mechanical disturbances.

3. Results and discussion

There are two cases to be considered for the mechanism of phase-conjugate wave generation associated with the erioglaucine dye-sensitized gelatin film. The first one is due to the third-order nonlinearity ($\chi^{(3)}$), and the second is due to absorption saturation and photobleaching which lead to the formation of a laser-induced grating in the medium.

We consider a three-level system for the erioglaucine dye in gelatin matrix. Figure 4 shows an energy level diagram of erioglaucine dye in which S_0 and S_1 are the ground and excited singlet states, respectively, and level T_1 is the triplet state. Absorption of a photon by erioglaucine results in transition of the erioglaucine to the first excited singlet state ($S_0 \rightarrow S_1$). If the singlet-to-triplet cross-over is considerable, the dye molecules will switch over to the triplet state ($S_1 \rightarrow T_1$), where it will remain for a relatively longer time as the triplet-to-singlet transition is inhibited, and, consequently, this molecule will not be available for further absorption from the ground state. This will result in a saturation of absorption if the triplet lifetime is long enough. Thus, in the medium, the absorption becomes a function of intensity. Therefore, when the two write beams interfere, the intensity pattern modulates the complex refractive index, which results in the formation of a grating [14]. The fringe period (Λ) can be determined by the well-known formula $\Lambda = \lambda/2\sin(\theta/2)$, where λ is the laser wavelength, and $\pm\theta$ is the forward-pump and probe beam incident angles with respect to the normal to the nonlinear medium.

Photobleaching of the dye molecules at the excitation wavelength also should be considered in this discussion. The existence of photobleaching can be inferred from

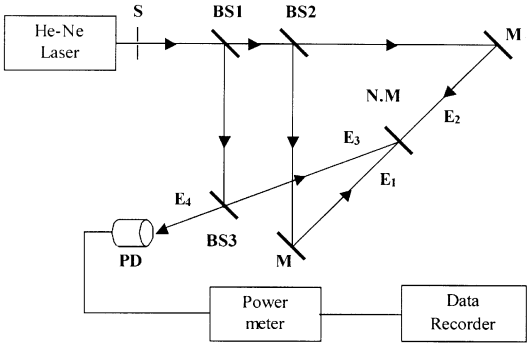

Figure 3. Experimental set-up for the observation of PC wave. S – shutter, BS1, BS2, BS3 – beam splitters, M – mirror, N.M – nonlinear medium, PD – photodetector.

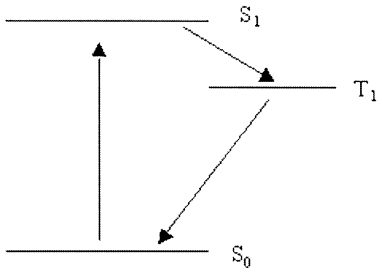

Figure 4. Schematic energy level diagram for erioglaucine dye saturable absorber.

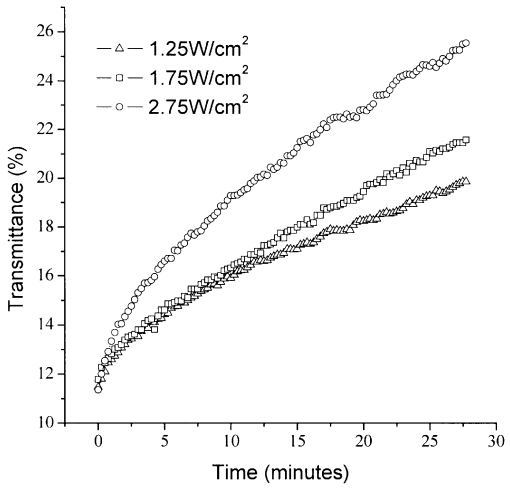

Figure 5. Measured transmittance of the erioglaucine dye-doped gelatin film as a function of time.

a simple experiment described as follows. Erioglaucine dye-doped gelatin film was illuminated with 633 nm radiations at three different incident intensities and the corresponding transmittance of the sample was measured with respect to time. We observed that the transmittance of the sample increases from 11.35 to 19.86%, 11.46 to 21.6%, and 11.72 to 25.57% in 28 min (see figure 5) corresponding to the incident light intensities of 1.25, 1.75 and 2.75 W/cm^2 respectively. The observed increase of transmission confirms the existence of a light-induced bleaching process associated with this dye-doped system. For lower intensities the bleaching may be reversible but when higher intensities are used it may result in complete decomposition of dye molecules and hence become irreversible, and such observations have been already reported [14].

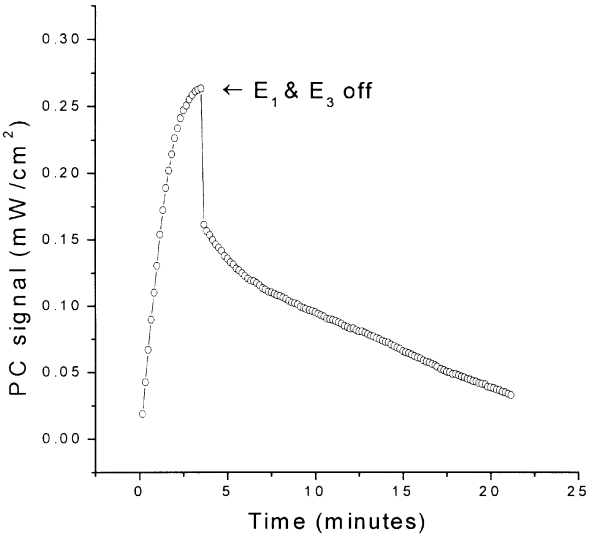

Figure 6. Measured PC signal as a function of recording time.

Dyes fixed in a solid matrix have the capability of generating a phase-conjugate wave by not only DFWM but also holographic process [20]. To distinguish the phase-conjugate wave generated by DFWM from that by the holographic process, the transient behavior of the PC signal was studied. For this, erioglaucine dye-sensitized gelatin film was first illuminated with three waves E_1, E_2 and E_3 for a specified duration, and afterwards, E_1 and E_3 were successively turned off, so that only E_2 was incident on the dye film. Here we call the duration for which all the three waves are incident on the dye film as the DFWM duration. Figure 6 shows the measured phase-conjugate signal as a function of time. The initial rise to a peak within a few minutes is due to DFWM and holographic processes; the sudden drop in the intensity of the PC signal after shutting off both the write beams E_1 and E_3 indicates the contribution from the fast DFWM process. Due to the holographic process the PC signal is present even after E_1 and E_3 are shut off, and it decays rather slowly. If the phase-conjugate wave was generated only by DFWM, the lack of only one of the three beams E_1, E_2 and E_3 would have stopped generation of the phase-conjugate wave. Therefore it is inferred that the rapidly decaying component corresponds to the phase-conjugate wave which is generated by the DFWM. On the other hand, if spatially modulated information formed by E_1 and E_3 can be recorded in the erioglaucine dye-sensitized gelatin film, the phase-conjugate wave can still be generated when E_2 tries to read this stored information, during the lifetime of the holographic grating.

4. Conclusions

To summarize, we have observed low-intensity optical phase-conjugation in erioglaucine dye-doped gelatin films using a degenerate four-wave mixing set-up,

employing 633 nm light radiation from a He–Ne laser. The mechanism of phase-conjugate wave generation associated with this dye-doped system is discussed. The phase-conjugate signal is found to have contributions from the DFWM and the holographic processes. The maximum phase-conjugate beam reflectivity observed in these dye films is about 0.24%. Since the erioglaucine dye-doped gelatin film is used at 633 nm and this may be suitable for low-power semiconductor lasers in the red wavelength region, erioglaucine dye-doped gelatin film may be a promising material for real-time double-exposure phase-conjugate interferometry.

Acknowledgement

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