Generation of phase-conjugate wave in acid blue 7 dye-doped gelatin film

T. Geethakrishnan and P. K. Palanisamy*

Centre for Laser Technology, Department of Physics, Anna University, Chennai 600 025, India

We present optical phase-conjugation based on the degenerate four-wave mixing (DFWM) arrangement in gelatin films doped with acid blue 7 dye. The 633 nm beam from a low-power He–Ne laser was split to form two counter-propagating pump beams and one probe beam in the DFWM geometry. Experiments were carried out by varying the parameters (angle of separation between the forward-pump and the probe beam, concentration of the dye in the gelatin film, writing beam intensities) that influence the phase-conjugate beam reflectivity. High reflectivity optical phase-conjugation is hereby reported for low-power lasers.

Keywords: Acid blue 7 dye, four-wave mixing, gelatin film, induced holographic gratings, optical phase-conjugation.

OVER the past several years, an exciting field in the area of coherent optics has been nonlinear optical phase-conjugation (NOPC)[1]. The field of OPC has stimulated a lot of research and development activities that cover both fundamental and applied parts of the field of laser optics. The important new aspects of OPC which are of prime interest are the following: OPC is a nonlinear mechanism that reverses both direction of propagation and phase of an aberated wavefront; generation of the conjugate beam can be viewed as a dynamic holographic recording process in a medium that exhibits third-order optical nonlinearity. Such an unconventional optical device is now known as a phase-conjugator or a nonlinear phase-conjugate mirror. The mechanism of dynamic holography and phase-conjugation has stimulated great interest in the research laboratories for nearly two decades. When the nonlinear optical material is illuminated by interference of a pump and probe beams, complex dielectric constant of the material is spatially modulated, which generates amplitude or a phase volume grating. The third-order effects occur even in isotropic transparent media, and there is no restriction regarding lack of inversion symmetry centre as for the second-order nonlinearities. In particular, the most established $\chi^{(3)}$ nonlinear mechanisms for OPC are Kerr effects, Brillouin scattering and Raman scattering. However, other effects that may also provide an efficient index or amplitude modulation through mechanisms with a response time ranging from several nanoseconds to seconds are of interest for OPC. Some potential applications[1] of NOPC include real-time adaptive optics, optical signal processing, optical computing, ultra-low noise detection and interferometry as well as nonlinear laser spectroscopy[2]. The analogy of phase-conjugation with dynamic holography was outlined by Yariv[3] and in early experiments with photorefractive crystals[4]. This contributed to extend the field of applications, thus including optical correlation for pattern recognition, holographic interferometry for non-destructive testing, incoherent to coherent image conversion, and novelty filters for moving object detection[3].

Organic dyes have emerged as materials with good potential for OPC even at relatively low light intensities because of their large third-order nonlinearity[5]. OPC has already been reported in several types of materials doped with organic dyes[6–19]. With the rapid growth of photonics technology, considerable effort is being directed towards the development of new materials. Better materials are still needed for implementation of ideas.

Here we demonstrate OPC effect in gelatin films doped with acid blue 7 dye (alphazurine A), which belongs to the triphenylmethane[20] group. The aim of this work is to characterize the details of OPC (PC reflectivity, kinetics of PC signal build-up and decay, its intensity dependence) and explain the results obtained.

Materials and film preparation

Synthetic dyes of the triphenylmethane group are either green, blue or violet in colour. Since 633 nm radiation from a He–Ne laser was used for this study; the red sensitive dye acid blue 7 (C.I.42080, alphazurine A) was chosen. Acid blue 7 is in the form of a dark bluish powder and is highly soluble in water, giving a dark bluish-green solution. The general structure and formula of the triphenylmethane dye acid blue 7 (alphazurine A) are shown in Figure 1 The optical absorption of the acid blue 7 dye in water with 0.01 mM concentration shows an absorption peak (λ_{max}) at 637 nm, as shown in the Figure 2.

In our experiment we used Agfa-Gaevert 10E75 holographic plates to prepare the gelatin plates. The silver halide grains of the photographic plates were removed by dipping them in sodium thiosulphate solution (fixer). After fixing, the plates were washed in running water and then dried in methanol. The thicknesses of the gelatin films

*For correspondence. (e-mail: profpkpannauniv@yahoo.co.in)

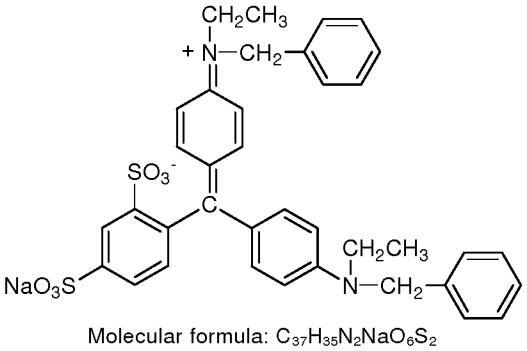

Figure 1. General formula and molecular structure of acid blue 7 dye.

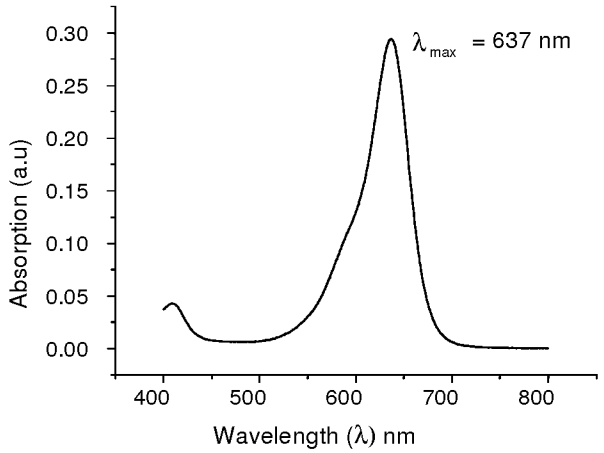

Figure 2. UV–VIS absorption spectrum of acid blue 7 dye in water.

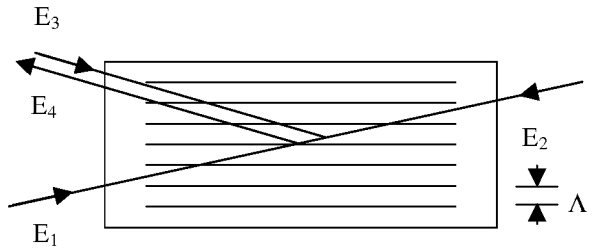

Figure 3. Schematic of phase-conjugate wave generation by degenerate four-wave mixing.

were of the order of 10 μm. Photosensitive plates at various optical densities (ODs) were obtained by soaking the gelatin plates in acid blue 7 dye solutions of different concentrations, keeping the soaking time constant. The plates were then dried at room temperature and used for study without any further processing.

Phase-conjugation via degenerate four-wave mixing

In the basic geometry for phase-conjugation, two counter-propagating plane waves called the pump beams of amplitudes E_1 (forward-pump) and E_2 (backward-pump) and a probe beam of amplitude E_3 pass through the nonlinear medium, as shown in Figure 3. In general, the beams have the same polarization state; the probe beam and the forward-pump beam meet at a small angle on the recording medium having third-order nonlinearity. A clear interpretation of conjugate-beam generation is shown in Figure 3. The probe beam E_3 and the forward-pump beam E_1 interfere in the nonlinear material and create a periodic interference pattern that modulates the physical properties of the nonlinear medium. The resulting grating wave vector amplitude is $k = 2\pi/\Lambda$. The fringe period (Λ) is given by the formula $\Lambda = \lambda/2\sin(\theta/2)$, where λ is the laser wavelength and $\pm\theta$ are the forward-pump and probe beam incident angles with respect to the normal to the nonlinear medium; it results in a complex volume hologram due to a spatial distribution of the refractive index (Kerr-like media), or of the absorption (saturable absorbers), or of the gain when the conjugator is the laser medium itself. The second antiparallel pump (backward-pump) beam E_2 is then diffracted under Bragg conditions by the dynamic volume hologram; following the classical formalism of holography, it generates a backward conjugate wavefront whose complex amplitude can be written as $E_4 = E_3^* E_1 E_2$. It is named as the conjugate image beam in holography. Since this geometry involves waves E_1–E_4, which are simultaneously present in the medium, it is known as four-wave mixing. The phase-conjugate reflectivity (R_{pc}) is defined as the ratio of the amplitude of the PC wave to the probe beam amplitude.

Experimental set-up for observation of PC signal

Figure 4 shows the experimental configuration used to realize OPC. In this case, the standard degenerate four-wave mixing (DFWM) configuration was used. A He–Ne laser

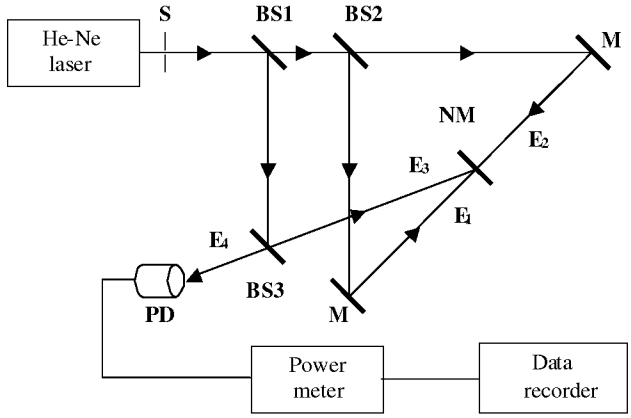

Figure 4. Experimental set-up for observation of PC wave. S, Shutter; BS1–BS3, Beam splitters; M, Mirror; NM, Nonlinear medium; PD, Photo-detector.

(Melles Griot 05-LHP-928) beam at 633 nm wavelength was used to generate the PC wave from the acid blue 7-doped gelatin films. The output beam from the laser was first split by a beam-splitter BS1 (~ 5 : 90). The beam reflected off from BS1 was used as the probe beam E_3 after reflected by the beam-splitter BS3; the transmitted beam from BS1 was further divided by another beam-splitter BS2 (50 : 50) to provide the counter-propagating pump beams, called forward-pump wave E_1 and backward-pump wave E_2 respectively. Beam-splitter BS3 was used to direct the probe beam to the dye-doped sample and to transmit the PC signal, which was opposite to the direction of the probe beam. The intensity of PC wave was measured by a photodetector fed to the digital power meter (Field Master^{TM} GS – Coherent). The constant intensity ratio of the probe beam (E_3), forward-pump beam (E_1), and backward-pump beam (E_2) used in this study was ~ 1 : 10 : 10. The spot size of each of the three (unfocussed) beams at the nonlinear medium was 1.25 mm in diameter. The incident angle (θ) between the probe beam and forward-pump beam was varied between 5 and 10°. The optical path lengths of all the three beams were made equal, so that they were coherent at the sample.

Results and discussion

Dye-doped gelatin plates of various optical densities were prepared by dipping the gelatin plates in the dye solution at different concentrations (1, 1.25, 1.5 mM, etc.) by maintaining the soaking time constant (2 min). Figure 5 shows the variation of optical densities of gelatin plates (at 633 nm) as a function of concentration of the dye solutions in which the plates were soaked. As expected, it was found that the OD of the gelatin plates increases as the concentration of the dye solution increases. However, beyond certain values of dye concentration, absorption of the dye by the gelatin plates gets saturated.

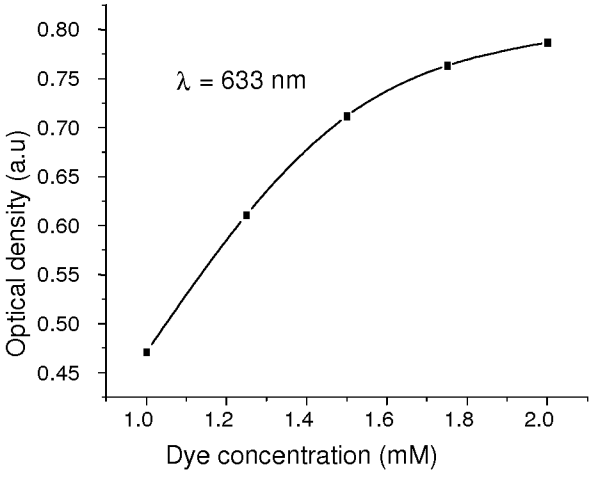

Figure 5. Optical density of gelatin films at 633 nm vs concentration of dye solutions in which plates were soaked for 2 min.

Figure 6 shows the PC reflectivities observed in acid blue 7 dye film as a function of the recording angle between the forward-pump and the probe beam. It is known that the PC signal disappears when the angle between the beams is large enough[21]. The highest PC reflectivity was observed at an angle of 7°. This is because of less overlap between probe and pump beams in the dye film and small grating periods[22].

Figure 7 shows the intensity of the PC signal as a function of time for acid blue 7 dye film with optical density of about 0.7. It is inferred that the PC signal build-up is not instantaneous but takes several minutes, and it decays slowly with respect to time. For a recording angle of 7°, the highest PC signal was reached approximately after 70 min

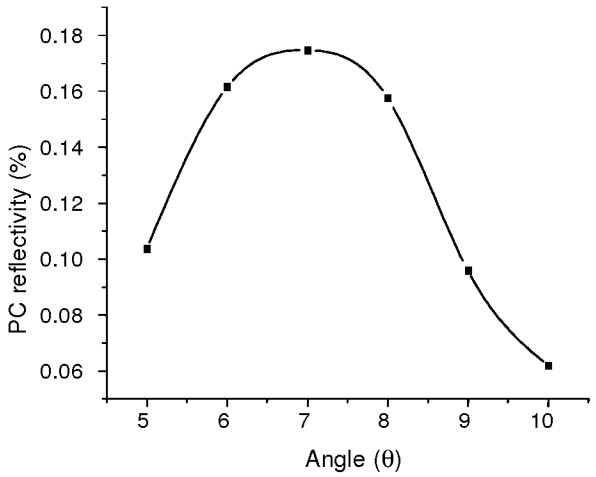

Figure 6. Measured PC reflectivity vs angle between probe and forward-pump beams.

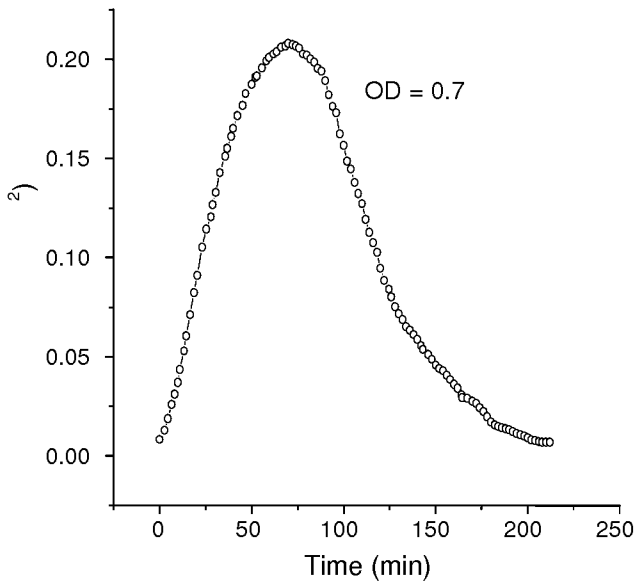

Figure 7. Growth of PC signal as a function of time.

and then it starts decreasing. The decrease in PC signal after reaching a maximum value may be due to photo-bleaching.

The existence of photo-bleaching can be inferred from a simple experiment described as follows. Acid blue 7-doped gelatin film was illuminated with 633 nm radiations at two different incident intensities and the corresponding transmittance of the sample was continuously monitored with respect to time. From the Figure 8, it is clear that the transmittance of the sample increases from 7.6 to 12.7% and 7.87 to 15.25% in a time duration of about 30 min, corresponding to incident light intensities of 1.22 and 2.52 W/cm^2 respectively. The observed increase in 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. Such observations have been already reported[15].

A systematic study has been made to investigate the influence of dye concentration of gelatin film on intensity of the PC signal. Figure 9 shows the PC reflectivity versus time for different dye concentrations of gelatin film. For higher concentration of the film, the rate of grating formation is initially slow, but the maximum value of PC signal obtained is high compared to lower concentration of the film. These results confirm that the PC reflectivity increases with increase in concentration of the dye. However,

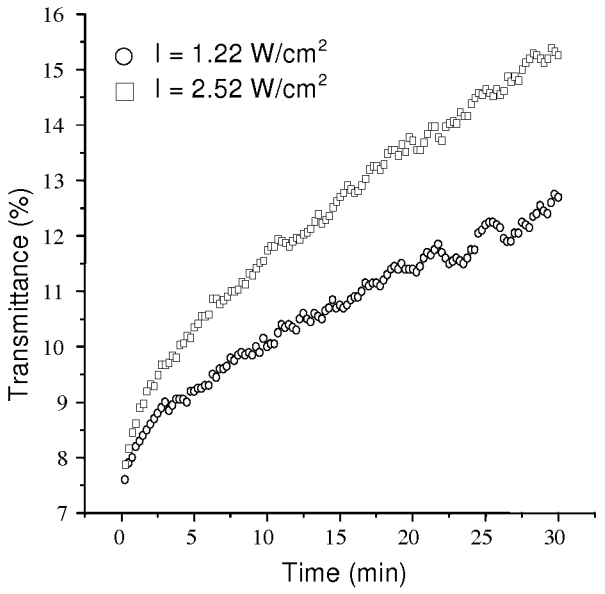

Figure 8. Transmittance of acid blue 7 dye-doped gelatin film with respect to time.

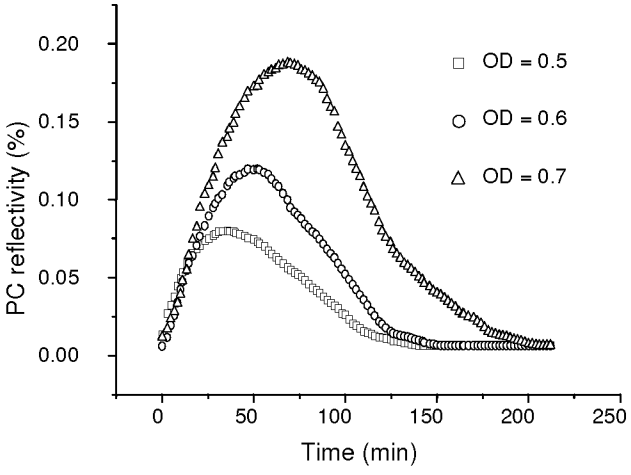

Figure 9. Measured PC reflectivity as a function dye concentration of acid blue 7-doped gelatin films.

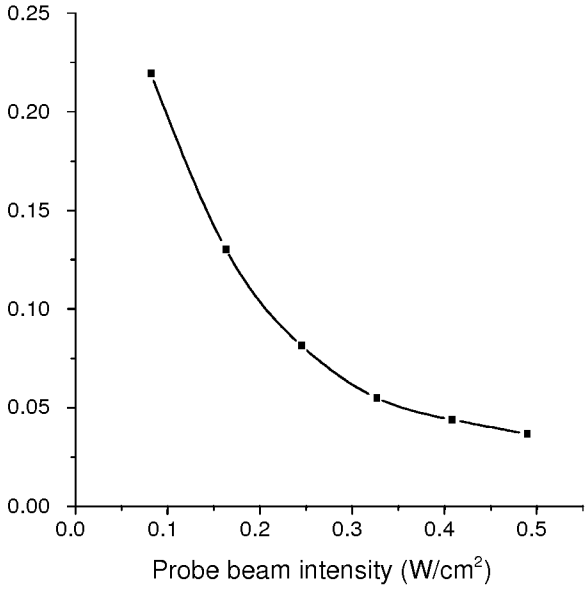

Figure 10. Conjugate reflectivity as a function of probe beam intensity.

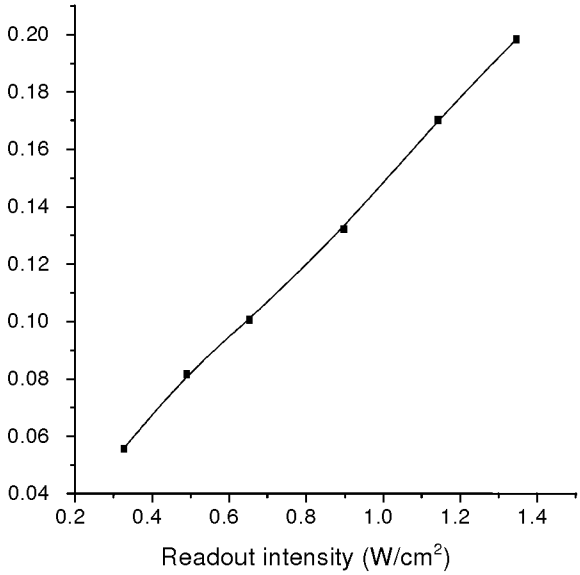

Figure 11. Dependence of PC reflectivity on readout beam intensity.

higher concentrated plates need longer exposure time; this may lead to mechanical instability and hence reduction in PC reflectivity.

The results of Figure 10 reveal the influence of the input probe beam intensity on the conjugate beam reflectivity. We observed a maximum reflectivity value of 0.22% for probe beam intensity $\cong 0.11$ W/cm^2, and further increase in probe beam intensity leads to decrease in PC reflectivity. Similar observations have been reported in other kinds of material doped with organic dyes[23].

Investigations were carried out to study the variation of PC reflectivity as a function of read-out (backward-pump) beam intensity, maintaining both the forward-pump beam and the probe beam intensities constant. From Figure 11, it is seen that higher readout intensities are necessary to achieve higher reflectivity.

Conclusions

Thus we have observed low-power OPC in acid blue 7 dye-doped gelatin film using 633 nm He–Ne laser radiation. We studied the various experimental parameters which influenced PC signal generation in acid blue 7 dye-doped films. Existence of light-induced bleaching process associated with this dye-doped system is also discussed. This kind of dye-doped system in red wavelength region may have potential applications in designing nonlinear optical devices and in interferometry studies.

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