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STUDY OF LOW POWER DEGENERATE FOUR-WAVE MIXING IN DISPERSE YELLOW DYE-DOPED POLYMER FILM

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Abstract:

Nonlinear optical phase conjugation by degenerate four-wave mixing (DFWM) is an important technique with applications in many fields of science and technology including image transmission, optical image processing, optical filtering, and laser resonators. In optimum condition, when two counter-propagating and intense light beams interact with a nonlinear medium, together with a less intense third beam, a fourth beam is generated from the medium, which will be the phase conjugation of the third beam. This technique is called four-wave mixing. In this paper, the optical phase conjugation property of disperse yellow (DY-7) dye-doped polymer films is studied using degenerate four-wave mixing method and the dependence of phase conjugated signal reflectivity on various parameters viz., dye concentration, the intensity of backward pump, forward pump, and inter-beam angle between the probe and forward pump beam on phase conjugation reflectivity are studied and presented using a low power continuous wave laser at 532 nm wavelength.

1. Introduction:

Nonlinear optical phase conjugation by degenerate four-wave mixing (DFWM) is an important technique with applications in many fields of science and technology including image transmission, optical image processing, optical filtering, and laser resonators [1-3]. When two counter-propagating and intense light beams interact with a nonlinear medium, together with a less intense third beam, a fourth beam is generated from the medium, which will be the phase conjugation of the third beam. This technique is called four-wave mixing. The unique feature of a pair of phase-conjugate beams is that the aberration influence contained in the forward (signal) beam passed through an inhomogeneous or disturbing medium (nonlinear medium) can be automatically removed from the backward (phase-conjugated) beam passed through the same disturbing medium [4]. The degenerate four-wave techniques are mainly used in nonlinear spectroscopy, real-time holography, and phase conjugation mirrors. Optical phase conjugation by degenerate four-wave mixing has been demonstrated in many organic and inorganic materials using a light beam of pulsed or continuous-wave (CW) lasers. [5-12].

The phase conjugate light beam has a variety of characteristics and properties which are not seen with normal light beam [13] and they can be described as follows:

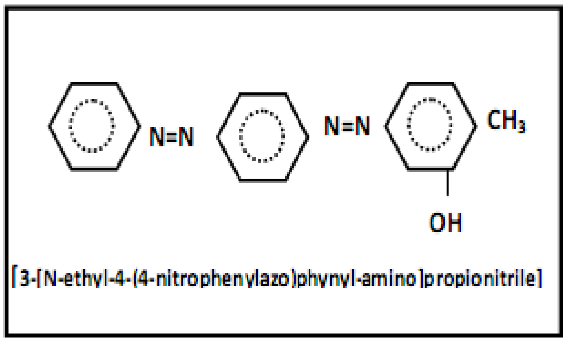
- ✓ **Phase Compensation Effect:** The phase distortion of the wave is compensated if is re-propagated through the medium.
- ✓ **Space Domain Multiplicative Interaction:** Since phase conjugate waves are produced by interaction in a nonlinear medium, the spatial multiplicative effect among the interacting light waves appears in the phase conjugate waves.

- ✓ **Intensity-Dependent Phase Shift:** The phase shift of the electric field of phase conjugated beam depends on the wave intensity due to the optical Kerr effect in a nonlinear medium.
- ✓ **Dependence Detuning:** The reflectance of a phase conjugate mirror strongly depends on the detuning between the pump and probe wave frequencies.
- ✓ **Time Inversion:** A phase conjugate light beam can be regarded as a time inverted light beam since its direction of propagation of the beam is exactly opposite to that of the probe beam and the wave-front of the light beam is identical to the wave-front of the probe beam.
- ✓ **Multiplicative Time Domain Interaction:** When pulsed laser light beams are used in the production of phase conjugate light the resulting polarization and hence the generalized phase conjugate light intensity depends on the product of the interacting light waves in the waves' temporal domains.
- ✓ **Quantum Correlation:** When the quantum mechanical analysis is used to correlate the forward and backward waves theoretically, the probe and phase conjugate waves are regarded as the photon annihilation and creation operators respectively.

In this paper, the optical phase conjugation property of disperse yellow (DY-7) dye-doped polymer films is studied using degenerate four-wave mixing method and the dependence of phase conjugated signal reflectivity on various parameters viz., dye concentration, the intensity of backward pump, forward pump, and inter-beam angle between the probe and forward pump beam on phase conjugation reflectivity are studied and presented at 532 nm wavelength.

2. Preparation & Linear Optical Properties of DY-7 Doped Polymer Films:

Commercially available Disperse Yellow -7 (DY-7) (Aldrich Chemical Co.) is purified by recrystallization twice with spectrogradeethanol and by vacuum sublimation. The purity is determined spectroscopically. Purified chloroform is used as the solvent. To prepare the film, Polymethyl methacrylate – metacrylic acid is used as a polymer matrix. The thin films of DY-7 doped in PMMA-MA are prepared using hot press technique [14]. Thin films of variable thickness (10μm) with 1 mM, 2 mM, and 5 mM dye concentration are prepared between two glass slides and are used as samples for DFWM experiment.



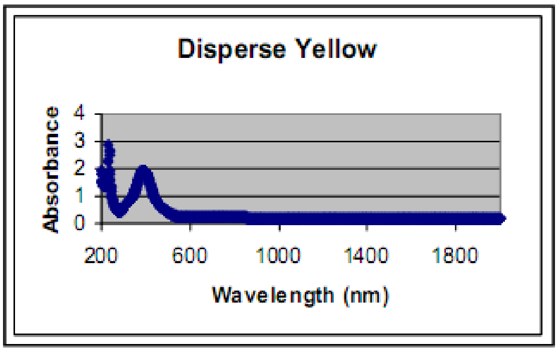


Figure 1: (a) Molecular structure of DY-7. (b) Linear absorption spectrum of DY-7.

The linear absorption spectrum of DY-7 doped in PMMA-MA is measured on a VARIAN Cary UV-vis-IR recording Spectrophotometer. The figure 1 shows the linear absorption spectrum of the film. The spectral curve has shown that there is a strong absorption band with peak absorption located at 468 nm with a bandwidth of 80 nm, a medium absorption peaked at 270 nm with a bandwidth of 60 nm and no linear absorption is observed in entire spectral range of 580 to 1200 nm. Z-scan technique is used to study the nonlinear optical properties of the sample [5]. In open aperture Z-scan, DY-7 has shown decrease in transmittance with increase in irradiance due to reverse saturation absorption [6-8].

3. Experimental Configuration for Degenerate Four-Wave Mixing:

The ray diagram of phase-conjugate wave generation by degenerate four-wave mixing is shown in figure 2. The probe beam E_3 and the forward-pump beam E_1 interfere in the nonlinear material and create a periodic interference pattern [13]. The resulting grating wave vector amplitude is $k = 2\pi/\Lambda$. The fringe period (Λ) can be determined by the well-known formula: $\Lambda = \lambda/2 \sin(\theta/2)$ where, $\lambda$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. The backward-pump beam E_2 is then diffracted under Bragg conditions by the dynamic volume hologram and generates a backward conjugate wave-front whose complex amplitude can be written as E_4.

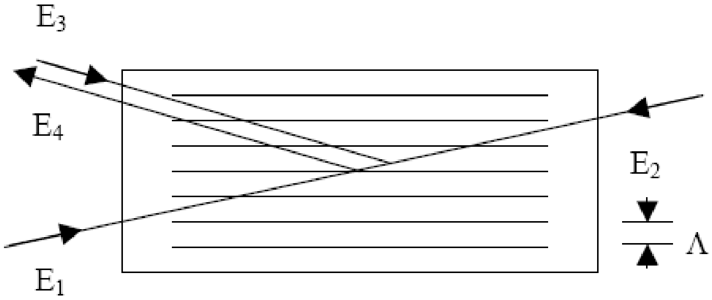


Figure 2: Ray diagram of phase-conjugate wave generation by degenerate four-wave mixing [13]

4. Experimental Set-up for Degenerate Four Wave Mixing:

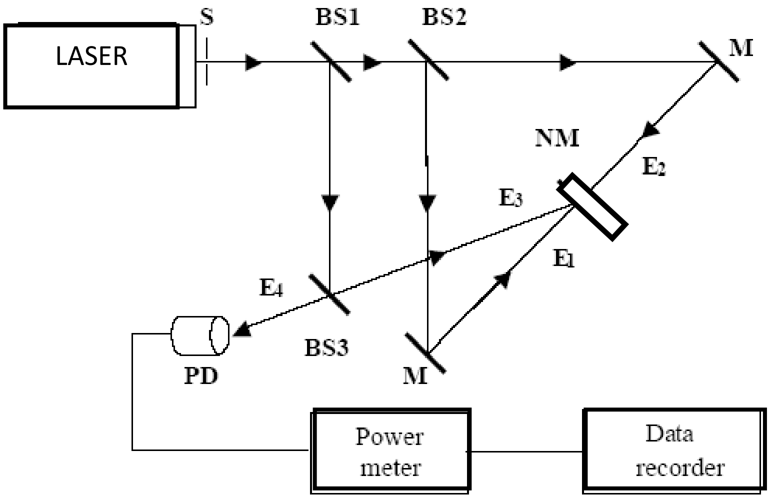


Figure 3: Experimental set-up for observation of Phase Conjugated wave using Degenerate Four-Wave Mixing configuration. S, Shutter; BS1–BS3, Beam splitters; M, Mirror; NM, Nonlinear medium; PD, Photo-detector [7]

The schematic diagram of the experimental setup to study phase conjugation effect is shown in Figure 3. A Semiconductor Diode laser (20 mW) beam at 532nm is divided into three beams, two counter-propagating pump beams E_1 and E_2 namely forward-pump and backward-pump beams respectively and E_3 is a probe beam to form the DFWM configuration. The spot size of each of these three unfocussed beams at the nonlinear medium is 1.20 mm in diameter. The constant power ratio of the forward-pump beam (E_1), the backward-pump beam (E_2), and the probe beam (E_3), used in this work is ≈10 : 10 : 1. The angle between the probe beam and the forward-pump beam can be varied and adjusted to a specific angle θ. The sample is exposed to all these three beams simultaneously. The optical path lengths of all the three beams are made equal so that they were coherent in the sample. The phase-conjugate wave retraces the path in the opposite direction to that of the probe beam E_3 and is detected with the help of a photo-detector and processed by a power meter and data recorder system. The experimental set-up is mounted on a vibration isolation table to avoid the destruction of the laser-induced gratings formed in the DY-7 dye-doped polymer matrix due to mechanical disturbances.

5. Procedure for Phase Conjugated Signal Generation & Study:

After generating the phase conjugated waves using DFWM experimental set-up shown in figure 3, the dependence of phase conjugated signal reflectivity on various parameters viz., dye concentration, intensity of backward, forward pump and inter-beam angle between the probe and forward pump beam on phase conjugation reflectivity are studied for all the three samples. The graphs are drawn between two parameters as listed below:

- ✓ Maximum PC Reflectivity at different dye concentrations.
- ✓ Phase conjugated signal recording time at different concentrations.
- ✓ Phase conjugated signal reflectivity as a function of the angle between the probe and forward pump beam.
- ✓ Dependence of phase conjugated signal reflectivity on backward pump beam intensity.
- ✓ Transmission of phase conjugated signal as a function of Time.
- ✓ Phase conjugated signal reflectivity as a function of the probe beam intensity.
- ✓ Phase conjugated signal reflectivity as a function of forward pump intensity.

In the case of Disperse Yellow dye-doped PMMA-MA films, the PC signal measurements are taken by varying the parameters which influence the PC signal during the DFWM process. Figure4shows the PC signal versus the time for different dye concentration of the doped polymer films. PC intensity rises linearly to a maximum and then starts decreasing. The phase grating formed is transient. To get maximum reflectivities, it is necessary that there be a perfect overlap of the probe and the pump beams in the nonlinear medium. Figure5shows the influence of the input probe beam intensity on the conjugate beam reflectivity. A maximum reflectivity value of 0.16% is observed for probe beam intensity at 0.11 W/cm^2, and further increase in probe beam intensity resulted to decrease in PC reflectivity. Similar observations have been reported in other kinds of material doped with organic dyes [15-18]. Figure6 shows the PC reflectivity as a function of recording angle between the forward pump and probe beam. It seems from the figure that, as the angle between the probe beam and the forward pump beam increases, initially the PC reflectivity increases and then decreases. This may be because as the angle increases, the probe beam becomes elliptical shape and only a fraction of its area falls within the interaction region. Because of two-wave coupling, the maximum PC reflectivity is achieved when the angle is 7 degrees. The effect of the backward pump beam power on the PC reflectivity by keeping the power of the forward pump and probe beams constant and varying the backward pump beam is shown in figure7. Figure8 shows the variation of reflectivity for the different power of forward pump beam. The PC reflectivity increases linearly with the power of forward pump beam. There are two main processes which must be considered in the discussion of the origin of OPC in dye-doped PMMA-PA films: (1) the formation of thermal grating and (2) third-order nonlinear optical processes 19-20]. The DY-7 film illuminated with

532 nm radiation of a variable intensity and the transmittance of the sample is measured simultaneously by using photo detector. If the effect observed in our experiment is of purely thermal nature, bleaching of the sample film should have been observed. The results obtained for the sample are shown in figure9. It is clearly demonstrated that the transmission of the sample increases with time. The experiment described above indicates that the third order nonlinear processes like reverse saturable absorption (RSA) is mainly responsible for OPC in the case of DY-7 doped PMMA-MA polymer films.

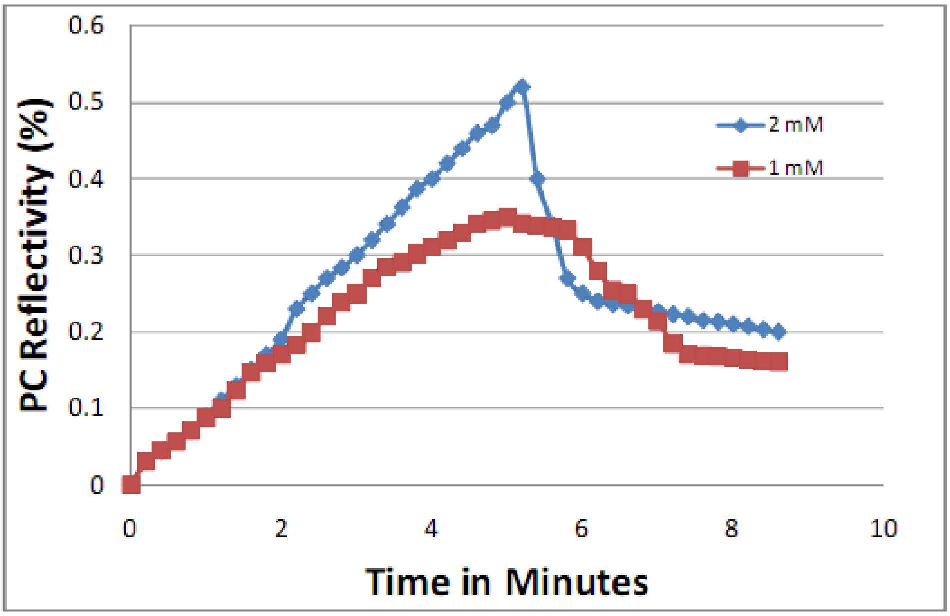


Figure 4: PC signal versus recording time for different concentration for DY-7 at 532 nm.

Disperse Yellow dye doped in a polymer matrix have the capability of generating a phase-conjugate wave by not only by third-harmonic nonlinearity using DFWM but also by the holographic process [21]. To distinguish the phase-conjugate wave generated by third-harmonic nonlinearity using DFWM from that by the holographic process, the transient behavior of the PC signal is studied. For this, the DY-7 dye doped in PMMA – MA polymer matrix is first illuminated with three waves E_1, E_2, and E_3 for a specified duration, and afterward, E_1 and $E_3$are successively turned off, so that only $E_2$is incident on the dye film. Figure4 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 switching 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 is 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 DY-7 dye in PMMA – MA polymer 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.

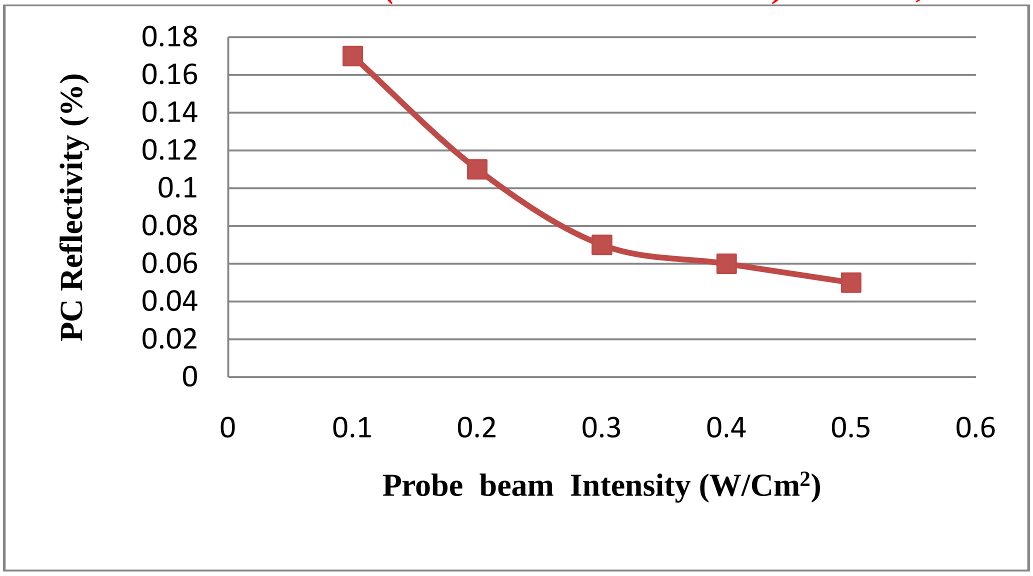


Figure 5: Conjugate reflectivity as a function of probe beam intensity for DY-7 at 532 nm.

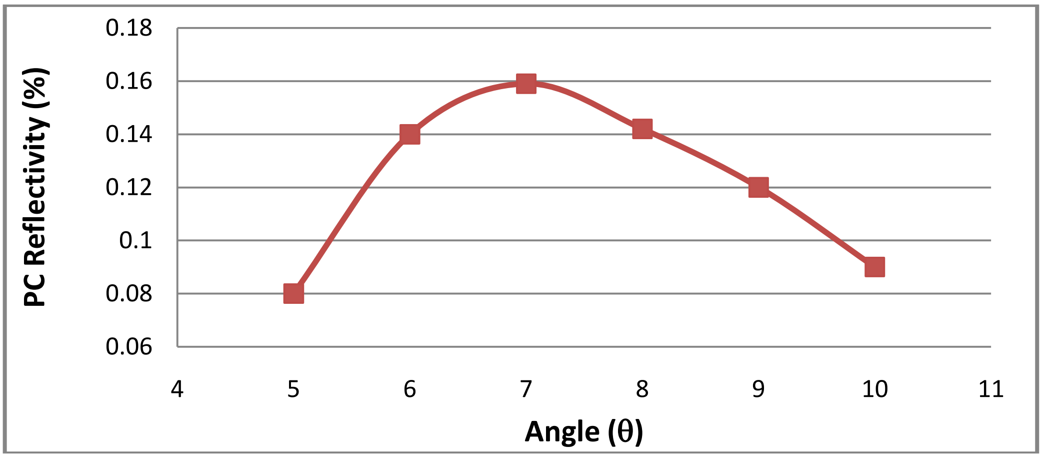


Figure 6: PC Reflectivity as function of angle between the probe and forward pump beams for DY-7 at 532 nm

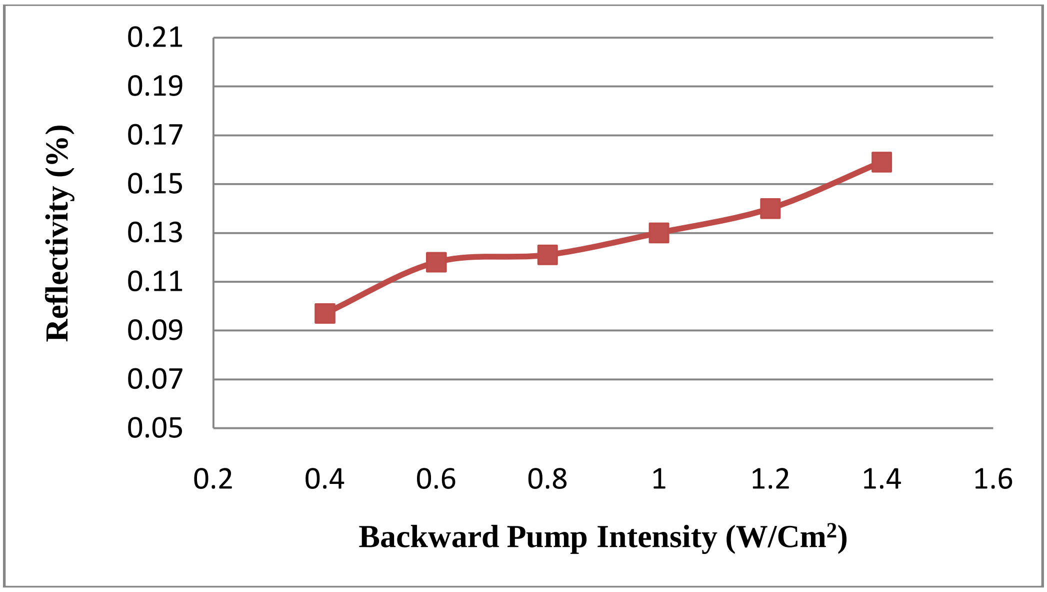


Figure 7: Dependence of PC reflectivity on backward pump Intensity for DY-7 at 532 nm.

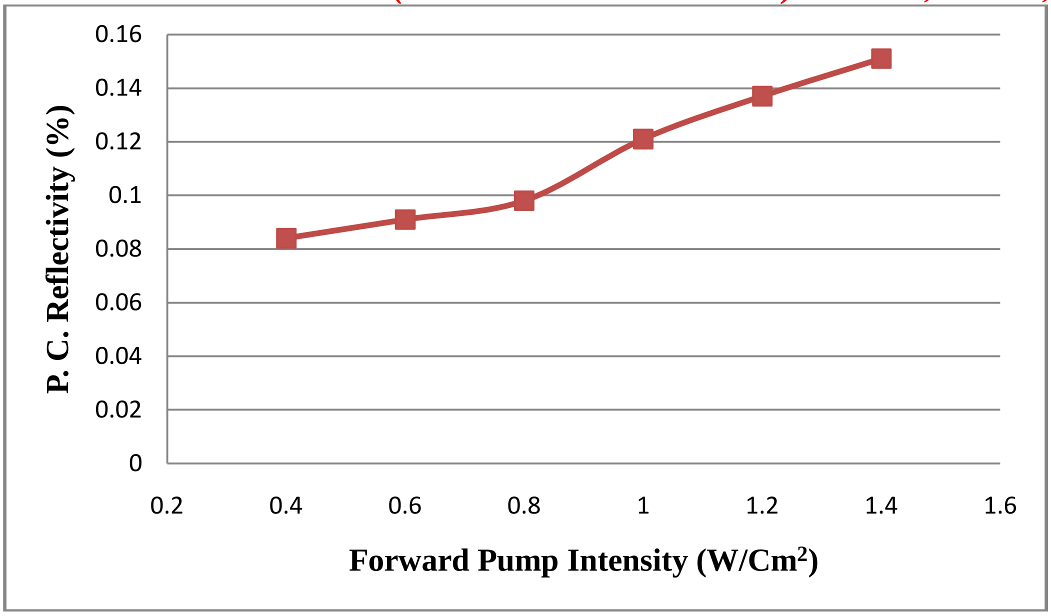


Figure 8: Dependence of PC reflectivity on forward pump power for DY-7 at 532 nm.

6. Results and Discussion of OPC Study:

The dependence of phase conjugated signal reflectivity on different parameters viz., dye concentration, intensity of backward, forward pump and inter beam angle between the probe and forward pump beam on phase conjugation reflectivity are studied. The graphs of the different parameters in relation to optical phase conjugation study are depicted as shown below.

- ✓ Maximum PC Reflectivity at different dye concentrations.
- ✓ PC signal recording time at different concentrations.
- ✓ PC reflectivity as a function of the angle between the probe and forward pump beam.
- ✓ Dependence of PC reflectivity on backward pump intensity.
- ✓ Transmission as a function of Time.
- ✓ PC Reflectivity as a function of the probe beam intensity.
- ✓ PC Reflectivity as a function of forward pump intensity.

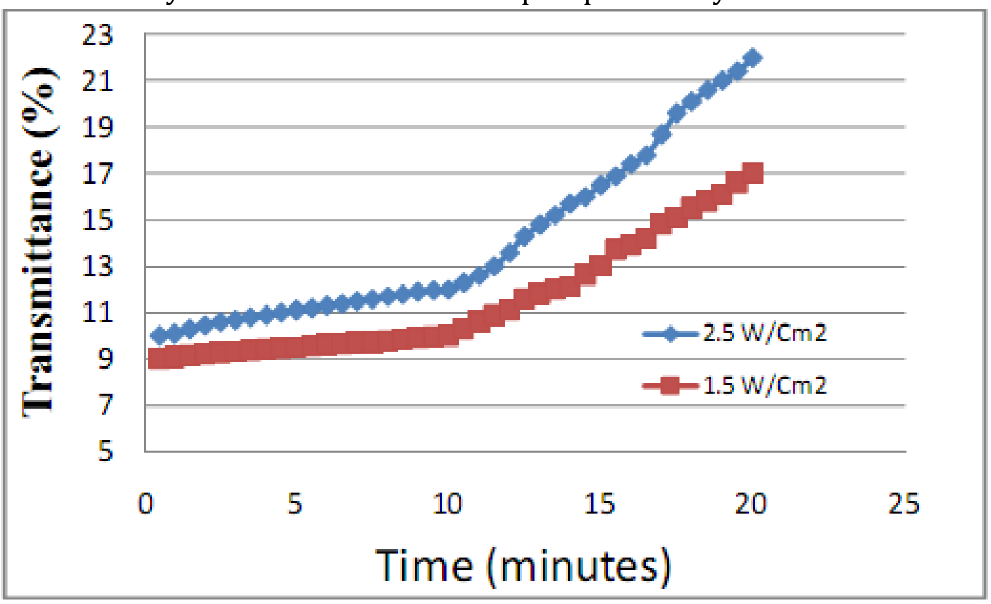


Figure 9: Transmission as a function of time for DY-7 at 532 nm.

Table 1 contains the results of PC reflectivity at different pump beam, probe beam and angle between them for DY-7 doped PMMA-MA films at the input wavelength 532 nm. The nonlinear PC reflectivity (R) which is the ratio of the incident signal beam intensity and the backward phase-conjugate beam intensity and is determined for all the three samples. $R = I_1/I_2$. In azo dye-doped polymers, the chemical linking between the dye and the polymer is negligible and hence can be used as optical data storage media [22]. Another advantage of azo dye-doped polymers is their nonlinear optical behavior. Therefore, they can be used as modulators or optical frequency doublers and switches while showing good mechanical characteristics and processability [23 -27].

Table 1: PC reflectivity at different pump beam, probe beam and angle between them at the input wavelength 532 nm

S.No	Parameter	Measured Value
1	Absorption range & absorption peak wavelength	80 nm & 468 nm
2	Film thickness	10 µm
3	Optimum dye concentration	2 mM
4	Sample transmittance at 532 nm	0.6
5	Maximum PC reflectivity at pump beam intensity of 1.5 W/cm^2	0.14%
6	Maximum PC reflectivity at probe beam intensity 0.11 W/cm^2	0.16%
7	Angle between probe & pump beam for maximum PC reflectivity (in degrees)	7

7. Conclusion:

The optical phase conjugation property of Dy-7 dye-doped polymer film is studied using degenerate four wave mixing method and the dependence of phase conjugated signal reflectivity on various parameters viz., dye concentration, intensity of backward, forward pump and inter-beam angle between probe and forward pump beam on phase conjugation reflectivity are studied and the results are depicted and analysed. We have observed low-intensity optical phase-conjugation in DY-7 dye in PMMA – MA polymer matrix using a degenerate four-wave mixing set-up, employing 532 nm light radiation from a Diode laser. The mechanism of phase-conjugate wave generation associated with this dye-doped system is discussed. The maximum phase-conjugate beam reflectivity observed in these dye films is about 0.16%. The maximum PC reflectivity is achieved when the angle between the probe and forward pump beam is 7 degrees. The effects of dye intensity backward, forward pump and interbeam angle between the probe and forward pump beam on phase conjugation reflectivity are also studied. PC signal reflectivity first increases and then decreases with time. PC reflectivity is increased by increasing the intensity of the backward and forward pump beam. The polarization and intensity profile are verified to be preserved in the conjugate signal. The predominant phase conjugation signal is attributed to the fact that the sample shows third harmonic nonlinearity even at low light intensity due to reverse saturable absorption (RSA) and thermal effect on the dye molecules. The decay time of the recorded grating at fixed pump intensity also decreases with decreasing temperature. Since the sample film is used with low power CW laser beam at 532 nm, this DY-7 dye in PMMA – MA polymer film may be a promising material for real-time phase-conjugate interferometry.

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