

Degenerate Four-Wave Mixing Experiments In Rose Bengal Dye Doped Gelatin Film.

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Abstract

Optical phase-conjugation (OPC) was observed in Rose Bengal dye-doped gelatin films via Degenerate four-wave mixing (DFWM) using continuous wave of SHG of Nd-YAG laser radiation ($\lambda=532$ nm) of total power 50 mW. Various parameters which influence the phase-conjugate (PC) signal during the DFWM process were studied. The PC signal contributions from induced holographic transmission and reflection gratings were measured. It observed a maximum PC beam reflectivity of 0.15% in these dye-doped gelatin films.

Keywords: *Nonlinear optics, Optical Phase Conjugation, Dye*

1. Introduction

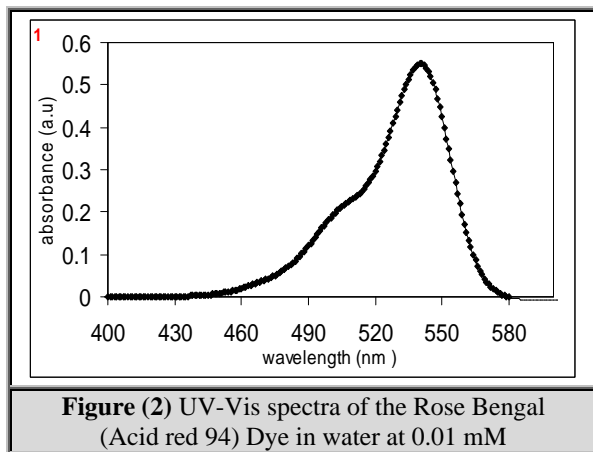
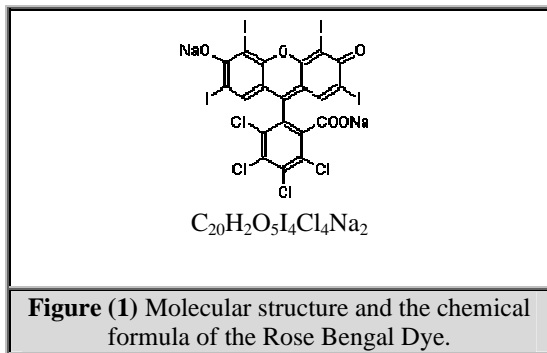
Optical phase-conjugation (OPC) has potential applications in image transmission, optical image processing, optical filtering, and laser resonators [1-3]. Generation of PC wave through degenerate four waves mixing (DFWM) has been investigated extensively in the past two decades in films or bulk media. OPC defines usually a special relationship between two coherent optical beams propagating in opposite directions with reversed wave front 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 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 [4-13]. Glasses and other solid matrices doped organic dyes emerged as promising materials for OPC because of their large third-order nonlinearity χ^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 Fluorone [14] dye-doped gelatin films using low-power continuous-wave laser excitation. The nonlinear medium (NM) used in this work is gelatin films doped with Rose Bengal dye as the photosensitive chromophore.

2. Experimental

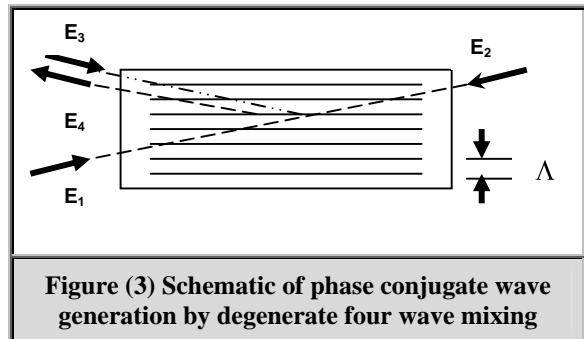
2.1 Materials

The organic dye Rose Bengal (Acid red 94) belongs to the Fluorone group. The chromophore of this class is the quinonoid group; the chemical structure and molecular formula of Rose Bengal dye are shown in fig.1. The UV-Vis absorption spectra of Rose Bengal (acid red 94) dye was studied using UV-Vis spectro-photometer (Perkin Elmer-Lambda 35) and it exhibits the peak absorption at 540 nm as in fig. 2.



In this work, Agfa-Gaevert 10E75 holographic plates are used to prepare the gelatin coated films on a glass plate. The silver halide is removed from holographic plates by fixing the unexposed plates in sodium thiosulphate and washing with water. The thickness of the gelatin films obtained are of $7\mu\text{m}$. Photosensitive plates with different optical densities are obtained by soaking the gelatin plates in aqueous solutions of Rose Bengal dyes of different concentrations by maintaining the soaking time constant (5 minutes). The plates are then dried at room temperature and used for the study without any further processing. The thicknesses of the films were measured using (AFM) (Atomic force microscope) it was of the order of 10 microns.

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 fig.4. A (SHG) of Nd-YAG laser (COHERENT – Compass 215M diode-pumped laser - 50 mW) beam at 532 nm was divided into three beams, two counter-propagating pump beams E1 and E2 namely forward-pump and backward-pump beams respectively and a probe beam E3 to form the DFWM configuration. The spot size of each of these three unfocussed beams at the nonlinear medium was 0.32 mm in diameter. The constant power ratio of the probe beam (E3), forward-pump beam (E1) and backward-pump beam (E2) used in this work was (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 E3 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 Rose Bengal dye-doped gelatin film due to mechanical disturbances.

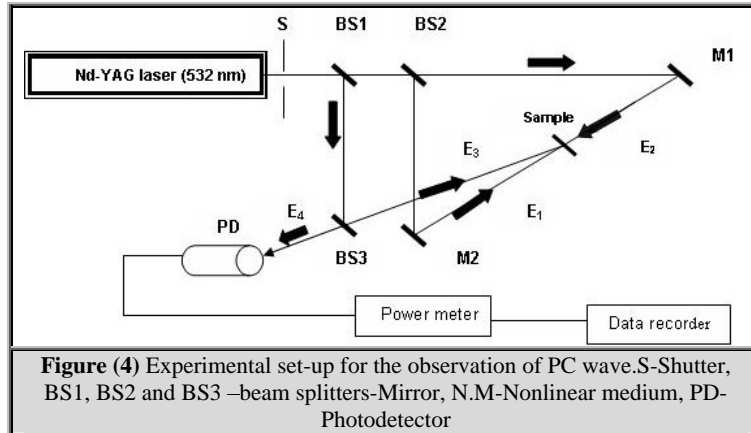


Figure (4) Experimental set-up for the observation of PC wave.S-Shutter, BS1, BS2 and BS3 –beam splitters-Mirror, N.M-Nonlinear medium, PD-Photodetector

3. Results and discussion

There are two cases to be considered for the mechanism of phase-conjugate wave generation associated with the Rose Bengal dye-sensitized gelatin film. The first one is due to the third-order nonlinearity χ^3 and the second is due to absorption saturation and photobleaching which lead to the formation of a laser induced grating in the medium. It considers a three-level system for the Rose Bengal dye in gelatin matrix. Fig. 5 shows an energy level diagram of Rose Bengal 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 dye molecule results in transition of the dye molecules to the first excited singlet state ($S_0 \rightarrow S_1$). If the singlet-to-triplet crossover 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 ($\Lambda = \lambda/2 \sin \theta/2$) fig.3 illustrate the phase conjugation signal generation by degenerate four wave mixing from nonlinear material), where λ is the laser wavelength, and θ is the forward-pump and probe beam incident angles with respect to the normal to the nonlinear medium.

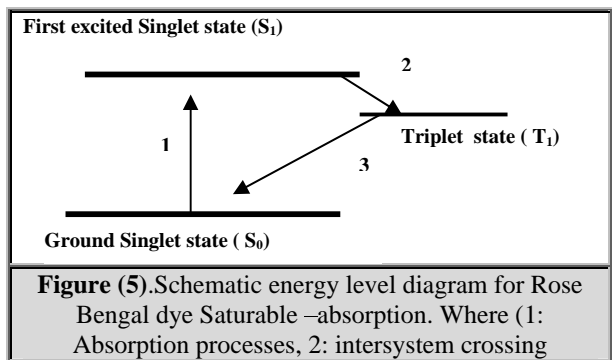


Figure (5).Schematic energy level diagram for Rose Bengal dye Saturable-absorption. Where (1: Absorption processes, 2: intersystem crossing

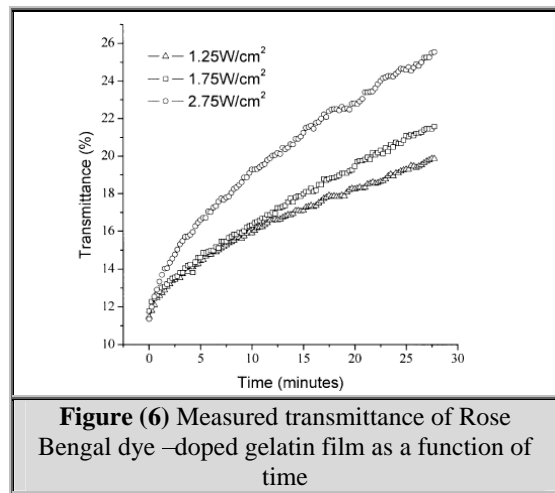


Figure (6) Measured transmittance of Rose Bengal dye-doped gelatin film as a function of time

Photobleaching of the dye molecules at the excitation wavelength also should be considered in this discussion. The existence of photobleaching can be inferred from simple experiment described as follows. Rose Bengal dye-doped gelatin film was illuminated with 532 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 fig. 6) corresponding to the incident light intensities of 1.25, 1.75 and 2.75 W/cm² 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 [10]

Dyes fixed in a solid matrix have the capability of generating a phase-conjugate wave by not only DFWM but also holographic process [15]. 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 Rose Bengal dye-sensitized gelatin film was first illuminated with three waves E1, E2 and E3 for a specified duration, and afterwards, E1 and E3 were successively turned off, so that only E2 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. Fig. 7 shows the measured phase-conjugate signal as a function of time.

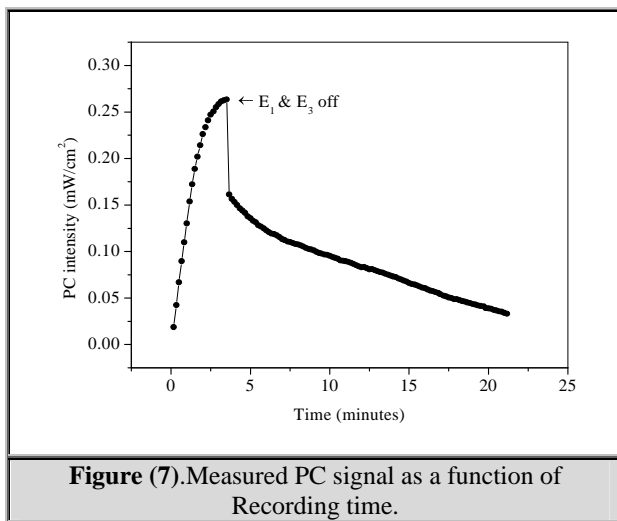


Figure (7). Measured PC signal as a function of Recording time.

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 E1 and E3 indicates the contribution from the fast DFWM process. Due to the holographic process the PC signal is present even after E1 and E3 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 E1, E2 and E3 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 E1 and E3 can be recorded in the Rose Bengal dye-sensitized gelatin film, the phase conjugate wave can still be generated when E2 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 Rose Bengal dye-doped gelatin films using a degenerate four-wave mixing set-up, employing 532 nm light radiation from a (SHG) of Nd-YAG 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 0.15%. Rose Bengal dye-doped gelatin film may be a promising material for real-time double-exposure phase-conjugate interferometry.

5. References

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الخلاصة :

في هذا البحث تمت ملاحظة الاقتران الطوري البصري (Optical Phase Conjugation) لصبغة الـ (Rose Bengal) المطعمه بأغشية الجيلاتين بواسطة تقنية انحلال اربعة امواج ضوئية متمازجة (Degenerate Four Wave Mixing) باستخدام اشعة ليزرية ذات موجه مستمره للتولد التوافقي الثاني من ليزر النديميوم - ياك ذي الطول الموجي (٥٣٢) نانومتر وبقدرة كليه مقدارها ٥٠ ملي واط.ان العديد من المعلمات والتي تؤثر على اشارة الاقتران الطوري (phase conjugated signal) خلال عملية الاقتران الطوري البصري قد تمت دراستها . كذلك تم قياس أثر مساهمة الحيود (الانتقالي والانعكاسي) على اشارة الاقتران الطوري ، كما تم ملاحظة اعظم انعكاسيه لحزمة الاقتران الطوري بمقدار ٠.١٥ % لهذه الصبغه المطعمه بأغشية الجيلاتين.

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