

## Dye-doped gelatin films for phase-conjugation studies in undergraduate optics laboratory

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### Abstract

In the present paper, an overview of optical phase-conjugation and the experimental observation of optical phase-conjugation based on the degenerate four-wave mixing are presented. A continuous wave 633 nm He–Ne laser of total power 35 mW is used to generate phase-conjugate signal from the Acid blue 7 (Alphazurine) dye-doped gelatin film. A maximum phase-conjugate reflectivity of 0.22 % has been observed in these dye films. Degenerate four-wave mixing based optical phase-conjugation can easily be demonstrated for undergraduate students in any optics laboratory using low power lasers.

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**Key words:** Holography, Optical Phase-conjugation, Dye-doped gelatin films.

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### 1. Introduction

Optical phase-conjugation (OPC) is a technique that incorporates nonlinear optical effects to precisely reverse both the direction of propagation and the overall phase factor of a light wave. The process can be regarded as a unique kind of mirror with very unusual image-transformation properties. Optical phase-

conjugation was first observed by Zel'dovich et al [1] in 1972, but its roots can be traced back further to earlier work in static and dynamic holography [2,3].

Optical phase-conjugation is an important technique with applications in many fields of science and engineering such as spectroscopy, adaptive optics, and real-time image processing or phase-conjugate mirrors. In general, OPC is a highly useful and very unique

approach to explore the nonlinear optical properties of various materials and to investigate different physical processes occurring in those media under the action of strong coherent optical fields. Phase-conjugation (PC) by degenerate four-wave mixing (DFWM) has been demonstrated in numerous nonlinear media [4–8] and most of the experiments utilized high peak power pulsed lasers in order to obtain measurable signals. The majority of published works in the field of optical phase-conjugation and Four-wave mixing (FWM) had been based on studying the nonlinear optical properties of a great variety of materials which include dye solutions [9-12] or dye-doped matrixes [13-16], impurity-doped glasses [17-18], crystals [19,20] and fullerenes (e.g. C60) related materials [21,22].

This paper, presents the fundamentals of optical phase-conjugation and the experimental demonstration in Acid blue 7 dye-doped gelatin films through degenerate

four-wave mixing geometry using low power continuous wave He–Ne laser.

## 2.Theory

Two optical beams are considered to be phase-conjugate to each other if they have same wavefronts but propagating in opposite directions. This means that the k-vectors of the two beams have the opposite sign and that the amplitude functions are complex conjugate of each other. When an optical beam is incident on a conventional mirror, only the component of the k-vector that is perpendicular to the plane of the mirror is reversed. If a divergent beam is incident on a conventional plane mirror its reflection will diverge in the same manner.

Hence for a beam reflecting from a flat mirror located in the x–y plane, reflection is given by

$$A \exp i \left( (k_x + k_y + k_z).r - \omega t \right) \Rightarrow A' \exp i \left( (k_x + k_y - k_z).r - \omega t \right) \quad (1)$$

However, the beam incident on a phase-conjugate mirror (PCM) reverses all three components of the k-vector and is given by

$$A \exp i \left( (k_x + k_y + k_z).r - \omega t \right) \Rightarrow A' \exp i \left( -(k_x + k_y - k_z).r - \omega t \right) \quad (2)$$

This causes the reflected wavefront to exactly retrace the path of the incident beam regardless of the initial spatial structure of the beam. There are many nonlinear optical processes that are capable of generating phase-conjugate (PC) waves. Holography can

be considered as a static form of phase-conjugation [4,5]. The reconstructed field that forms a real image in conventional holography is actually a phase-conjugate replica of the original object field. Stimulated scattering processes are another important class of

interactions that can be used for optical phase-conjugation. Degenerate four-wave mixing (DFWM) is one of the most convenient processes for the generation of high-quality PC waves.

### 3. Optical phase-conjugation through DFWM

There is a close analogy between the degenerate four-wave mixing (DFWM) process and the real time holography. As shown in the Fig.1(a), two counter-propagating pump waves ( $E_1$  and  $E_2$ ) pass through the nonlinear medium, and a probe wave ( $E_3$ ) is incident on the nonlinear medium at an angle with respect to the pump wave  $E_1$ . The wave vectors of these three light beams are  $k_1$ ,  $k_2$  and  $k_3$  respectively ( $k = 2\pi/\lambda$ ). Under this

experimental arrangement the backward propagating (phase-conjugate) wave can be generated through the reflection or transmission from two possible induced gratings. In the first case, the interference between the forward-pump beam  $E_1$  and the signal beam  $E_3$  that produces nearly parallel interference fringes (transmission grating) along the bisector direction of the crossing angle are considered. Since the refractive-index change in the medium is proportional to the local light intensity, one may realize that the interference fringes can produce an induced holographic grating within the nonlinear medium. In this case, the backward-pump beam acts as a reading beam during its passage through the induced grating. A diffracted (or reflected) wave  $E_4$  is created and the wave vector corresponding to this newly generated wave is  $k_4$ .

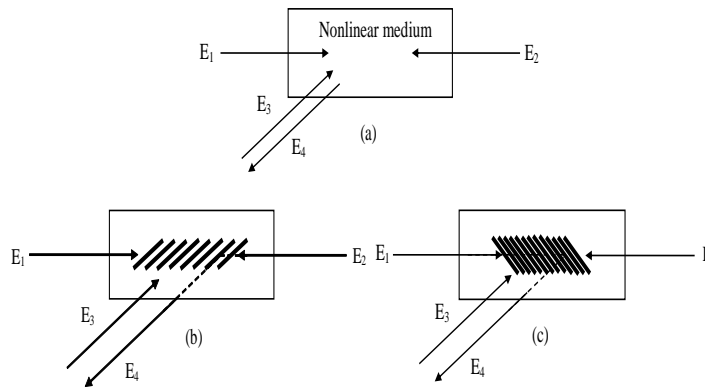


FIG.1: (a) A schematic of four-wave mixing, (b) a transmission grating formed by  $E_3$  and  $E_1$  (c) a reflection grating formed by  $E_3$  and  $E_2$

According to the holographic principle, it is known that this diffracted wave  $E_4$  has the spatial information carried by the incident signal beam  $E_3$ . In other words, the waves  $E_3$  and  $E_4$  are phase-conjugated to each other. To

further justify this conclusion one can treat the nonlinear medium as a holographic medium whose transmission function is determined by the interference-induced refractive index

modulation and can phenomenologically be expressed as

$$T \propto (E_1 + E_3)(E_1 + E_3)^* = |E_1|^2 + |E_3|^2 + E_1^* E_3 + E_1 E_3^* \quad (3)$$

where  $E_1$  and  $E_3$  denote the complex amplitudes of the forward-pump wave and the probe waves respectively, in the grating plane. As assumed the waves  $E_1$  and  $E_2$  are two counter-propagating plane waves and  $E_2 = E_1^*$ , so that the transmitted field of reading wave is given by

$$E_2' \propto T E_2 = T A_1^* = [|E_1|^2 + |E_3|^2] E_2 + E_3 (E_1^*)^2 + E_1 E_2 E_3^* \quad (4)$$

Here on the right-hand side of the equation, the first term proportional to  $E_2$  represents the zero-order diffracted wave that does not involve any spatial information and therefore of no interest to us. The contribution from the second term that actually involves a phase factor of  $\exp[-i(2k_1 \cdot r -$

As shown in Fig. 1(c), the backward wave  $E_4$  can also be generated through the diffraction of the holographic grating (reflection grating) induced by the waves  $E_2$  and  $E_3$ . In this case, the forward-pump wave  $E_1$  plays the role of the reading plane wave, and the reflected wave  $E_4$  is still phase-conjugated to the signal wave  $E_3$ . Since both these gratings contribute to the generation of the phase-conjugate wave  $E_4$ , the total conjugate field  $E_4$  is due to the coherent superposition of both

$k_3 z]$  can be neglected because its phase matching condition could not be fulfilled. The third term corresponds to the diffracted wave that involves the spatial information carried by the signal wave  $E_3$  and can be written separately as

$$E_4 \propto E_1 E_2 E_3^* \quad (5)$$

the processes. The period ( $\Lambda$ ) of these two gratings are different and can be written as

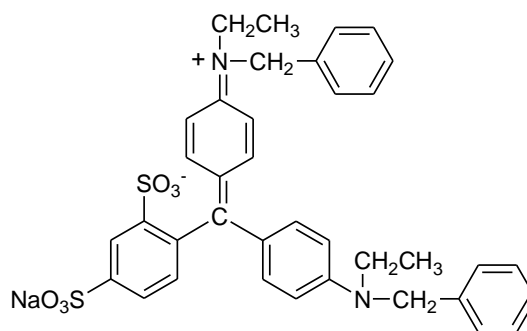
$$\Lambda_{13} = \frac{\lambda}{2 \sin(\theta/2)} \quad (6)$$

$$\Lambda_{23} = \frac{\lambda}{2 \cos(\theta/2)} \quad (7)$$

where  $\lambda$  is the wavelength of the waves in the medium. It is obvious that these periods of the induced gratings are determined by the corresponding spacing of the interference fringes formed by two appropriate waves.

## 4. Experimental studies

In this work, a commercially available synthetic dye, Acid blue 7 (Alphazurine A-C.I. 42080) was used as the nonlinear agent, which belongs to the triphenylmethane [23] groups. The general structure and formula of acid blue 7 dye are shown in Fig.2.



Molecular formula:  $C_{37}H_{35}N_2NaO_6S_2$

FIG. 2: Chemical structure and formula of Acid blue 7 dye

The UV-visible absorption spectra of acid blue 7 dye were studied using UV-2401 PC spectrophotometer and it exhibits the peak absorbance at 637 nm as shown in the Fig.3.

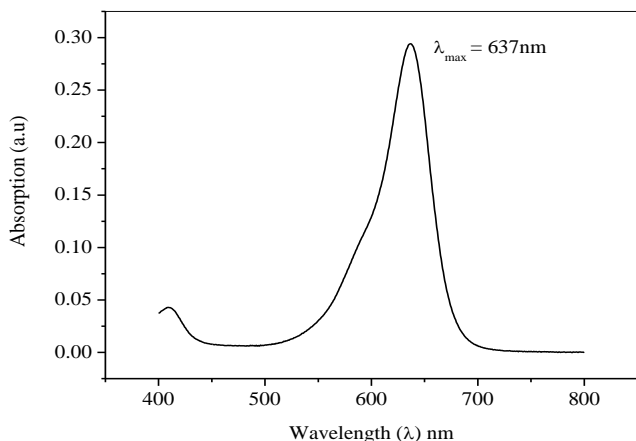


FIG. 3: UV-Vis absorption spectra of Acid blue 7 dye in water

Dye-doped gelatin films were prepared by removing silver halide from 10E75 Agfa Gaevert holographic plates and soaking in aqueous dye solutions of appropriate dye concentrations for 2 minutes time duration and dried at room temperature. The thickness of the dye film used in this study was of the order of 10 microns. These films were used without any further process for this study.

Fig.4 and Fig.5 respectively show the schematic and the photograph of the experimental setup, which were used to realize the optical phase-conjugation. In this case, the standard degenerate four-wave mixing (DFWM) configuration was used. A He-Ne laser (Coherent, 31-2140-000)

beam at 633 nm wavelength was used to generate the phase-conjugate wave from the Acid blue 7-doped gelatin films. The output beam from the laser was first split by a beam 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  $E_3$  to the dye-doped sample and to transmit the phase-conjugated signal, which was opposite to the direction of the probe beam.

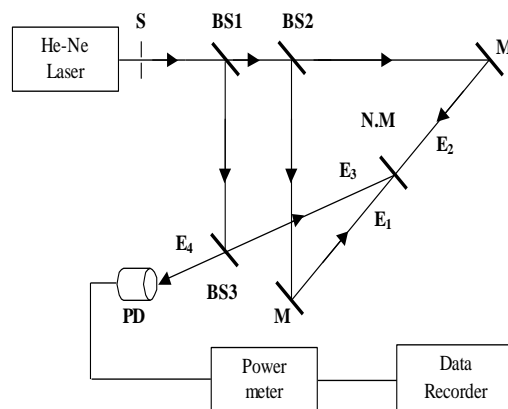


FIG. 4: Experimental set-up for the observation of PC wave, S-shutter, PD-photo detector, BS1, BS2, BS3-beam-splitters, M-mirror, N.M-nonlinear medium

The intensity of the phase-conjugated (PC) wave was measured by a photo detector fed to the digital power meter (Field Master™ GS – Coherent). The optical path lengths of all the

three beams were made equal, so that they were coherent at the sample in the DFWM geometry. The constant power ratio of the probe beam ( $E_3$ ), forward-pump beam ( $E_1$ ), and backward-pump beam ( $E_2$ ) used in this study was  $\sim 1:10:10$ .

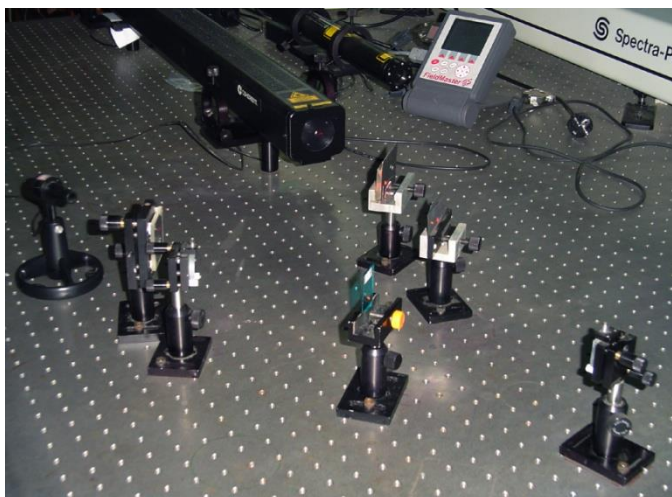


FIG.5: Photograph of the DFWM experimental set-up used for the generation of phase-conjugate waves from the Acid blue 7 dye-doped gelatin films

The spot size of each of the three (unfocussed) beams at the nonlinear medium (NM) was 1.25 mm in diameter. Interbeam angle between the probe beam and the forward-pump beam was varied between  $5^\circ$  and  $10^\circ$ .

The grating spacing ( $\Lambda$ ) of periodic lines can be determined according to the well-known formula (6) and (7). A shutter was introduced before the beam-splitter  $BS_1$  to control the start of DFWM process. The phase-conjugate reflectivity is

defined as the ratio of intensity of the phase-conjugate wave to the probe beam intensity.

## 5. Results and discussion

A systematic study has been made to investigate the influence of the dye concentration of the gelatin films on the intensity of the PC signal. Fig. 6 shows the PC reflectivity versus recording time for dye film with an optical density approximately equal to 0.7. For higher concentration of the films, the rate of formation is initially very slow, but the maximum value of PC signal obtained is high compared to the lower concentration films. These results confirm that the PC reflectivity increases with the increase in the concentration of the dye in the gelatin films. However, much higher concentrated plates need longer exposure time; this may lead mechanical instability and hence the reduction in PC reflectivity. A maximum phase conjugate reflectivity of 0.22% has been observed for probe beam intensity  $\cong 0.1W/cm^2$  and further increase of probe beam intensities leads to decrease of phase-conjugate reflectivity.

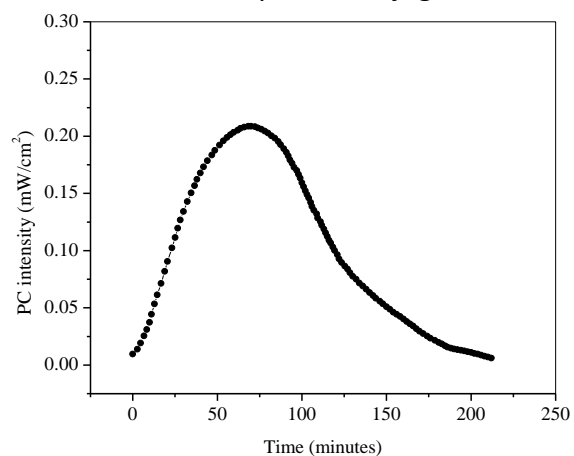


FIG. 6: Measured PC reflectivity in the Acid blue 7 dye-doped gelatin films as a function of recording time

As discussed earlier, two kinds of gratings are developed during the DFWM process towards phase-conjugate wave generation. To quantify their individual contributions, we measured the intensity of phase-conjugate signal by sequentially shutting and opening both the forward-pump ( $E_1$ ) and backward-pump ( $E_2$ ) beams incident onto the sample at various stages of the DFWM process. As shown in the Fig.7 when the reading beam  $E_1$  is switched off, the phase-conjugate signal obtained is only due to transmission grating. Similarly when the reading beam  $E_2$  is switched off, the corresponding phase-conjugate signal obtained is only from reflection grating. We measured independently both transmitted and reflected components of the phase-conjugated signal strength at three different stages of DFWM process in progress and the results are seen in the Fig.7. From this plot itself it is very clear that the main contribution to the phase-conjugate signal comes from the  $E_2$  diffraction from the transmission grating and the growth rate of transmission grating is comparatively very fast than the growth of reflection grating.

There are two main processes, which contribute effectively to the origin of phase-conjugation in acid blue 7 doped gelatin films. First one is formation of thermal grating and another is due to photo bleaching of dye molecules. We have experimentally verified these facts adopting the literatures [24] for the acid blue 7 dye-doped systems. Observation of decaying phase-

conjugate signal gives information on whether or not there is a thermal grating contribution to the phase-conjugate signal.

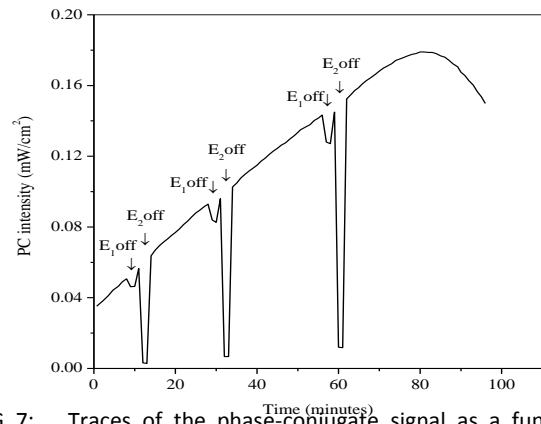


FIG. 7: Traces of the phase-conjugate signal as a function of time for Acid blue 7 dye films. A sequence of timed shutting and opening of the beams is shown in the Fig.

## 6. Conclusions

This paper reports the demonstration of optical phase-conjugation based on the degenerate four-wave mixing in Acid blue 7 dye-doped gelatin films by using a 633 nm He–Ne laser of total power 35 mW. Since this work is focused on the undergraduate level experimentation, the fundamentals of optical phase-conjugation are discussed. The individual contributions from the induced holographic gratings to the phase-conjugate signal were quantified and discussed. The inexpensive dye-doped films can easily be prepared in the students laboratories for optical phase-conjugation and other nonlinear optics related studies using low power laser sources available in the market. The authors hope this article help the students who are interested in

studies related to optical phase-conjugation, nonlinear optics and materials.

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