

## Nonlinear optical wave mixing and switching in dye-doped polymer saturable absorbers

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### Abstract

Dye-saturable absorbers are an important class of nonlinear optical materials where nonlinear interactions like four- and two-wave mixing can be observed even at low laser intensities. The modulation of the nonlinear absorption coefficient of the dye medium by an appropriate interaction of laser radiation can be utilized to generate phase conjugate signals both in the backward and the forward FWM geometries. By probing this population grating at time scales less than the dye relaxation times, transient PC signals can be observed. Here we report the generation of the forward and backward PC signals using azo dye-doped polymer films. By modulating the intensity of one of the pump beams an exchange of energy between these two PC signals was observed. The behaviour of phase conjugator as an optical switch due to this pump intensity modulation is also discussed.

**Keywords:** Optical phase conjugation, wave mixing, transient phenomena, saturable absorption, optical switching.

### 1. Introduction

Optical phase conjugation which deals with the generation of the phase conjugate (PC) wave has been investigated by many workers. This process of exactly reversing the phase and direction of an input object wave through nonlinear optical interactions has been utilized in a variety of applications like distortion correction, real-time holography, interferometry and image processing. The ability of saturable media to generate PC signals of useful intensities, in a four-wave mixing (FWM) geometry, while providing reasonably fast modulation capability has also been known.

For light inputs in which one or more of the beams is amplitude modulated the maximum modulation rate is determined by the decay rate of the medium's population. Many earlier studies<sup>1,2</sup> have been performed on time scales which are much longer than the relaxation times of saturable media. This state is called the steady state since the population distribution of the dye is expected to reach an equilibrium and there is no further redistribution of the population. On the other hand, Silberberg and Bar-Joseph<sup>3</sup> showed that the value of transient reflectivity is significantly different from the results obtained from a steady-state analysis. In particular, at an intensity much higher than the saturation intensity of the dye medium the transient reflected conjugate wave can be much stronger than the steady-state value. The duration of this intense pulse is equal to or shorter than the lifetime of the relevant transition. The transient phase conjugation phenomenon has been further investigated in both degenerate<sup>4</sup> and non-degenerate<sup>5</sup> FWM geometries. These studies have dealt with the behaviour of saturation-type non-

linearity exhibited by absorbing dyes under different input conditions. The interplay of these wave-mixing effects could be utilized for a number of interesting applications involving the modulation of the PC intensity by manipulating one of the input beams at fast rates. The fast relaxation or ground state recovery times of dye media makes it possible to demonstrate optical switching.

Here we discuss the ability of saturable dye media to generate high-transient PC reflectivities in a delayed FWM geometry where the read beam is delayed with respect to the grating writing beams. The time for which the read beam is delayed is also utilized in the observation of a self-diffraction interaction between the pump and the probe beams. The switching of energy between the backward propagating PC beam and the forward propagating self-diffracted beam is discussed.

## 2. Photoinduced dye population distribution

A typical three-energy level model (Fig. 1) is chosen to describe the saturable dyes fixed in a polymer matrix, used in our experiments as the nonlinear medium. We assume that the lifetime of level 3,  $T$ , is relatively long and that the transition from level 2 to 3 is fast and forbidden. These are reasonable assumptions for organic dyes which exhibit saturation of absorption. Under these assumptions only levels 1 and 3 can be appreciably populated. The population density in level 3,  $n_3$ , can be approximated by  $n_3 = n_0 - n_1$ , where  $n_0$  is almost equal to the total population density of saturable dye molecules and  $n_1$ , the population density in level 1. The population distribution under a uniform illumination of intensity  $I$  is governed by the rate equation

$$\frac{dn_1(t)}{dt} = -\frac{I}{I_s T} + \frac{(n_0 - n_1)}{T} \quad (1)$$

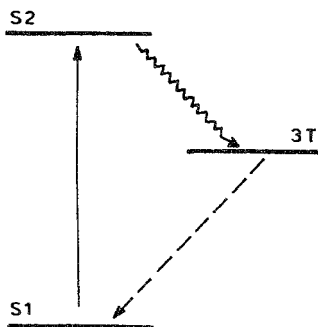


FIG. 1. Three energy-level diagram to describe a saturable absorber dye molecule; S: Singlet, T: Triplet levels.

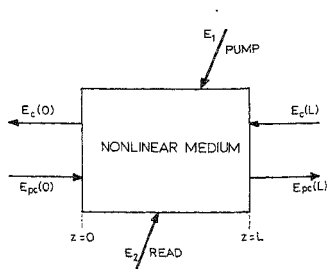


FIG. 2. Typical four-wave interaction geometry.

The saturation intensity of the dye at the line-centre is defined as  $I_s = \frac{\hbar\omega}{\sigma QT}$ , where  $\sigma$  is the absorption cross-section for the 1-2 transition,  $Q$ , the triplet yield, and  $T$ , the triplet lifetime. If the operating frequency is detuned from the line-centre frequency then the saturation intensity increases by a factor  $1 + \delta^2$  where  $\delta$  is the detuning parameter. The effect of off line-centre operation is that the saturation occurs at higher incident intensities.

### 3. Transient phase conjugation

Here we consider an FWM geometry which deals with the interaction of the three beams in which the read beam is delayed with respect to the grating recording beams, *i.e.*, the pump and the probe. This is different from the cases discussed by Silberberg and Bar-Joseph<sup>3</sup> and also by Fujiwara and Nakagawa<sup>4</sup>. Suppose that two light beams of intensities  $I_1$  and  $I_p$ , referred to as the probe and the pump beams, are interacting in a nonlinear medium. Suppose again that these intensities rise instantaneously from zero to a constant intensity at time  $t = 0$ . The read beam of the same frequency  $\omega$  is incident on the medium and is assumed to rise instantaneously from zero to constant intensity  $I_2$  at a time  $t = t_1$ .

The population of the dye at the instance  $t_1$  is given by  $n_1(t_1)$ , and is found by integrating eqn 1 from zero to  $t_1$  during which  $I_2 = 0$ . The solution is given by

$$n_1(t_1) = \frac{n_0}{I_s + I_1} (I_s + I_1) e^{-\left(1 + \frac{I_1}{I_s}\right) \frac{t_1}{T}}. \quad (2)$$

After  $t = t_1$  the population of the ground state is given once again by the integration of eqn 1 from  $t_1$  to  $t$ . The total field is a combination of the pump and read fields which form a standing wave pattern in the nonlinear medium. Now using  $n_1(t_1)$  as the initial population density the ground state population can be obtained at any time later than  $t_1$  as

$$n_1(\tilde{t}) = \frac{n_0}{(I_s + I_1 + I_2)} \left[ I_s + n_1(t_1)(I_1 + I_2) e^{-\left(\frac{I_s + I_1 + I_2}{I_s}\right) \frac{\tilde{t}}{T}} \right] \quad (3)$$

where  $\tilde{t} = (t - t_1)$  and  $n_1(t_1)$  is given by eqn 2.

The typical DFWM geometry shown in Fig. 2 is chosen for the theoretical analysis and in particular the grating picture which involves interference of the pump and probe beams is invoked. Since the medium absorbs at the incident frequency, the population is spatially modulated by making the intensity pattern incident on the dye medium spatially periodic. This modulation gives rise to an absorbing grating which in turn would diffract the read beam to form the PC signal. The path difference between pump and probe beams is assumed to be within the coherence length of the laser source.

The electric fields are taken as plane waves represented by

$$E_i(r_i, t) = \frac{1}{2} E_i(r_i) e^{-(i\omega - k_i \cdot r_i) + c.c} \quad (4)$$

where  $r_i$  is the distance measured along  $k_i$  and  $i = 1, 2, P, PC$ .

It is clear from Fig. 2 that  $E_1$  and  $E_2$  the intense pump and read waves respectively are of same frequency  $\omega$ , and are counterpropagating. The propagation axis is taken to be  $z$ -axis.  $E_P$  and  $E_{PC}$  are two weak waves of frequency  $\omega$  also counterpropagating to each other but in a direction different from the pump waves. The wave directions imply that  $k_1 + k_2 = 0$  and  $k_P + k_{PC} = 0$ . The total field can be taken as  $E = E_0 + \Delta E$ , where  $E_0 = E_1 + E_2$  and  $\Delta E = E_P + E_{PC}$ . It is assumed that the pump and read intensities are not depleted and that the slowly varying envelope approximation holds. It can be also assumed that the time derivatives can be neglected within the transit time in the phase conjugator and also within the transverse relaxation time of the dye.

Substituting the above equations into the wave equation, we get a set of equations for  $E_P$  and  $E_{PC}$  in terms of  $\alpha$  which account for the saturated absorption and dispersion of the weak fields in the presence of strong fields and  $\kappa$ , the coupling coefficient. These parameters are defined as<sup>6</sup>

$$\alpha = \left\langle -\frac{|k|}{2i} \chi_0 - \frac{|k|}{2i} \chi'_0 \left[ I_1 + I_2 + 2(I_1 I_2)^{1/2} \cos 2\phi \right] \right\rangle \quad (5a)$$

$$\kappa^* = \left\langle -\frac{|k|}{2i} \chi'_0 \left[ I_1 e^{2i\phi} + I_2 e^{-2i\phi} + 2(I_1 I_2)^{1/2} \right] \right\rangle. \quad (5b)$$

In these equations,  $\chi_0$  is the susceptibility due to pump and read beams and  $\chi'_0 = \frac{d\chi_0}{dl}$ .

The brackets  $\langle \rangle$  denote averaging over the phase term  $\phi = k_i \cdot r_i$ , which is necessary to account for the standing wave distribution generated by the counterpropagating pump and read beams.

The time-dependent phase conjugate reflectivity  $R(t)$  is given by

$$R(t) = \frac{|E_{PC}(0)|^2}{|E_P(0)|^2} = \frac{|\kappa \sin(wL)|^2}{|w \cos(wL) + \alpha \sin(wL)|^2} \quad (6)$$

where  $w = (|k|^2 - \alpha^2)^{1/2}$ .

Thus, once the susceptibility of the nonlinear dye medium due to wave mixing is known all the other parameters including PC reflectivity and their time evolution can be calculated.

For dye-saturable absorbers the imaginary susceptibility is deduced by considering an ensemble of the saturable absorbers and using a density matrix approach. If the line-centre small signal absorption cross-section is  $\sigma$  then the susceptibility is given by

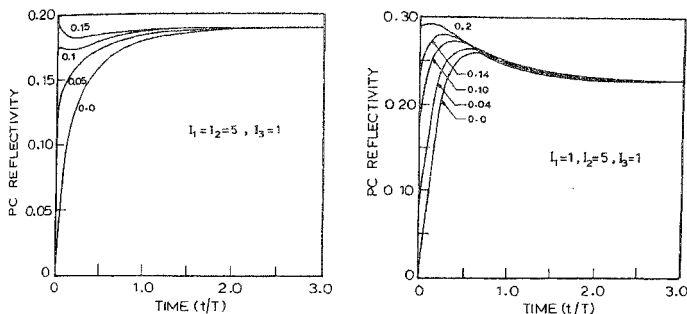


FIG. 3. Transient phase conjugate reflectivity as a function of normalized time under different intensity ratios. The numbers against each curve indicate the normalized read beam time delays.

$\chi = \frac{-i2\sigma n_1(t)}{|k|}$  which can be used in the calculation of the nonlinear optical parameters of the dye.

As mentioned earlier the interaction in which the pump and probe beams arrive simultaneously and the read beam is delayed relative to them is discussed. The read beam time delay is governed by the delay parameter  $t_1$  normalized with respect to the triplet relaxation time. As usual the spatial averaging is performed over the  $k, r$  plane by taking an arbitrary angle parameter  $\phi$  and varying it from 0 to  $2\pi$ .

The results obtained by using the above analysis for a few values of the incident intensities and saturation parameter are shown in Fig. 3. In these figures the temporal evolution of the PC reflectivity is plotted for several time delays and different intensity ratios. All the time parameters are normalized with respect to the triplet relaxation time. The small signal absorbance  $\alpha_0 L$  is taken to be 1 for ease of calculation. Here, at a saturation parameter value of 0.5 the PC reflectivity is a smoothly increasing function of the normalized time. The normalized time is the time after the arrival of the read beam, *i.e.*,  $\frac{\tilde{t}}{T} = \frac{(t-t_1)}{T}$  where  $t_1$  is the instant at which the read beam is switched ON. At nonzero time delays it can be noticed that as the readout delay is increased, the initial reflectivity increases significantly and that all the curves reach a common steady state as is expected. Very high transient PC gains can be realized by read beam time delay along with a large read to pump intensity ratio.

#### 4. Two-wave mixing

For the duration of time during which the read beam is cut off, the pump and probe beams interact and form an interference pattern which modulates the population of the

ground state and forms a grating. This is akin to two-wave mixing (TWM). This grating can assist self-diffraction and generate higher diffraction orders. The  $-1$  order diffraction beam can also be viewed as a forward phase conjugate wave<sup>6</sup>. The time evolution of the diffraction efficiency is once again governed by the susceptibility and the incident intensity and is briefly discussed here.

The total intensity in a TWM geometry is described by the intensity modulation pattern due to pump  $I_1$  and probe  $I_p$  beams, given by  $I = I_1 + I_p + 2(I_1 I_p)^{1/2} \cos(Kx)$  where  $K$  is the sinusoidal grating vector defined as  $K = \frac{2\pi}{\Lambda}$ , where  $\Lambda$  is the grating period and the grating vector is taken to be parallel to  $x$ -axis.

It is more convenient to define a modulated saturation parameter  $S(x)$ , assuming that the angle of interaction between the two beams is  $2\theta$ , as  $S(x) = S_0 + S' \cos 2\theta$ , where  $S_0 = \frac{(I_1 + I_p)}{I_s}$  and  $S' = \frac{2(I_1 I_p)^{1/2}}{I_s}$ . The absorption coefficient associated with level 1 is given by  $\alpha_s(t) = \sigma_s n_1(t)$  where  $\sigma_s$  is the small signal ground state absorption cross-section at the incident wavelength  $\lambda$  and  $n_1(t)$  is the time-dependent population of the ground state governed by the rate equations<sup>7</sup>.

The excitation grating remains harmonic at moderate intensity levels and the saturation parameter can be Fourier expanded. The first two terms in the expansion of the periodic distribution<sup>7</sup>  $\alpha_s(t)$  yield the time-dependent bias term  $\alpha_{0s}(t)$  and a modulation term responsible for the self-diffraction phenomenon,  $\alpha_{1s}(t)$ . Thus the bias and the modulation components of the absorption coefficient are given, respectively, by  $n=0$  and  $n=1$  Fourier coefficients as

$$\alpha_{0s}(t) = n_0 \sigma_s C_0 = \alpha_0 C_0; \quad (7a)$$

$$\alpha_{1s}(t) = n_0 \sigma_s C_1 = \alpha_1 C_1; \quad (7b)$$

$$\text{where } C_n = \frac{1}{\pi} \int_{-\pi}^{\pi} \left[ \frac{1 + S(x) e^{-\beta(1+S(x)) \frac{t}{T}}}{1 + S(x)} \right] dx.$$

The diffraction efficiency  $\eta$  of this absorption distribution, in either case, is obtained by analysing thick-absorbing<sup>8</sup> holographic transmission grating and is given by

$$\eta = \exp \left[ \frac{-2\alpha_{0s}d}{\cos\theta} \right] \sin^2 \left[ \frac{\alpha_{1s}d}{\cos\theta} \right] \quad (8)$$

where  $d$  is the thickness of the nonlinear medium.

The time evolution of the first-order diffraction efficiency of such a population grating is plotted in Fig. 4. The pump and probe intensities have been chosen to be less than

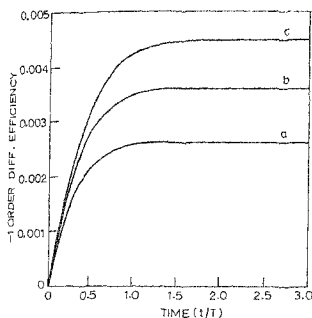


FIG. 4. Time evolution of the  $-1$  order diffraction efficiency in a two-wave mixing geometry at different intensities; a:  $I_1 = I_2 = 0.5$ ,  $I_s = 2$ , b:  $I_1 = I_2 = 1$ ,  $I_s = 2$ , c:  $I_1 = I_2 = 1$ ,  $I_s = 1$ .

the saturation intensity of the medium. It is also possible to obtain high transient forward phase conjugate efficiencies by choosing the saturation parameter to be greater than 1. The figure shows a smooth increase in the incident intensity. The steady-state values can be seen to be the same as is expected.

## 5. Experimental

The experimental layout to observe the optical wave mixing and switching effects is shown in Fig. 5. A fast risetime mechanical shutter M is used for blocking and releasing the read beam. The phase conjugate signals are detected with a fast photodiode whose output is given to dual channel storage oscilloscope (L&T Gould) through a preamplifier. The oscilloscope has a sampling rate of 400 MS/s. The stored output could then be

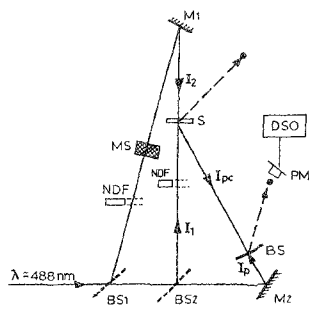


FIG. 5. Experimental layout to observe optical switching and modulation; M: Mirror, BS: Beam splitter, PM: Powermeter, DSO: Digital storage oscilloscope, MS: Mechanical shutter, NDF: Neutral density filter.

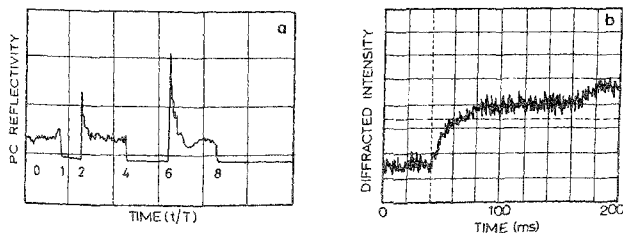


FIG. 6. Experimentally observed temporal development of: (a) Transient PC signals, (b)  $-1$  order forward PC signal.

downloaded to a computer for analysis. The 488 nm wavelength output of the argon ion laser (Spectra Physics) is used for the experiments. An azo-dye methyl yellow (N, N-dimethyl, 4-phenylazo-aniline) doped into PMMA films has been used as nonlinear media. The mechanical shutter is closed for a certain duration after the probe and the pump beams are allowed to be incident on the sample. The oscilloscope is triggered before the arrival of the read beam and the PC signal is stored. The experimental results are shown in Fig. 6. Typical transient FWM signals obtained when the shutter is closed for a certain time and released is shown in the figure. It can be clearly seen that the transient PC reflectivity is higher than the steady-state value. The increase in the transient reflectivity with the increase in the read beam delay time can also be seen. The typical TWM signals obtained in the  $-1$  order diffraction direction, once the read beam is switched OFF, are also shown. Since the intensities used for the TWM experiment are much smaller than the saturation intensity of the dye, transient effects are not observed. The figures clearly demonstrate the ability of the dye media to generate very useful transient PC signals.

## 6. Switching behaviour

One of the most interesting outcomes of the experimental observations is the exchange of energy between the phase conjugate beam and the first-order diffraction beam (Fig. 5). In this experiment, the intensities of both the PC signal and the  $-1$  order diffracted signals are recorded simultaneously by the oscilloscope. In the event of all the three beams being present simultaneously the PC signal reaches a steady-state value and the first-order signal is of very low intensity.

The mechanical shutter is then closed for a certain amount of time. This being a TWM geometry the first diffracted order grows in intensity till a steady state is reached. But once the shutter is opened after a finite time it is observed that the PC intensity increases and a transient peak is observed. The first-order signal decays simultaneously and proportionately loses in intensity as can be seen from Fig. 7. As soon as the shutter is closed again the PC signal grows to a maximum. Thus, the modulation of the read

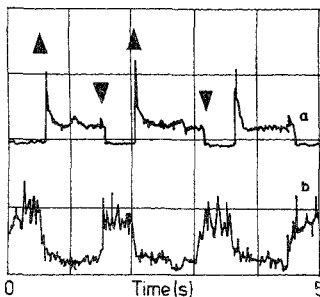


FIG. 7. Switching behaviour or energy exchange observed between PC signal (a) and first-order diffracted signal (b) due to modulation of the read beam. ▲ and ▼ show the times at which the read beam is switched ON and OFF, respectively.



beam can be used for switching the energy from the FWM to the TWM signals and the switching times are governed by the dye response times.

The behaviour observed earlier can also be explained as due to the distortion of a sinusoidal population distribution in the dye medium by an intense read beam. As explained earlier, in the TWM geometry, pump and probe beams form the grating. At intensities below the saturation intensity they form a sinusoidal population distribution which then acts as a grating. Higher diffraction orders are formed due to the forward FWM phenomenon and hence the first-order diffracted pump signal grows in intensity. As soon as the intense read beam is released it destroys the existing population modulation and distorts it for a brief period which is less than the lifetimes associated with the dye media. Thus transient peak is observed in the backward FWM signal. It should be noted that the population distribution is now governed by the pump and read intensities only as the probe is considered to be much less intense. Thus, the backward FWM process dominates and the PC intensity increases and simultaneously the  $-1$  order signal intensity decreases. Though both are third-order nonlinear optical processes the dominant mechanism is determined by the relative intensities of the beams which govern the population distribution between the energy levels of the dye medium. If all three beams are intense then the first-order diffraction is always present as the background and the energy is evenly distributed between the  $-1$  order diffraction signal and the PC signal in the steady state. Thus, the read beam intensity can be modulated to observe exchange of energy between the two processes, due to which an optical switching phenomenon can be observed.

## 7. Conclusion

Here we report the observation of transient phase conjugate signals when the read beam is operated in a pulsed manner, *i.e.*, using the delay in the arrival times of the read beam with respect to pump and probe beams. It is shown that in the absence of the read beam the TWM process leads to self-diffraction and higher diffraction orders are visible. The theoretical analysis and the corresponding experimental observations are also presented. It is also pointed out that if the read beam is more intense than the probe beam then a switching behaviour can be observed. If the read beam intensity is modulated, either backward or forward FWM becomes the dominant process. An energy exchange is observed between the backward PC signal due to FWM and the forward PC signal due to the TWM process leading to a light-induced switching phenomenon.

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