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Saturable absorbers for low power optical phase conjugation

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Abstract. Properties of a few saturable absorbers as candidates for low power optical phase conjugation (OPC) are examined. The specific systems studied are the dyes eosin, erythrosin B and Rose Bengal, doped in gelatin, polyvinyl alcohol and boric acid glass films. A method for quickly checking saturated absorption is suggested and demonstrated. Examples for OPC with these systems are cited and limitations are briefly discussed.

Keywords. Optical phase conjugation; saturable absorber; degenerate four-wave mixing; absorption spectra; triplet lifetime.

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1. Introduction

Ever since the first experimental observation of optical wavefront reversal was reported (Stepanov *et al* 1971), scientists are engaged in identifying and characterizing suitable materials for optical phase conjugation (OPC). Being a third order non-linear optical phenomenon, OPC was earlier expected only for the case of high power pulsed lasers (Yariv and Pepper 1977). However, it was later found that phenomena like photorefractive effect and saturable absorption enhance the effective third order optical susceptibility making it possible to achieve OPC using low power lasers (Huignard *et al* 1980; Liao *et al* 1978).

Saturable absorbers are known in literature, mainly in connection with Q-switching and mode-locking of pulsed lasers (Svelto 1976). These have high saturation intensities—a few MW/cm^2 and above, which can be generated only by pulsed lasers. But there are materials which can be saturated at much lower intensity levels—a few mW/cm^2 or even less. These are certain organic dyes characterized by large triplet yields. Since triplet → singlet transitions are forbidden, these molecules remain in the triplet states for long time durations, resulting in a depletion of the number of molecules available for absorption in the ground state. If the triplet lifetimes are high enough, this "bottle-necking" will result in saturable absorption at low incident intensity levels. An analysis via rate equations gives (Hercher 1967),

$$I_s = \hbar\omega/\sigma_0 t, \tag{1}$$

where I_s is the saturation intensity, σ_0 the ground state absorption cross-section and t the triplet lifetime.

Triplet lifetimes of dyes have earlier been measured by flash photolysis and it is known that the lifetimes increase as the rigidity of the solvent increases. Dyes

embedded in solid matrices like gelatin, polyvinyl alcohol (PVA), boric acid glass (BAG) etc show triplet lifetimes ranging from microseconds to seconds (e.g. Buettner 1964; Greggs and Drickamer 1960). A few dyes of the Xanthene family, viz eosin, erythrosin and Rose Bengal, which have good absorption at 514·5 nm are reported to possess lifetimes of the order of milliseconds in films of gelatin and PVA (Beuttner 1964). Therefore, these dyes can be used as saturable absorbers at the main argon ion laser wavelength.

Silberberg and Bar-Joseph (1981) demonstrated the use of eosin and erythrosin in films of gelatin for low power OPC at 514·5 nm. We have earlier reported successful use of Rose Bengal in gelatin and PVA films (Moosad *et al* 1988). In the present paper, we summarize the different considerations on these dyes as materials for low power OPC.

2. Choice of a solid matrix

Efficiency of OPC using saturable dyes depend on the proper choice of the solid matrices. The absorption spectra of the above three dyes in three solid matrices, viz gelatin, PVA and BAG are shown in figures 1(a) to (c). It is immediately evident that the BAG samples are not suitable for work at the wavelength 514·5 nm, due to the weak absorption.

3. Detection of saturable absorption

As mentioned earlier, absorption saturation depends on the ground state absorption cross-section and triplet lifetime. It is usual to measure the saturation intensity directly by observing the transmitted and incident intensities, at various incident intensities, in the case of pulsed lasers (e.g. Penzkofer 1986). This is difficult in the case of CW lasers due to damage problems. It has been observed that in the case of the above three dyes, taken in gelatin films, permanent bleaching occurs at intensities slightly above the

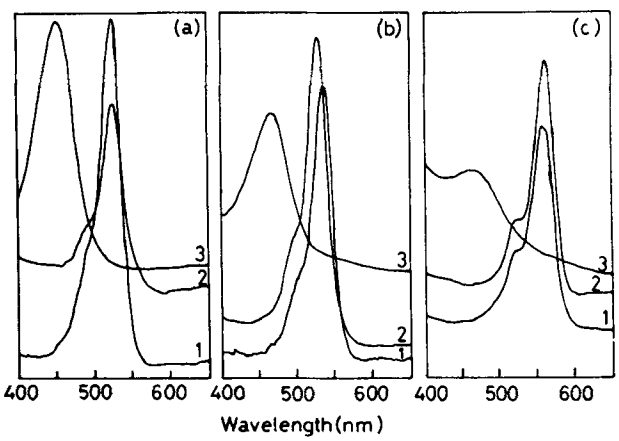

Figure 1. Absorption spectra of (**a**) eosin, (**b**) erythrosin B and (**c**) Rose Bengal in (1) gelatin, (2) PVA and (3) BAG films.

saturation intensity (Moosad *et al* 1987). This makes it difficult to distinguish the region where saturation actually occurs. This is true in the case of the PVA samples as well. A quick method of looking for the saturation of absorption is to observe the time evolution of the fluorescence emission from the sample, under constant excitation. Following the analysis of the response of fluorescent systems to transient excitation by Bartolo (1968), the fluorescence intensity in such a system for any time t, such that $0 \leqslant t \leqslant \tau, \tau$ being the duration of the excitation, can be given in the form

$$s_{1,2} = \tfrac{1}{2}\{-(p_1 + p_2 + p_3 + 2w) \pm (p_1 + p_2 - p_3 + 2w)^2 - 4wp^2\}^{1/2}, \tag{2}$$

where A, B, C, s_1 and s_2 are functions of the various lifetimes involved, which are given as

$$s_{1,2} = \tfrac{1}{2}\{-(p_1 + p_2 + p_3 + 2w) \pm (p_1 + p_2 - p_3 + 2w)^2 - 4wp_2\}^{1/2}, \tag{3}$$

$$A = wN_0p_3/s_1s_2, \tag{4}$$

$$B = wN_0(s_1 + p_3)/s_1(s_1 - s_2) \tag{5}$$

and

$$C = wN_0(s_2 + p_3)/s_2(s_2 - s_1) \tag{6}$$

p_1, p_2 and p_3 are the rates of fluorescence emission, triplet cross-over and phosphorescence emission respectively. w is the constant rate of excitation and N_0 is the ground state number density.

In systems which are expected to behave as saturable absorbers at low intensities, p_2 and p_3 have to be sufficiently high. p_1 is generally of the order of 10^8. For a case with $p_2 = 10^7$ and $p_3 = 2 \times 10^4$ with an excitation rate $w = 10^4$, the fluorescence intensity for a time duration of 5 ms would look like that shown in figure 2(a). The actual observation in the case of eosin in a gelatin film, excited by the 514·5 nm line of argon ion laser, at 500 mW power, is shown in figure 2(b). The appearance of the initial peak indicates its capability to function as a saturable absorber at this intensity level. Figure 2(c) shows the emission from Rhodamine 6G under the same conditions and 2(d) shows that from eosin at a much lower incident intensity. In either case the initial peak is absent indicating that saturation does not occur. These oscillograms are recorded using the set-up shown in figure 3.

4. Determination of triplet lifetimes

Triplet lifetimes in these systems can be determined directly by observing the phosphorescence emission. It is known that the phosphorescence emission is exponential in time, with a rate constant equal to the triplet lifetime and hence can be obtained from a log (intensity) vs time plot.

5. Low power OPC using saturable absorber films

We have already reported (Moosad *et al* 1988) OPC in the above dyes in films of gelatin and PVA. OPC can be achieved in the usual counter-propagating geometry of the

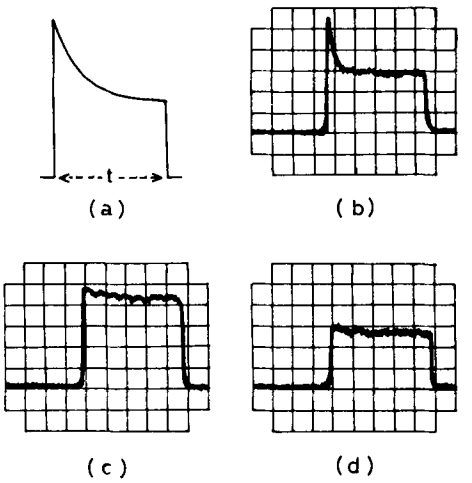

Figure 2. (**a**) Calculated fluorescence emission from a sample with $p_1 = 10^8$, $p_2 = 10^7$, $p_3 = 2 \times 10^4$ and $w = 10^4$, for a time $t = 5$ *ms*. (**b**) Fluorescence emission from an eosin/gelatin film, excited by 514·5 nm of argon ion laser at 500 mW power. Sweep rate: 1 ms/div. (**c**) Fluorescence emission from a Rhodamine 6G/gelatin film under the same conditions. (**d**) Fluorescence emission from an eosin/gelatin film, excited by the above laser at about 20 mW power.

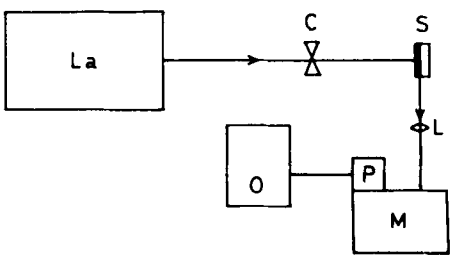

Figure 3. Experimental set-up for recording the oscillograms of the fluorescence emission, shown in figures 2(b) to (d). C, chopper; S, sample; L, lens; P, photomultiplier tube; M, monochromator; O, oscilloscope; La, laser.

beams as shown in figure 4. Phase conjugate reflectivities of 10^{-5}–10^{-3} are achieved. The beam spot can be observed on a white screen kept in the retro-direction of the probe beam. Detection can be done by a PMT preceded by a filter to cut-off the intensity due to fluorescence. An example of the variation of the OPC signal with the laser power is shown in figure 5. The reflectivities in these cases are found to be varying in accordance with the theory proposed by Abrams and Lind (1978a, b).

6. Variation of OPC efficiency with wavelength

Since saturation intensity depends on the ground state absorption cross-section, OPC efficiency of saturable absorbers is expected to roughly follow the absorption spectrum. As an example, we give the relative OPC signal strengths at the different argon ion laser wavelengths, in the case of erythrosin B in PVA film, in figure 6.

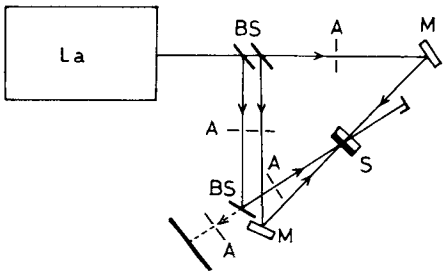

Figure 4. Beam geometry for OPC by degenerate four-wave mixing. M, mirror; BS, beam splitters; S, sample; A, aperatures; La, laser.

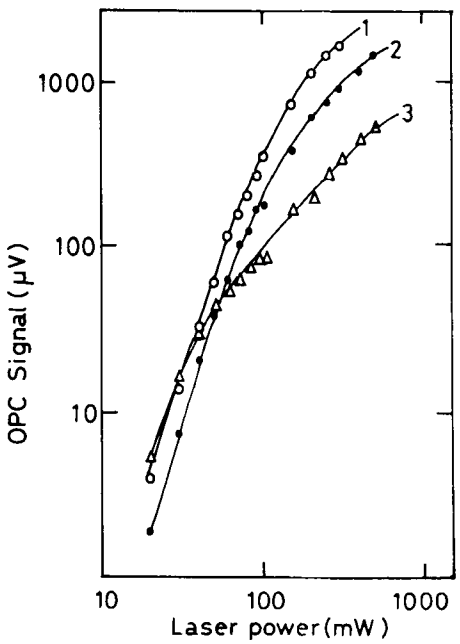

Figure 5. OPC signals from (1) erythrosin B, (2) Rose Bengal and (3) eosin in PVA films vs laser power.

7. Limitations

The main limitations in using these systems for OPC applications come from two effects: (i) permanent bleaching of the dyes due to continued exposure to the laser beams. This limits the usable intensity levels to those below the saturation intensities which in turn limits the maximum efficiency achievable. (ii) Wash-out effects due to vibration pick-ups: since these species get saturated in a time scale of milliseconds, vibrations will cause wash-out effects, which will also limit the available efficiency. For obtaining more efficiencies, good vibration isolation set-ups will be necessary.

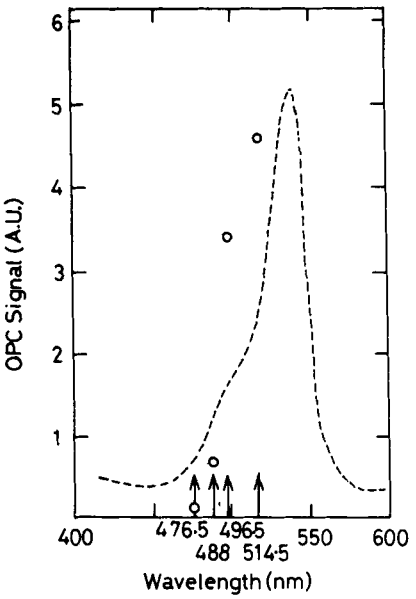

Figure 6. OPC signals from erythrosin B/PVA film, at the different argon ion laser wavelengths, plotted in the background of the absorption spectrum (broken line).

8. Conclusion

In this paper we have summarized the various considerations on saturable absorbers as materials for low power OPC. The case of three dyes, eosin, erythrosin B and Rose Bengal in films of gelatin, PVA and BAG are studied. An easy method of looking for saturated absorption is suggested and demonstrated. Examples for OPC with saturable absorbers are cited and limitations are briefly mentioned.

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