Invited Paper

Polarization Properties of Phase Conjugation by Degenerate Four-Wave Mixing in Saturable Absorbers

R. W. Boyd, D. J. Gauthier, M. Kauranen, M. S. Malcuit, and W. R. Tompkin
The Institute of Optics, University of Rochester, Rochester, New York 14627

Abstract

Dye molecules held rigidly in solid hosts display unusual polarization properties that can be exploited to produce nearly perfect vector phase conjugation with high sensitivity.

Introduction

Let us begin by distinguishing between vector phase conjugation (VPC) and scalar phase conjugation. If a field $\vec{E}_{inc}(\vec{r},t) = \vec{E}_o(\vec{r})\, e^{-i\omega t} + c.c.$ falls onto an ideal phase conjugate mirror (PCM), the field leaving the mirror will be given by $\vec{E}_{pc}(\vec{r},t) = \mu\, \vec{E}_o^{\,*}(\vec{r})\, e^{-i\omega t} + c.c.$, where μ is the amplitude reflectivity. In order to determine the significance of replacing the field amplitude $\vec{E}_o$ by its complex conjugate, we represent $\vec{E}_o$ as the product of a complex polarization unit vector $\vec{\varepsilon}$, a slowly varying field amplitude A_o, and an exponential phase factor $e^{i\vec{k}\cdot\vec{r}}$, i.e., by

$$\vec{E}_o(\vec{r}) = \vec{\varepsilon}\, A_o(\vec{r})\, e^{i\vec{k}\cdot\vec{r}} \quad . \tag{1}$$

By taking the complex conjugate of $\vec{E}_o(\vec{r})$, we see that the consequences of the phase-conjugation process are three-fold: $\vec{k}$ is replaced by $-\vec{k}$ implying inversion of the wavevector, A_o is replaced by A_o^* implying reversal of the wavefront, and $\vec{\varepsilon}$ is replaced by $\vec{\varepsilon}^{\,*}$ implying polarization conjugation, for example, RHC (right-hand circular) light remains RHC in reflection from a PCM instead of turning into LHC, as in the case of reflection from an ordinary mirror. Many devices which are known as PCM's do not possess this third property. We will refer to such devices as scalar PCM's and to devices that possess all three properties as vector PCM's.

In order to determine the conditions under which perfect vector phase conjugation will occur, it is necessary to consider the vector nature of the $\chi^{(3)}$ susceptibility used in the degenerate four-wave mixing process leading to phase conjugation. Terhune and Maker[1] have shown that for an isotropic nonlinear material, the third-order nonlinear polarization can always be written in the form

$$\vec{P}^{(3)} = A(\vec{E}^* \cdot \vec{E})\vec{E} + \frac{1}{2}B(\vec{E} \cdot \vec{E})\vec{E}^* , \tag{2}$$

where A and B are parameters that characterize a particular nonlinear material. Note that the second term has the vector nature of $\vec{E}^*$ and hence directly leads to VPC whereas the first term has the vector nature of $\vec{E}$ and leads to VPC only under special conditions. If A were to vanish, the four-wave mixing process would automatically lead to perfect polarization conjugation, but for typical nonlinear optical materials A is nonzero, as listed here for common nonlinear optical interactions:

B/A = 0 for electrostriction,

B/A = 1 for nonresonant electronic nonlinearities, and

B/A = 6 for orientational Kerr effect.

For certain two-photon allowed transitions A vanishes;[2] however, since A is non-zero in general, special care must be taken to achieve VPC.

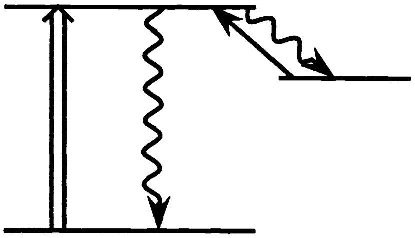

Fig. 1. Energy-level diagram showing the relevant optical interactions in fluorescein-doped boric acid glass.

VPC using Fluorescein-Doped Boric Acid Glass

We have been working on the development of highly sensitive nonlinear optical materials that can be used for phase conjugation. The technique we use is to dope organic molecules into glass hosts.[3,4] Due to the presence of triplet trap levels, in many cases such composite materials act as low-intensity saturable absorbers. The excitation sequence for this class of materials[5] is shown in Fig. 1. Following optical excitation, there is an appreciable chance that population will be transferred to the trap level. The material we have studied is fluorescein-doped boric acid glass which has a ground-state recovery time τ of ~0.1 sec. This long ground-state recovery time implies that the saturation intensity is low, since $I_s = \hbar\omega/\sigma\tau$ where σ is the absorption cross section, and hence that the $\chi^{(3)}$ susceptibility is large, since

$$\chi^{(3)} = \frac{-i n^2 c^2 \alpha_o}{96\pi^2 \omega I_s} , \tag{3}$$

where α_0 is the unsaturated absorption coefficient. In Table I we compare the nonlinear optical properties of fluorescein-doped boric acid glass to those of several other nonlinear optical materials. Note that $\chi^{(3)}$ for fluorescein-doped boric acid glass is approximately 10^{12} times larger than that of CS_2 and that its response time is ~10^{12} times longer than that of CS_2.

Table I, Nonlinear Optical Properties of Several Different Materials.

Material	$\chi^{(3)}$	I_s	τ
fluorescein-doped glass	1 esu	30 mW/cm^2	0.1 s
ruby	10^{-5} esu	1.5 kW/cm^2	3 ms
fused silica	2×10^{-14} esu	-	1 fs
CS_2	2×10^{-12} esu	-	2 ps
sodium (off resonance)	10^{-8} esu	-	~1 ns

We have studied the polarization properties of phase conjugation in fluorescein-doped boric acid glass. One way of performing VPC even for a material in which A is nonzero is to choose the pump waves to be circularly polarized and counterrotating.[6] To see why this interaction leads to VPC, we recall that four-wave mixing can be viewed as the simultaneous annihilation of two pump photons with the creation of a signal and conjugate photon. Since the two pump waves are counterrotating, the absorption of two pump photons removes no angular momentum from the input fields. Consequently, the signal and conjugate photons must be emitted with equal and opposite angular momenta, implying that the VPC process is perfect. We have studied this process using the setup shown in Fig. 2. The quarter-wave plate is used as a known polarization distorter. By controlling the orientation angle θ of the waveplate, we are able to impress a known polarization aberration onto the beam. Our results are shown in Fig. 3a. For the case of counterrotating pump beams, the intensity of that part of the conjugate field whose polarization is the complex conjugate of the probe beam (the "good" component) is seen to be much larger than that of the orthogonal component (the "bad" component) for any orientation of the waveplate and hence for any state of polarization of the probe beam at the PCM.

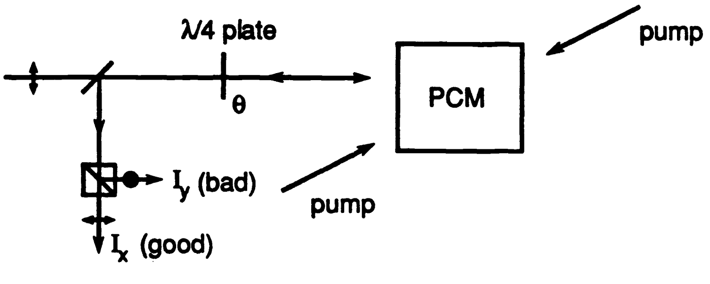

Fig. 2. Experimental setup.

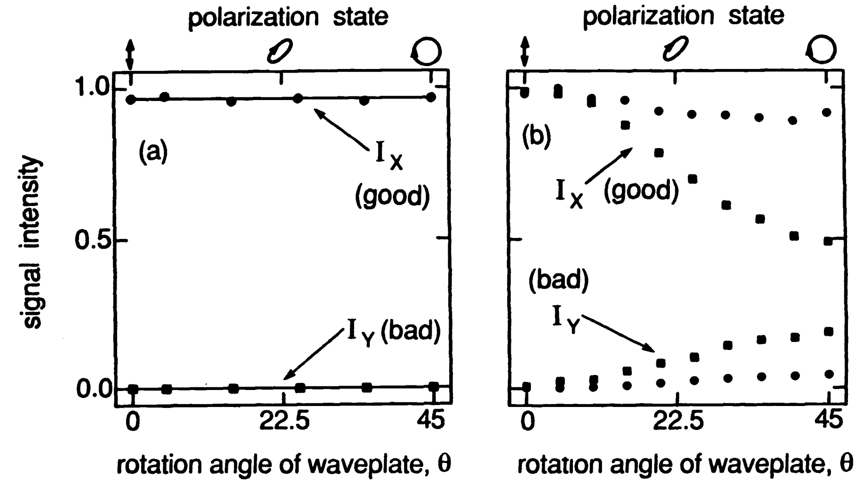

Fig. 3. Intensities of the polarization components parallel (good) and perpendicular (bad) to the initial polarization direction as functions of the rotation angle θ of the quarter-wave plate for (a) counterrotating and (b) linear pump beam polarizations.

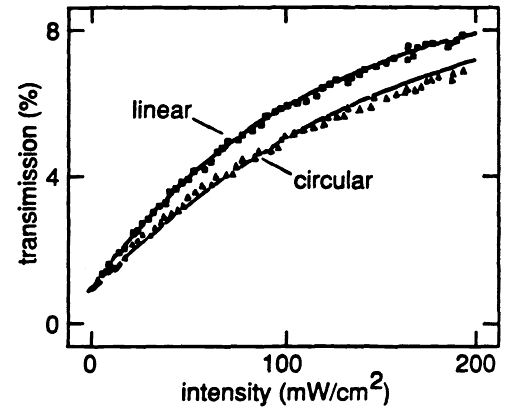

Fig. 4. Transmission of fluorescein-doped boric acid glass plotted as a function of the laser intensity for linearly and circularly polarized light at a wavelength of 457.9 nm. (After Kramer *et al.*, 1986)

Another technique for VPC in fluorescein-doped boric acid glass results from the fact that dye molecules held rigidly in solid hosts display unusual polarization properties.[4,7] An example of such behavior is that fluorescein-doped boric acid glass displays polarization-dependent saturation. In Fig. 4 we show the percent transmission of a sample of fluorescein doped boric acid glass as a function of the incident laser intensity. We see that the absorption saturates more rapidly for the case of linearly polarized light than for circularly polarized light. This behavior is quite unusual in that typically saturation behavior is independent of the state of polarization. For example, polarization-independent saturation is displayed by isotropic absorbers, such as by atoms or by dye molecules in liquid solutions which are free to reorient rapidly. The unique polarization properties of rigidly held dye molecules can be exploited to produce VPC even for the case of linear and parallel pump-wave polarizations. We have studied the polarization properties of phase conjugation for this case and the results, shown in Fig. 3b, are rather surprising. For low pump intensities, (i.e., well below the saturation intensity) we see that the polarization properties are very imperfect (shown by the data plotted as squares in Fig. 3b). However, when we increase the pump intensities to values comparable to the saturation intensity, we see that nearly perfect VPC is obtained (shown by the data plotted as circles in Fig. 3b).

We have developed the following theoretical model to explain these results. Due to the extremely low saturation intensity (30 mW/cm^2) of fluorescein-doped boric acid glass, we must include higher-order contributions to the nonlinear polarization. Since the dye molecules are held fixed, we assume that the molecules can absorb only that component of the incident radiation which is polarized along the chromophore of the molecule, and we perform an angular average over all possible orientations of the molecules. These assumptions lead to a polarization of the form

$$\vec{P} = \frac{K}{|E_s|^2}\int d\Omega \, \frac{\vec{\mu}(\vec{\mu}\cdot\vec{E})}{1+|\vec{\mu}\cdot\vec{E}|^2/|E_s|^2}\,, \tag{4}$$

where the unit vector $\vec{\mu}$ points along the direction of the chromophore, $|E_s|^2$ is the saturation intensity for light polarized along the chromophore, $\int d\Omega$ denotes an orientational average over solid angle, and K is a proportionality constant that depends on the particular molecular system. For the model illustrated in Fig. 1, the saturation intensity and proportionality constant are given by $|E_s|^2=\gamma\hbar^2(\Delta_1^2+\Gamma_1^2)/4\Gamma_1\mu^2$ and $K=(N/4\pi)[\gamma\hbar(\Delta_1+i\Gamma_1)/4\Gamma_1]$, where γ^{-1} is the ground-state recovery time, Δ_1 is the detuning of the applied field from the singlet-singlet transition frequency, $(\Gamma_1)^{-1}$ is the dipole-relaxation time for the singlet-singlet transition, and N is the number density of molecules. We now let the total field amplitude be the sum of the pump and probe fields so that $\vec{E}=\vec{E}_o+\vec{E}_p$ and linearize the expression for $\vec{P}$ about the pump field strength $\vec{E}_o$ to obtain that part of the nonlinear polarization that can act as a source for the conjugate field:

$$\vec{P}_c = \frac{-K}{|E_s|^4}\int d\Omega \, \frac{\vec{\mu}(\vec{\mu}\cdot\vec{E}_o)^2(\vec{\mu}\cdot\vec{E}_p^*)}{1+|\vec{\mu}\cdot\vec{E}|^2/|E_s|^2}\,. \tag{5}$$

We specialize to the case of equal pump wave intensities with linear and parallel polarizations and allow the probe field to have x and y polarization components:

$$\vec{E}_o = A_o(e^{ikz}+e^{-ikz})\vec{x} \qquad \text{and} \qquad \vec{E}_p = (E_x\vec{x}+E_y\vec{y})e^{ikz}\,. \tag{6}$$

We then find that we can perform the angular average analytically, and obtain the result:

$$\vec{P}_c = P_x \vec{x} + P_y \vec{y} \tag{7a}$$

where

$$P_x = E_x^* \left\{ \frac{-2\pi K}{\alpha^2 |E_s|^2} \left[\frac{2\alpha^2+3}{\alpha^2+1} - \frac{3}{\alpha}\tan^{-1}\alpha \right] \right\} \quad \text{and} \quad P_y = E_y^* \left\{ \frac{-\pi K}{\alpha^2 |E_s|^2} \left[\frac{2\alpha^2+3}{\alpha}\tan^{-1}\alpha - 3 \right] \right\} \tag{7b}$$

where $\alpha = 2A_0 \cos(kz)/|E_s|$. We then derive coupled amplitude equations, which for the case of a thin medium take the simple form

$$\frac{d\vec{A}_c}{dz} = 2\pi i k \vec{P}_{pm} \quad \text{where} \quad \vec{P}_{pm} = \frac{1}{\lambda} \int_0^{\lambda} \vec{P}_c e^{ikz} dz. \tag{8}$$

Note that we have extracted the phase matched part of $\vec{P}_c$ by performing a spatial average. Finally, we decompose the output field into its good and bad components by introducing the unit vector $\vec{\varepsilon}_g$ describing the state of polarization conjugate to that of the probe field and the unit vector $\vec{\varepsilon}_b$ describing the polarization orthogonal to $\vec{\varepsilon}_g$, that is,

$$\vec{A}_c(0) = A_g \vec{\varepsilon}_g + A_b \vec{\varepsilon}_b \quad \text{where} \quad \vec{\varepsilon}_g = \vec{A}_p^* / |A_p| \quad \text{and} \quad \vec{\varepsilon}_b \cdot \vec{\varepsilon}_g^* = 0 \ . \tag{9}$$

The results of this calculation are shown in Fig. 5a, where we plot the reflectivity associated with the good and bad components as functions of the pump intensity. We see that the good component has a broad maximum near the the saturation intensity, and that the bad component has a sharp dip at nearly the same intensity. The sharp dip in the bad component can be understood intuitively from the following point of view. Dipoles oriented along the polarization direction of the pump waves respond more strongly but saturate more quickly than dipoles oriented perpendicular to this direction. Hence both linear components of the response become equal for some intensity near the saturation intensity. The solid circles are experimental data points. Note that there is good qualitative agreement between theory and experiment, but that the dip in the bad component is not as pronounced as that predicted by theory. Even still for intensities slightly above the saturation intensity the good component is two orders of magnitude larger than the bad component.

In Fig. 5b we have modified the theoretical analysis by assuming that there is a 30% wide spread of saturation intensities. Note that the agreement is now extremely good. We believe that site-to-site variations[5,8] in the glass host leads to this spread of saturation intensities.

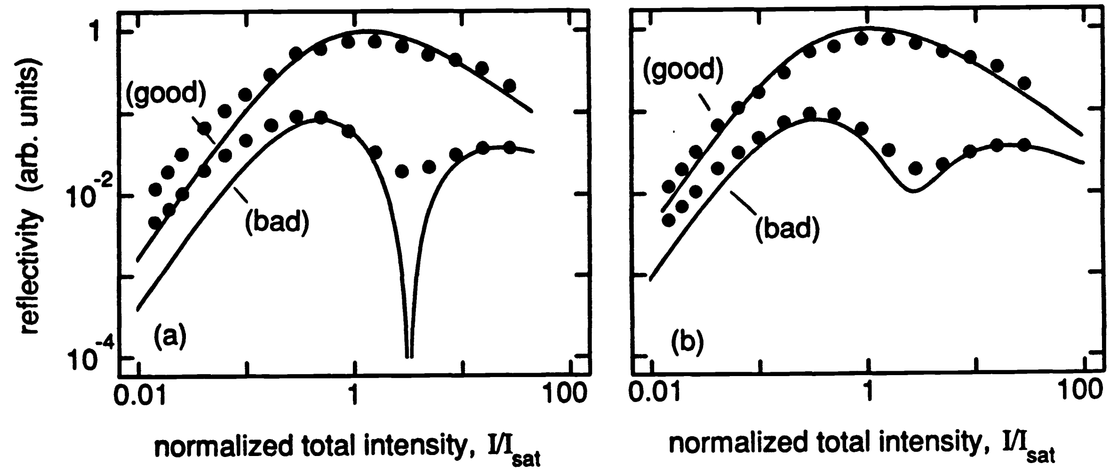

Fig. 5. Reflectivity associated with each polarization component plotted as a function of normalized pump intensity I/I_s.

Summary

Rigidly held dye molecules display unusual polarization behavior. This behavior can be exploited to produce nearly perfect VPC using linearly polarized pump waves. Fluorescein-doped boric acid glass has a very small saturation intensity and hence is useful for phase conjugating weak signals. We achieved ~0.5% phase-conjugate reflectivity using fluorescein-doped boric acid glass. The phase conjugate reflectivity could be improved substantially through the use of a dye/host system that does not display excited state absorption. To improve the vector character of the phase-conjugation process, one should use a dye/host system with smaller site-to-site variations.

Acknowledgements

We acknowledge fruitful discussions with Prof. J. E. Sipe. This work was supported by the U.S. Office of Naval Research (contract number N 00014-86-K-0746), and by a NATO travel grant.

References

1. P. D. Maker and R. W. Terhune, Phys. Rev. **137**, A801 (1965).
2. G. Grynberg, Opt. Commun. **48**, 432 (1984); M. Ducloy and D. Bloch, Phys. Rev. A **30**, 3107 (1984); and M. S. Malcuit, D. G. Gauthier, and R. W. Boyd, Opt. Lett. **13**, 663 (1988).
3. T. A. Shankoff, Appl. Opt. **8**, 2282 (1969); Y. Silberberg and I. Bar-Joseph, Opt. Commun. **39**, 265 (1981); I. Bar Joseph and Y. Silberberg, Opt. Commun. **41**, 455 (1982); M. A. Kramer, W. R. Tompkin and R. W. Boyd, J. Lumin. **31/32**, 789 (1984); H. Fugiwara and K. Nakagawa, Opt. Commun. **55**, 386 (1985); T. Todorov, L. Nikolova, N.

Tomova and V. Dragostina, Opt. Quantum Electron. **13**, 209 (1981); IEEE J. Quantum Electron. **QE-22**, 1262 (1986); and W. R. Tompkin, R. W. Boyd, D. W. Hall, and P. A. Tick, J. Opt. Soc. Am. B. **4**, 1030 (1987).

4. M. A. Kramer, W. R. Tompkin and R. W. Boyd, Phys. Rev. A. **34**, 2026 (1986).
5. G. N. Lewis, D. Lipkin, and T. T. Magel, J. Am. Chem. Soc. **63**, 3005 (1941); and M. Frakowiak and H. Waylerys, Acta Phys. Polon. **18**, 93 (1959).
6. V.N. Blaschuk, B. Ya. Zel′dovich, A. V. Mamaev, N. F. Pilipetsky, and V. V. Shkunov, Sov. J. Quantum Electron. **10**, 356 (1980); and G. Martin, L. L. Lam, and R. W. Hellwarth, Opt. Lett. **5**, 185 (1980).
7. M. Montecchi, M. Settembre, and M. Romagnoli, J. Opt. Soc. B **5**, 2357 (1988).
8. T. Tomashek, Ann. Phys. **67**, 622 (1922); D. W. Greggs and H. G. Drickamer, J. Chem. Phys. **35**, 1780 (1960); and V. A. Pilipovitch and B. T. Sveshnikov, Opt. Spektrosk. **5**, 290 (1958).