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Optical Phase Conjugation in Iodine Solutions

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Abstract

Results of experimental investigations on the optical phase conjugation in iodine solutions are reported in this paper. Degenerate four wave mixing is performed at two different wavelengths - one within the resonant regime and the other away from it. The third-order susceptibility $\chi^{(3)}$ shows an enhancement by two orders of magnitude in the resonant regime.

Key words: optical phase conjugation, degenerate wave mixing

Introduction

Iodine, a simple acceptor molecule forms charge transfer (CT) complexes with many donors (Mulliken 1969). CT complex of iodine in the solvent 1,2 dibromoethane is studied here for its nonlinear optical (nlo) behaviour, by Degenerate Four Wave Mixing (DFWM) experiments.

Theory

The nonlinear polarization $\vec{P}_{NL}$ generated in a four wave mixing experiment is given by (Fisher 1983).

$$\vec{P}_{NL}(\omega_4) = \epsilon_0 \chi^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3)\vec{E}_1(\omega_1)\vec{E}_2(\omega_2)\vec{E}_3(\omega_3) \quad (1)$$

$\chi^{(3)}$ is the third order susceptibility tensor and ϵ_0 is the permittivity of free space.

A fourth wave is generated ($\vec{E}_4$) at ω_4 due to $\vec{P}_{NL}$. The energy and momentum conservation conditions are represented by:

$$\omega_4 = \omega_1 + \omega_2 + \omega_3 \quad (2)$$

$$\vec{k}_1 + \vec{k}_2 - \vec{k}_3 - \vec{k}_4 = 0 \quad (3)$$

where ω_1 and $\vec{k}_1$ are the respective angular frequency and propagation vectors of the four waves with electric fields $\vec{E}_1$ referred to above. $\vec{E}_1, \vec{E}_2$ and $\vec{E}_3$ are the electric fields associated with the input waves. Two of the input waves are called pump waves and the third is the probe. The output $\vec{E}_4$ that is generated is the phase conjugate replica of the probe. In the case of DFWM experiment, all the frequencies ω_1 are equal (ω) and $\vec{k}_2 = -\vec{k}_1$. So $\vec{k}_4 = -\vec{k}_3$, i.e., the phase conjugate beam travels in a direction opposite to that of the probe. The phase conjugate reflectivity R is the ratio of the intensities of the conjugate to the probe (I_4/I_3). In the resonant part of the spectrum, the third-order susceptibility $\chi^{(3)}$ is estimated from the phase conjugate reflectivity R by the following expression (Caro & Gower 1982)

$$\chi^{(3)} = \frac{4c^2n^2\epsilon_0\alpha}{3\omega e^{-\alpha L sec\theta}(1 - e^{-\alpha L})} \times \frac{R^{1/2}}{I_1} \quad (4)$$

Here L is the thickness of the sample, θ is the angle between pump 1 and the probe beam, c is the velocity of light, α is the absorption coefficient, n is the refractive index of the material, and I_1 is the intensity of pump 1.

When the absorption coefficient $\alpha \to 0$, this expression gets modified to

$$\chi^{(3)} = \frac{4c^2n^2\epsilon_0}{3\omega L} \times \frac{R^{1/2}}{I_1} \quad (5)$$

Experiment

The experimental setup is the same as given in (Vijaya et al. 1990). Two laser wavelengths are employed here - (i) second harmonic of a Nd: YAG laser (532 nm) and (ii) fundamental of a Ruby laser (694 nm). The solvent used is 1,2 dibromoethane. The concentration range studied is 1×10^{-3} to 1×10^{-2} moles/litre. The iodine solution exhibits a visible absorption spectrum consisting of a band peaking at 490 nm. There is considerable absorption at 532 nm and practically no absorption at 694 nm. This study thus enables us to find the resonance contribution to $\chi^{(3)}$.

The intensity of the phase conjugate beam (I_4) is measured as a function of (i) pump beam intensity and (ii) concentration of iodine solution at the two wavelengths of study. The pump and probe intensities (I_1, I_2 and I_3) are measured for determining the reflectivity and $\chi^{(3)}$.

Results

All the samples give good phase conjugate reflectivities R varying between 0.1% and 0.3%. A reflectivity of 0.14% is obtained at a concentration of 3.9 $\times 10^{-3}$ moles /litre of iodine solution at 532 nm. The pump and probe energies for this reflectivity are 7.6 mJ and 0.9 mJ respectively with a pulse width of 40 ns and a beam area of 0.314 cm^2.

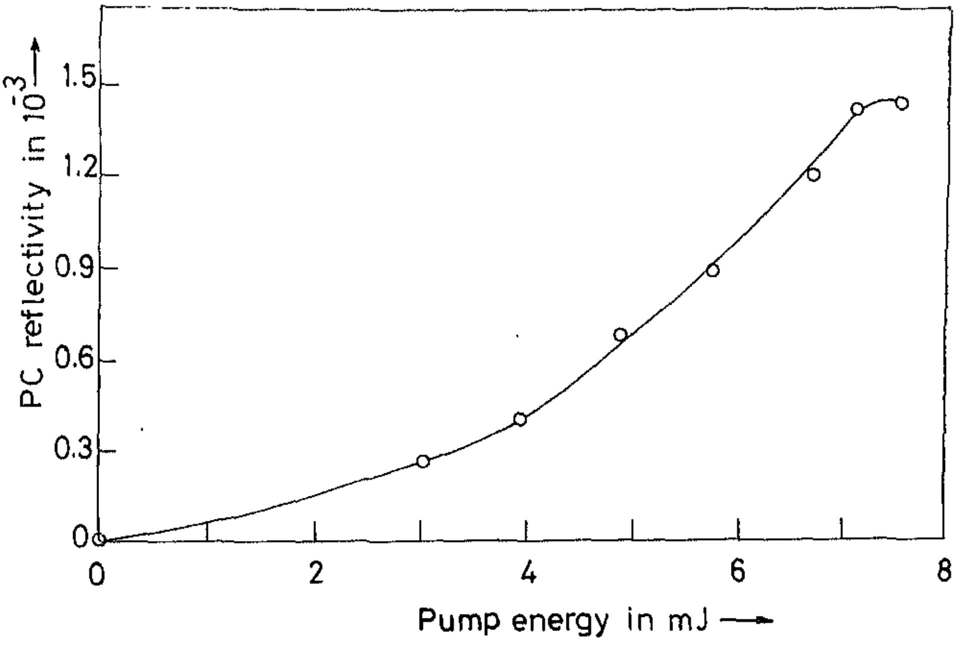

Figure 1. Phase conjugate reflectivity as a function of pump beam energy at 532 nm.

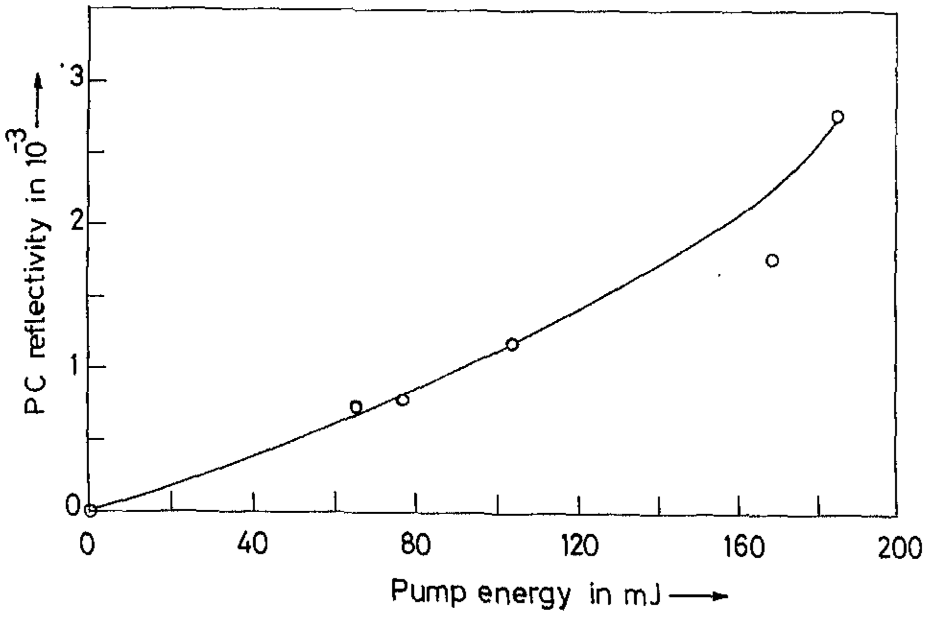

Figure 2. Variation of phase conjugate reflectivity with pump beam energy at 694 nm.

Fig. 1 shows the variation of phase conjugate reflectivity as a function of pump energy for this sample. The value of αL is 0.65 (α is taken at 532 nm). This is observed to be the optimum molar concentration that gives the maximum phase conjugate reflectivity. For concentrations that are either more or less than this concentration, the phase conjugate reflectivity is found to be lower.

A reflectivity value of 0.28% is obtained at a concentration of 6.5×10^{-3} moles/litre at 694 nm for pump and probe energies of 185 mJ and 13.5 mJ respectively, the laser pulse width being 20 ns and the beam area is 0.196 cm^2. In Fig. 2, we depict the variation of phase conjugate reflectivity for this sample at various pump energies. In contrast to the results at 532 nm, it is seen from these measurements that the reflectivity increases with increase in the concentration of the solution.

Discussion

In the calculation of $\chi^{(3)}$ from equations (4) or (5), the data of $R^{1/2}$ vs I_1 are fitted to a straight line using a least squares method. For the studies at 532 nm, Equation (4) is used and Equation (5) is the appropriate one at 694 nm. The values of $\chi^{(3)}$ thus obtained are listed in Table 1.

From Table 1, we may compare the values of $\chi^{(3)}$ of iodine in 1,2 dibromoethane at the two wavelengths. Clearly the resonance contribution enhances the $\chi^{(3)}$ by two orders of magnitude at 532 nm, in comparison to that at 694 nm which lies in a nonabsorbing region.

One may also note that Equation (4) predicts a maximum reflectivity for $\alpha L = 0.7$ (Basov 1988), which agrees well with our result (0.65).

Table 1. Third-order susceptibility of Iodine

Laser Wavelength (nm)	Concentration (moles/litre)	$\chi^{(3)}(m^2/V^2)$
532	3.9×10^{-3}	1.17×10^{-17}
694	6.5×10^{-3}	9.41×10^{-20}

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References

Basov, N.G.(Ed).1988,Phase conjugation of laser emission, Proc.Lebedev Phys.Inst., Acad. Sci. USSR, **172**, Nova Science, Commack.

Caro, R.G. and Gower, M.C. 1982,IEEE J.Quant.Electr.**QE-18**,1376.

Fisher, R.A. 1983, Optical phase conjugation, Academic, New York.

Mulliken, R.S. 1969, Molecular complexes, Wiley-Interscience,New York.

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