Optical phase conjugation in a soluble polymer

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Abstract. Degenerate four-wave mixing of picosecond light pulses in polydiacetylene dissolved in toluene has been observed. Phase conjugate reflectivities of over 100% were obtained. The origin of the nonlinear interaction was found to be a thermal refractive index change.

Subject terms: degenerate four-wave mixing; polymer; polydiacetylene; phase conjugation; picosecond.

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CONTENTS

1. INTRODUCTION

Nonlinear optical phase conjugation has found possible applications in fields as varied as laser fusion and image processing.[1]

Phase conjugate waves can be generated by a range of techniques including stimulated Brillouin scattering and degenerate four-wave mixing.[2] Both these techniques require a nonlinear medium with large third-order susceptibility $\chi^{(3)}$.

Organic dyes have been used to produce high phase conjugate reflectivities in the picosecond regime.[3] The possibility of finding other conjugators exhibiting large nonlinear susceptibilities provides motivation for investigating organic polymers as a new source of picosecond phase conjugate reflectors.

Polydiacetylenes have been shown to exhibit large values of $\chi^{(3)}$ both in crystalline form[4] and in solution.[5] While third-order frequency mixing has been measured in these materials, to our knowledge no observation of phase conjugation has been previously observed.

2. PICOSECOND EXPERIMENTAL TECHNIQUES

To obtain the high temporal resolution and the large peak intensities required for degenerate four-wave mixing at modest pulse energies, it is necessary for the laser pulse width to be sufficiently short. Pulses of 80 ps at 0.532 μm were generated with the laser system shown schematically in Fig. 1.

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A Quantronix 116R continuous-working mode-locked Nd^{3+}:YAG laser was mounted on an Invar 36 rail to increase thermal stability, and the variable frequency mode locker driver was replaced by a high stability single-frequency generator. The laser mirrors were mounted on differential micrometer translation stages to provide precise cavity length tuning.

The laser output characterized by $\lambda_L = 1.064$ μm, pulse width (τ_L) = 100 ps, pulse energy = 100 nJ, and repetition rate = 100 MHz was fed into a double Pockels cell single-pulse selector, which removed one pulse from the mode-locked pulse train at a repetition rate of 10 Hz. The residual pulses were frequency doubled using a KTP crystal and monitored using a synchroscan Photochron II streak camera.[6] The selected pulses were then passed into a regenerative ring amplifier with a gain of 10^4, yielding a 1 mJ pulse of 100 ps duration. These pulses were further amplified in a single-pass (gain = 10) amplifier and were frequency doubled, with pulse energies of 5 mJ.

Upon focusing with an f = 1 m lens, second-harmonic (0.532 μm) pulses with peak intensities of up to 10 $GW \cdot cm^{-2}$ (horizontally polarized) were easily obtained. Degenerate four-wave mixing was demonstrated experimentally using the setup shown in Fig. 2. The input intensity was varied using a half-wave plate (WP1) and a Glan polarizer (GP).

Counterpropagating pump beams ($\lambda = 0.53$ μm) were obtained using a retroreflection geometry, where the distance between the sample S and the retroreflecting mirror M was minimized, if maximum temporal pulse overlap was desired. The probe beam was split off from the pump beam by beam splitters BS1 and BS2, and the angle between the pump (I_1) and probe (I_3) beams was chosen to be 6° and the ratio between them 20:1. The synchronized arrival of the probe pulse with the two pump pulses was controlled by the prism combination DL. The polarizations of beams I_2 and I_3 could be rotated with respect to I_1 by inserting quarter-wave plate WP2 and half-wave plate WP3. The sample cell used in this experiment was made of quartz and was 1 mm in thickness.

The intensities of all three beams were increased at the sample by means of the 1-m-focal-length lens L1. Input

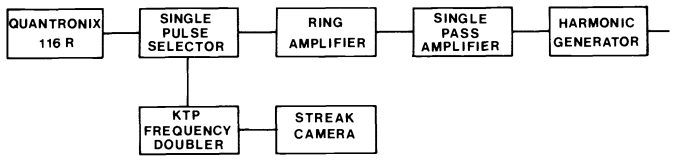


Fig. 1. Schematic diagram of the laser system.

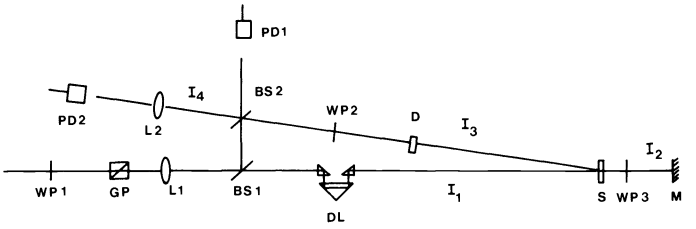


Fig. 2. Experimental setup for degenerate four-wave mixing using retroreflection geometry. All abbreviations are explained in text.

pulse energies were measured using a Scientec 362 power-energy meter PD1, and output energies I_4 were measured using a vacuum photodiode PD2. A second lens, L2, was placed in the beam I_4 so that the far-field pattern could be observed with and without a distorter D in the probe beam I_3.

3. NONLINEAR MEDIUM: POLYDIACETYLENE

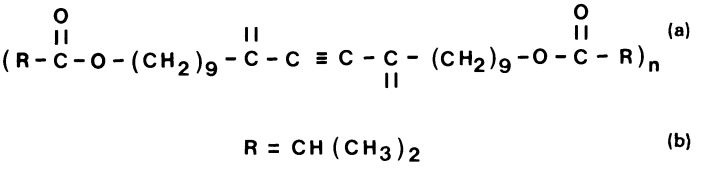
Recently, a range of polydiacetylenes have been produced that are highly soluble in a variety of organic solvents.[7] The polydiacetylenes have the general formula shown in Fig. 3(a). Of these, we chose the polymer with the R group as shown in Fig. 3(b). This polymer, which may be obtained as a free-standing red film, forms a yellow solution in toluene. Figure 3(c) shows the absorption spectrum of the used polydiacetylene in toluene. The laser wavelength that lies in the tail of the absorption band is also indicated.

4. RESULTS AND DISCUSSION

A four-wave mixing signal counterpropagating to the probe beam was easily observed. In accordance with four-wave mixing theory this signal vanished on being deprived of beams I_1, I_2, or I_3. The optimal concentration of polydiacetylene in toluene was found to be 2.3×10^{-3} M, corresponding to a sample transmission of 50% at the laser wavelength. The transmission remained constant over a range of intensities up to 5 GW•cm^{-2}, showing no tendency to saturable absorption. With this solution a phase conjugate reflectivity ($R_{PC} = I_4/I_3$) of 100% was found at a first pump pulse peak intensity of I = 1.5 GW•cm^{-2}. The phase conjugate reflectivity (R_{PC}) was measured as a function of incident pulse intensity (I_1) (Fig. 4) by rotating the half-wave plate WP1. Absolute peak intensities were obtained by measuring the intensity-dependent transmission of di-iodomethane.[8]

The experimental data were fitted to a quadratic dependence, yielding an effective nonlinear susceptibility of $\chi^{(3)}_{xxxx} = (7 \pm 3) \times 10^{-20}\ m^2 \cdot V^{-2}$ [$(5 \pm 2) \times 10^{-12}$ esu] from[9]

$$\chi^{(3)} = \frac{4c^2n^2\epsilon_0\alpha\sqrt{R_{PC}}}{3\omega I_1 T(1 - T)}, \quad (1)$$



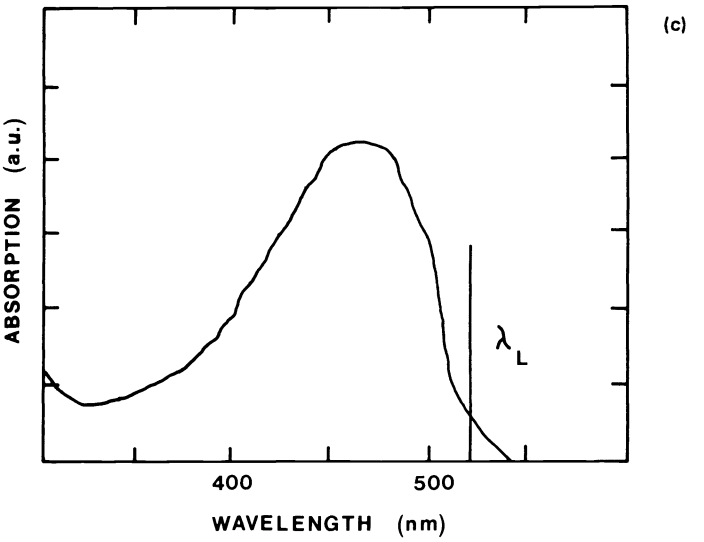


Fig. 3. Chemical formula of the used polydiacetylene. (a) The basic structure of the polymer, (b) the side chain of the sample used, and (c) an absorption spectrum in toluene.

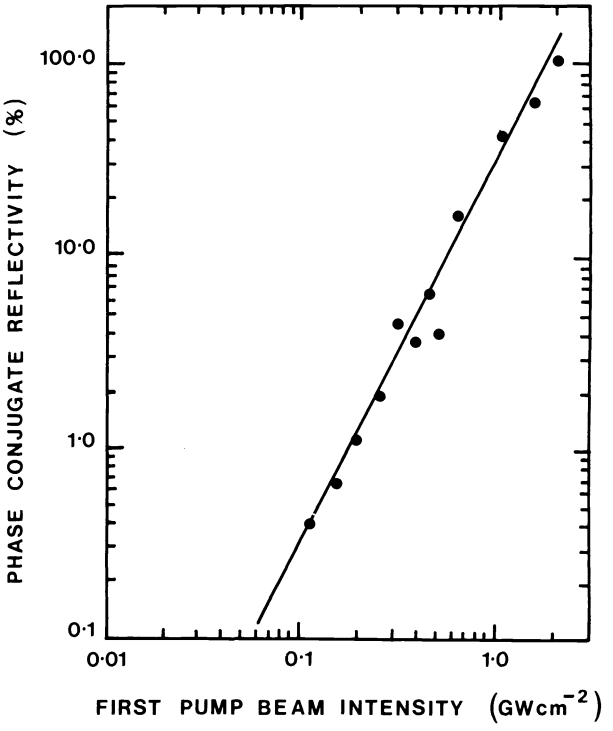


Fig. 4. Dependence of the phase conjugate reflectivity on the first pump beam peak intensity.

where n = refractive index, ω = laser frequency, α = extinction coefficient, T = sample transmission, and other symbols have their usual meanings or are explained in the

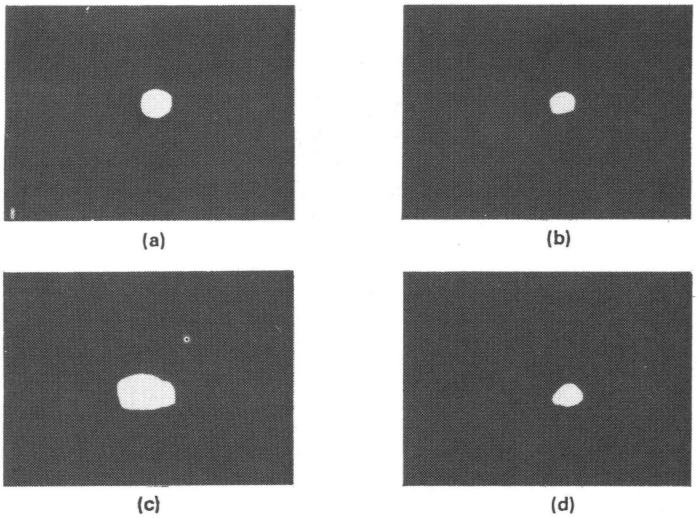


Fig. 5. Experimental demonstration of optical phase conjuction. (a) The undistorted probe beam, (b) the undistorted phase conjugate beam, (c) the distorted probe beam after reflection off an ordinary mirror and repassing the distorter, and (d) the cross section of the phase conjugate of the distorted probe beam after repassing the distorter.

text.

The various components of the nonlinear susceptibility tensor were evaluated by inserting suitable combinations of wave plates WP2 and WP3. Values of $\chi^{(3)}_{xyxy} = (5 \pm 2) \times 10^{-20}\ m^2 \cdot V^{-2}$, $\chi^{(3)}_{xxyy} = (2 \pm 1) \times 10^{-20}\ m^2 \cdot V^{-2}$, and $\chi^{(3)}_{xyyx} = 0$ were found. The fact that $\chi^{(3)}_{xxxx} = \chi^{(3)}_{xxyy} + \chi^{(3)}_{xyyx} + \chi^{(3)}_{xyxy}$ shows that the nonlinear medium is spatially isotropic.

Phase conjugate behavior of the signal beam was demonstrated by observing the distortion-undoing properties of the far-field pattern of the beam observed at the focus of an f = 0.5 m lens. Photographs of the beam (Fig. 5) show clearly the restoration of the incident probe-beam cross section after passing the phase conjugate of the distorted probe beam back through the distorter.

In the formalism of real-time holography the tensor components $\chi^{(3)}_{xyxy}$ and $\chi^{(3)}_{xxyy}$ are generated by laser-induced transient gratings. To clarify the nature of the interaction of the beams with the nonlinear medium, the decay of the laser-induced grating was measured by delaying the incidence of the second pump pulse (I_2). A signal could be observed for delays up to 40 ns, indicating that the process is a thermally induced refractive index change. The effective nonlinear susceptibility of such a process is described by[9]

$$\chi^{(3)} = \frac{dn}{d\theta} \left| \frac{4n^2 c \epsilon_0 \alpha \tau \Phi}{3\varrho\, C_p} \right| , \qquad (2)$$

where $dn/d\theta$ = rate of change of refractive index with temperature, ϱ = density of solvent, and C_p = specific heat capacity of solvent at constant pressure. The symbol $\Phi = 1 - \eta$ (where η = fluorescence quantum yield of solute) represents the fraction of absorbed light energy transferred to the solvent in the form of heat. This process is very efficient in polydiacetylene/toluene because $\eta = 2 \times 10^{-3}$. The resulting refractive index changes form two gratings with grating spacings $\Lambda_{large} = 5.1\ \mu m$ and $\Lambda_{small} = 0.178\ \mu m$, calculated from

$$\Lambda = \frac{\lambda_L}{2 n \sin(\theta/2)} , \qquad (3)$$

where θ is the angle between the two beams forming the grating. These gratings decay by thermal diffusion with time constants $\tau_{large} = 7.2\ \mu s$ and $\tau_{small} = 8.8$ ns, calculated from[10]

$$\tau_{large,\,small} = \frac{\varrho C_P \Lambda^2_{large,\,small}}{4\pi^2 \varkappa} , \qquad (4)$$

with $\varkappa$ = thermal conductivity of the solvent.

From Eq. (2) a theoretical value for the effective nonlinear susceptibility of $\chi^{(3)} = 9 \times 10^{-20}\ m^2 \cdot V^{-2}$ can be obtained, which compares favorably with values obtained experimentally.

5. CONCLUSIONS

In conclusion, we have demonstrated efficient phase conjugation by degenerate four-wave mixing of picosecond light pulses in a soluble polymer. The origin of the process is a thermally induced refractive index change, making it possible to tailor the nonlinear optical susceptibility and the possible repetition rate of phase conjugation by a suitable choice of the solvent.

6. ACKNOWLEDGMENTS

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