PICOSECOND PHASE-CONJUGATE REFLECTION FROM ORGANIC DYE SATURABLE ABSORBERS

J.O. TOCHO *, W. SIBBETT and D.J. BRADLEY
Optics Section, Blackett Laboratory, Imperial College, London SW7 2BZ, UK

Received 27 March 1980

Phase conjugation with conversion efficiences of up to 50% has been obtained in DODCI and other saturable absorbers with picosecond pulses from a mode-locked dye laser. Reflectivity shows a quadratic dependence on pump intensity up to ≈ 500 MW cm^{-2}.

1. Introduction

Phase conjugation reflection [1] by degenerate nonlinear mixing [2] involving local saturation of absorption [3,5] or gain [6,7] has been experimentally demonstrated in gaseous, solid and liquid media. These experiments have employed a *Q*-switched ruby laser pumping a saturable dye solution [2], a CW argon ion laser interacting with a ruby crystal [3], pulsed CO_2 lasers using germanium [4] and SF_6 [5] as saturable absorbers, and a pulsed dye laser pumping atomic sodium vapour [6]. Nonlinear mixing within the laser gain medium has been obtained with pulsed CO_2 [7] and Nd-YAG lasers [8]. Wavefront conjugation has been produced with picosecond pulses using stimulated Raman and Rayleigh Wing Scattering in benzene and cyclohexane [9], and the nonlinear Kerr effect in CS_2 [10]. We report achievement of high efficiency (up to 50%) phase conjugation reflection in organic dye saturable absorbers, produced by a train of picosecond pulses from a passively mode-locked flashlamp pumped dye laser. As with ruby [3,10] the conjugate pulses can be efficiently generated with broad-band and thus ultrashort duration, pumping pulses but since the lifetime of the excited electronic level of a dye is ~1 ns (compared with 4 ms for ruby) the transistory hologram creates is more suitable for rapid real-time holography and signal processing [1].

* Permanent address: Centro de Investigaciones Opticas, Casilla de Correo 124, 1900 La Plata, Argentina.

2. Experimental

A flashlamp passively mode-locked dye laser [11] generated a 1.2 μs train of pulses of duration ≈5 ps at the centre of the train and with ≈50 μJ of energy in each pulse. The laser was tuned to operate at 605 nm where mode-locking is optimum [12]. The pulses are separated by the 5 ns round-trip time of the laser cavity and the output was more than 95% polarized in the horizontal plane. A beam-splitter (BS1 in fig. 1) devided the laser beam to generate pulses of high intensity I_1 for pumping and low intensity I_2 probe pulses. The times of arrival of the pulses of each pair at the saturable absorber dye cell were controlled by varying the lengths of the prism optical delay lines. The interaction cell was of the type used in passively mode-locked dye-lasers [11] with a 100% reflectivity mirror immersed in the dye. The mirror was aligned to retroreflect the incident pumping beam so as to provide counter propagating, pump pulses for phase conjugation. The probe beam pulses were arranged to arrive at the cell at the same time as the pumping pulses. The resulting phase-conjugate reflected pulses were detected by a fast photodiode (ITL type FD125), after passing through the 50% reflectivity beamsplitter BS2. Alternatively, the reflected beam profile could be photographed with a lensless camera back, C, using another beamsplitter BS3. Neutral density filters, ND, were used to attenuate the pump and probe beams. When a half-wave plate was placed in

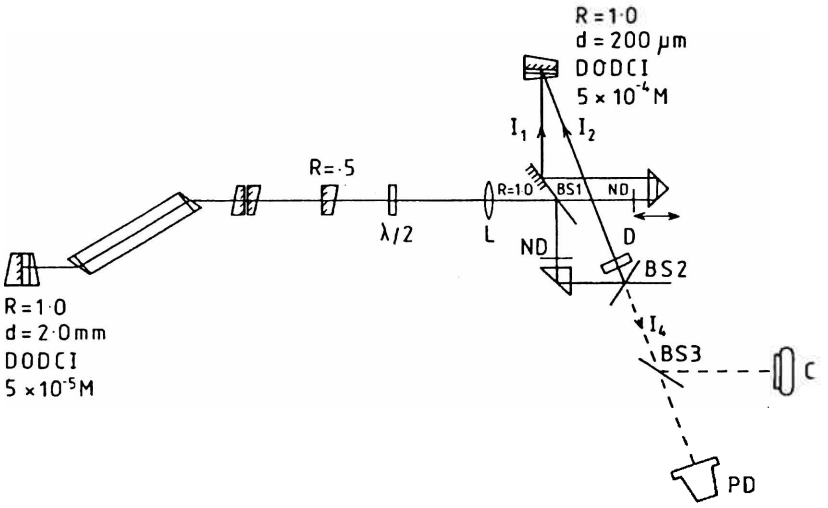

Fig. 1. Experimental arrangement.

the laser beam the plane of polarization was rotated. In this way two values for the ratio of the probe and pump beam intensities, I_2/I_1 were obtained. For horizontal polarization $I_2/I_1 \approx 0.009$ and for vertical polarization this ratio had the value 0.085.

The pulses were focused into the 200 μm thick dye layer by a 35 cm focal length lens, L, to give a peak pump intensity of ≈500 MW cm^{-2}, in an area of ≈1 mm diameter. To confirm that phase conjugation was occurring a cylindrical lens of 15 cm focal length was placed in the probe beam path at D. Solutions of DODCI, DQTCI and Oxazine 1, all absorbing at the dye laser wavelength, in a range of solvents (ethanol, methanol, and glycerol) were employed in the dye cell. The results were found to be independent of the solvent employed and phase conjugate reflectivity was produced by all three saturable absorbers. Most of the work was carried out with DODCI dissolved in ethanol and the results obtained with this dye are reported.

3. Results

To obtain backward wave reflectivity it was necessary to ensure that the difference in time of arrival of the object and pump pulses at the absorber cell was kept less than the coherence time of the laser beam. Fig. 2 shows the variation of reflected intensity as a function of the delay between the object and pump pulses. This is effectively a method of measuring the temporal coherence function of the laser beam [13]

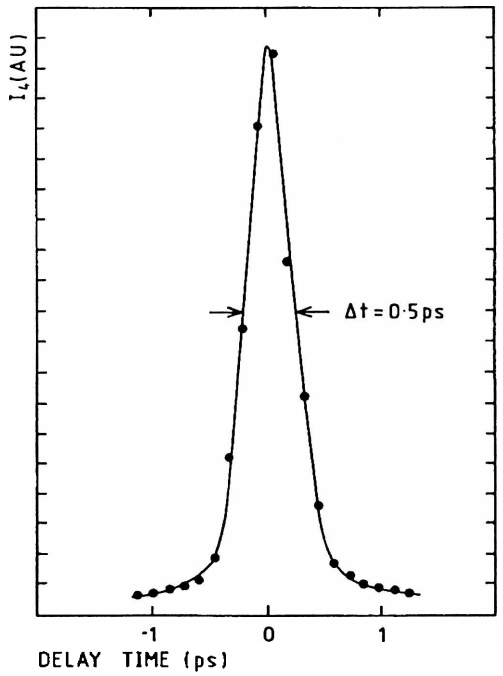

Fig. 2. Variation of reflected intensity I_4 (arbitrary units) with delay between arrival of pump and probe pulses at DODCI cell.

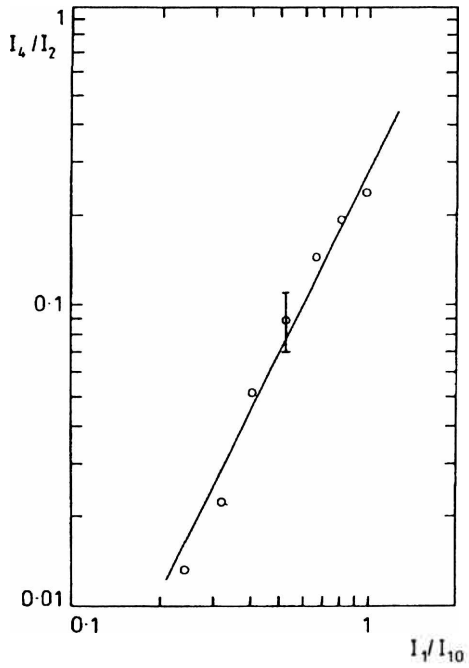

Fig. 3. Plot of phase conjugate reflectivity R as a function of pump intensity I_1 (normalized to maximum pump intensity I_{10} = 480 MW cm^{-2}). I_2 = 5 MW cm^{-2}.

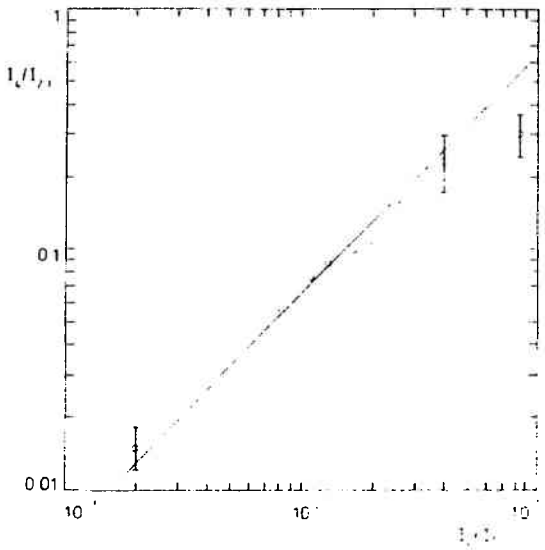

Fig. 4. Plot of phase conjugate reflected intensity I_4 (normalized to maximum probe intensity I_{20} = 36 MW cm^{-2}) as function of probe intensity I_2 (normalized to pump intensity I_1 = 440 MW cm^{-2}).

and the half-width of 0.5 ps gives the value of the coherence time. The spectral bandwidth of the pulse train was measured to be 2.3 nm which corresponds to a coherence time (or pulse duration for bandwidth-limited pulses) of 0.22 ps for gaussian shaped pulses. The discrepancy probably arises from spectral broadening due to self-phase modulation when the laser is pumped above threshold [14]. This broadening increases monotonically along the pulse train so that the recorded time-integrated bandwidth largely comes from pulses towards the end of the pulse train. Thus fig. 2 gives an average coherence time for the whole pulse train.

The reflectivity as a function of pump intensity is plotted in fig. 3 for horizontally polarized light. The maximum pump intensity employed was well above the saturation intensity $I_{sat} \approx 250$ MW cm^{-2} of DODCI for 5 ps pulses at 605 nm. The theory of conjugate-wave generation by nonlinear mixing has been treated for two-level [15] and three-level [3] absorbing media. Since the relaxation rates within the electronic singlet bands of the saturable absorber dye molecules are rapid ($\approx 10^{12}$ s^{-1}) it is possible to apply the two-level theory of [15] if triplet state population and absorption to higher singlet and triplet levels are neglected. Then the reflectivity R can be shown [3,15] to be proportional to $I_1^2 \exp(-2\alpha d)(1-\exp(-2\alpha d))^2$ where d is the dye cell thickness and α is the absorption coefficient. The expected quadratic dependence of reflectivity upon intensity is shown in fig. 3 up to the maximum pump intensity employed of ≈500 MW cm^{-2}. The corresponding value for the backward wave reflectivity was then ≈30%. In fig. 4 the reflected intensity for vertically polarized light is plotted as a function of the probe intensity for a constant pump intensity of ≈450 MW cm^{-2}. The output intensity is seen to increase linearly with the object wave intensity until this approaches 17 MW cm^{-2}, corresponding to an output power of ~8 MW cm^{-2}. Before saturation sets in the power reflectivity is ≈50%. The dependence of reflectivity upon the DODCI concentration is shown in fig. 5, measured for a pump intensity and a probe intensity similar to that used for fig. 4. The experimental points are seen to be a good fit to the theoretical function $\exp(-2\alpha d)(1-\exp(-2\alpha d))^2$ for the experimental parameters employed. At dye concentrations below the optimum value of 5.5 ×

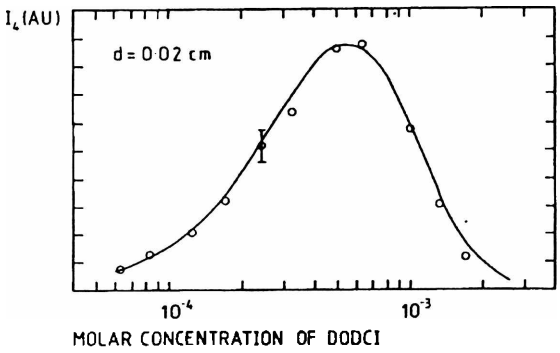

Fig. 5. Plot of reflected intensity I (in arbitrary units) as function of DODCI molar concentration for pump and probe intensities similar to fig. 4. The smooth curve corresponds to the theoretical relationship $I_4 \propto \exp(-2\alpha d)(1-\exp(2\alpha d))^2$ $\alpha = n\epsilon$; n = molar concentration, ϵ = extinction coefficnet (0.5×10^5 M^{-1} cm^{-1} at 605 nm) and $d = 0.02$ cm (the cell thickness).

10^{-4} M the coupling constant for nonlinear mixing is low [15] and the intensity of the backward wave is reduced. For higher dye concentrations the efficiency also drops because of increasing absorption of all the waves. For the optimum dye concentration but with a cell of 1 mm thickness there was a considerable decrease in efficiency (to ≈5% reflectivity).

Confirmation of wavefront phase conjugation in the backward reflected pulses is given by the photographs of the beam profiles of fig. 6. While the cylindrical lens distorter has only a minor effect on the beam profile reflected from the dye cell, replacing this with a plane mirror produces a major change. Phase-conjugate reflection has also been obtained when the dye laser was not mode-locked. The reflected laser pulse had a duration ≈270 ns which was much shorter than the expected value of 870 ns for the 1.5 μs pumping duration. This was probably due to the effect of photoisomer generation [16].

In conclusion our results show that saturable absorber dyes can be employed for efficient phase-conjugation of broad-band laser pulses. With a wide range of dyes absorbing in the ultra-violet, visible and near-IR, backward conjugation of picosecond laser pulses can thus be easily obtained throughout these spectral regions. Optimum conditions can be readily achieved by simply adjusting the dye concentration and the cell thickness.

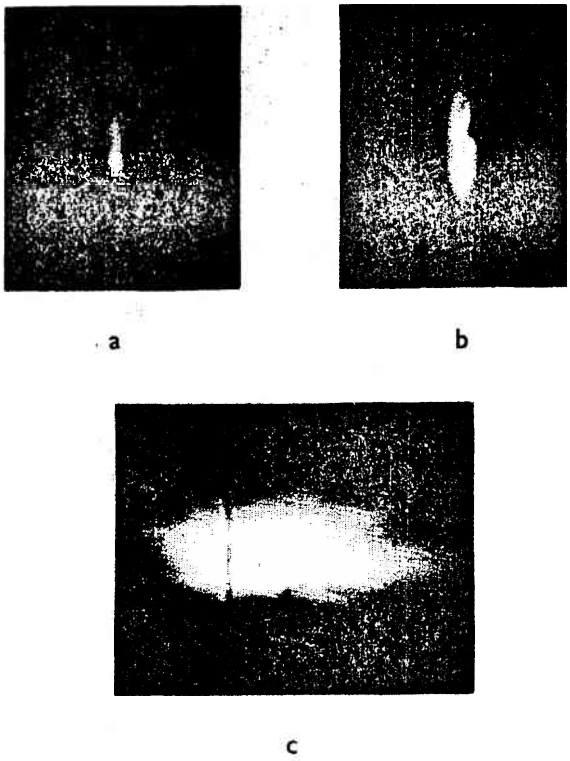

Fig. 6. (a) Reflected phase conjugate beam profile recorded on film in camera-back (C of fig. 1) with ×3 attenuation. (b) Reflected phase conjugate beam with 15 cm focal length cylindrical lens distorter with no attenuation. (c) Reflected beam with distorter when DODCI cell was replaced by plane mirror for ×10 attenuation.

We are pleased to acknowledge financial support from the Science Research Council and one of us (J.O.T.) is supported by the Consejo Nacional Investigaciones Cientificas y Técnicas de la República Argentina.

References

[1] A. Yariv, I.E.E.E.J., Quant. Electron QE-14 (1978) 650; QE-15 (1979) 524, and references therein.
[2] B.I. Stepanov, E.V. Ivakin and A.S. Rubanov, Sov. Phys. -Dokl. 16 (1971) 46.
[3] P.F. Liao and D.M. Bloom, Optics Lett. 3 (1978) 4.
[4] E.E. Bergmann, I.G. Bigio, B.J. Fledman and R.A. Fisher, Optics Lett. 3 (1978) 82.
[5] R.C. Lind, D.G. Steel, J.F. Lam and C.R. Guilionoa, Appl. Phys. Lett. 34 (1979) 457.

[6] D.M. Bloom, P.F. Liao and N.P. Economon, Optics Lett. 2 (1978) 58.
[7] R.A. Fisher and B.J. Feldman, Optics Lett. (May, 1979).
[8] A. Tomita, Appl. Phys. Lett. 34 (1979) 463.
[9] A.D. Kudriavtseva, A.I. Sokolovskaia, J. Gazengel, N. Phu Xuan and G. Rivoire, Optics Comm. 26 (1978) 446.
[10] D.M. Bloom, C.V. Shank, R.L. Fork and O. Teschke, Springer Series in Chemical Physics 4, Picosecond phenomena (1978) p. 96.
[11] D.J. Bardkey, in: Topics in applied physics, 18 Ultrashort light pulses, ed. S.L. Shapiro (Springer Verlag, Heidelberg, 1977) pp. 17–81.
[12] D.J. Bradley, Optical and Quantum Electronics 6 (1974) 25.
[13] H.J. Eichler, U. Klein and D. Langhams, Appl. Phys. 21 (1980) 215.
[14] E.G. Arthurs, D.J. Bradley and A.G. Roddie, Appl. Phys. Lett. 19 (1971) 428.
[15] R.L. Abrams and R.C. Lind, Optics Lett. 2 (1978) 94.
[16] E.G. Arthurs, D.J. Bradley and A.G. Roddie, Appl. Phys. Lett. 20 (1972) 125.