Picosecond Phase-Conjugation Reflection and Gain in Saturable Absorbers by Degenerate Four-Wave Mixing

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More reliable mode-locking and bandwidth-limited pulses of shorter durations are obtained when the high-reflectivity mirror is immersed in the saturable absorber solutions used to passively mode-lock pulsed and CW dye lasers, Neodymium and ruby lasers [1]. This result had been previously explained [2,3] by preferential saturation of the absorber at the antinodes of the standing waves in the neighbourhood of the mirror, particularly when the dye cell length is comparable to the laser pulse length. With the recent demonstration of phase conjugation reflection in many materials by degenerate nonlinear mixing [4] involving local saturation of absorption [5] it seemed timely to investigate picosecond phase-conjugation in DODCI and other saturable absorbers commonly used for mode-locking dye lasers. Our results indicate that, in addition to preferential saturation, phase conjugation effects contribute to the improved performance of the immersed mirror mode-locking dye cell, even when the laser beam is focussed on to the mirror as in the case of CW dye lasers [6]. Photoisomer effects [7] are also shown to be important in the phase-conjugation process, and play a dominating role as the laser is tuned to longer wavelengths.

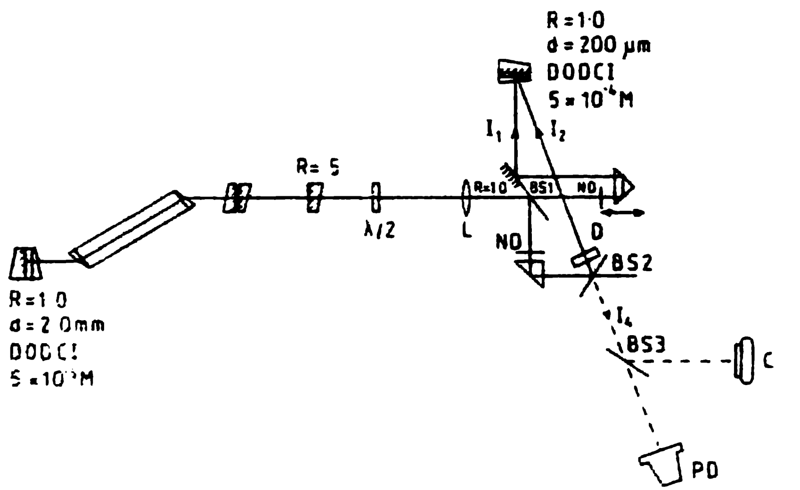


Fig.1 Experimental arrangement for extra-cavity phase conjugation

Experiment

A 1.2μs train of 50 μJ, 5ps pulses from a flashlamp pumped mode-locked dye laser [1] was divided by a beamsplitter to generate pumping pulses of high-intensity I_1 and low-intensity I_2 probe pulses, arranged to arrive simultaneously at the extra-cavity retroflecting cell by appropriate adjustment of the prism optical delay lines. The pulses were focussed into the 200 um path length dye cell to give peak intensity of $\sim$500 MW cm^{-2} in an area of $\sim$1mm diameter. DODCI, DQTCI and Oxazine I dissolved in a range of solvents (ethanol, methanol, and glycerol) were employed. The results were independent of the solvent used and phase conjugation reflectivity was produced by all three dyes. Most of the work was carried out with DODCI.

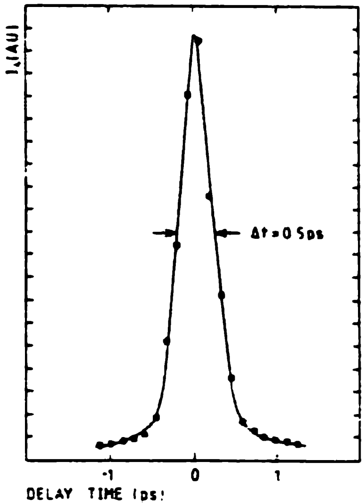


Fig.2(a) Reflected intensity dependence on delay between pump and probe pulses

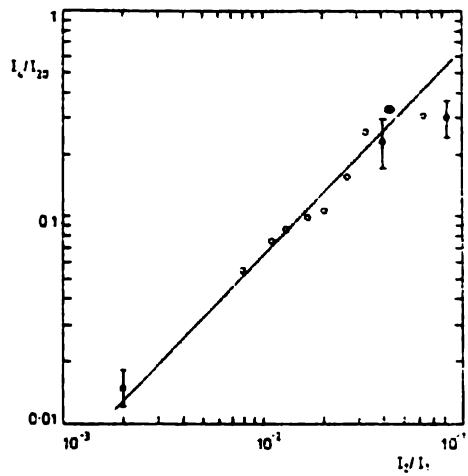


Fig.2(b) Reflected intensity I_4 dependence on probe intensity I_2 (I_{20} = 36 MW cm^{-2}; I_1 = 440 MW cm^{-2})

Figure 2(a) shows the variation of reflected intensity for DODCI as a function of the delay between the object and pump pulses. The half-width of 0.5ps, averaged over a complete pulse train, compares with a coherence time of 0.22ps for the 2.3 nm total bandwidth. Self-phase modulation spectral broadening along the dye lasers pulse train [7] accounts for the difference. As expected there was a quadratic dependence of reflectivity upon pump intensity [8,9]. The output intensity increased linearly with the object wave intensity up to $\sim$17 MW cm^{-2} when saturation set in at a power reflectivity of $\sim$50% (Fig.2(b)). The dependence of reflectivity upon DODCI concentration (Fig.3) is a good fit to the theoretical relation $(\exp - 2\alpha d)\ (1 - \exp(-2\alpha d))^2$ [9] with an optimum concentration of 5.5×10^{-4}M, the typical concentration giving optimum mode-locking performance. Confirmation of wavefront phase conjugation was obtained by correcting for the effect of a cylindrical lens distorter. Phase-conjugate reflection was also obtained when the laser was not mode-locked.

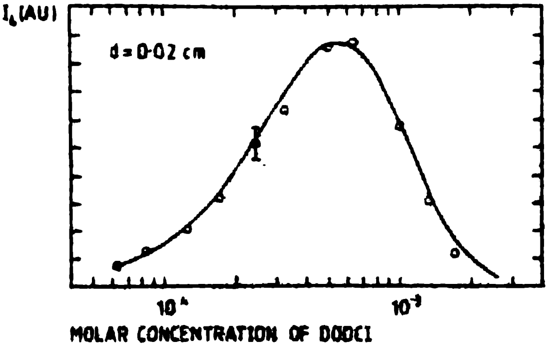


Fig.3 Reflected intensity dependence on DODCI concentration. Smooth curve $I_L \propto (\exp - 2\alpha d)\ (1 - \exp(-2\alpha d))^{2}$, α = absorption coefficient, d = 0.2mm

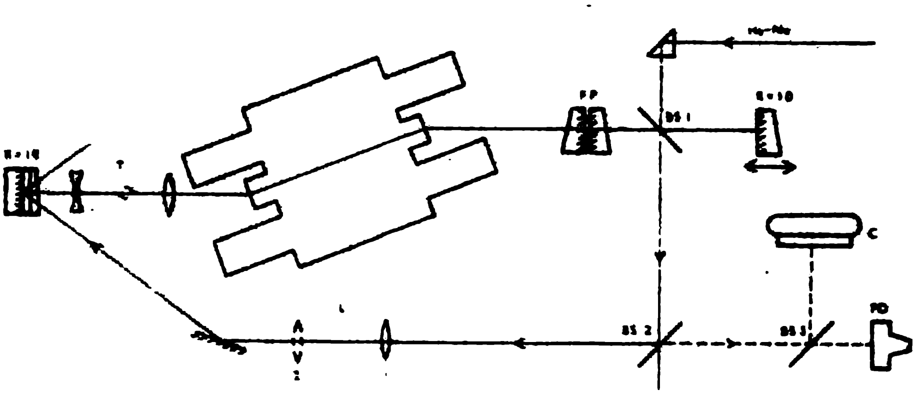
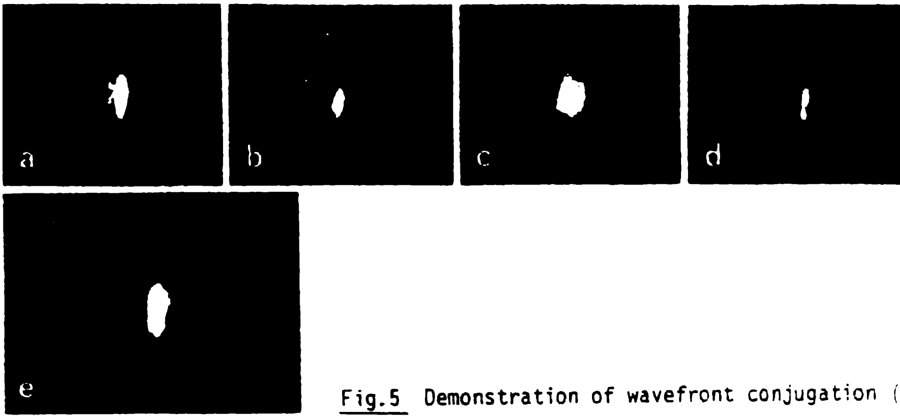
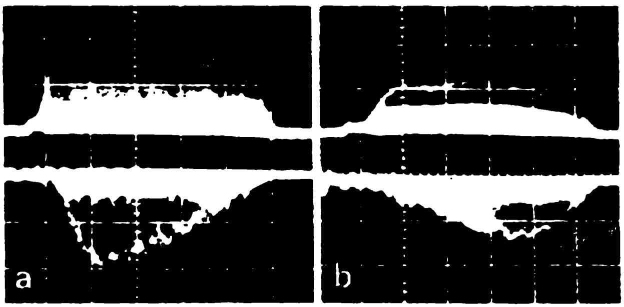


Fig.4 Intra-cavity phase conjugation experimental arrangement.

The experimental arrangement for intracavity phase conjugation is shown in Fig.4. At a DODCI concentration of 2.5×10^{-4}M in a cell of 0.2 mm thickness a reflectivity of 1% was obtained. Fig.5 shows the return beam profile when (a) the cell mirror was at the focus of lens L; (b) the lens was displaced by 5 cm; the fluorescence patterns (c and d) for these two positions respectively and (e) the probe beam reflected by a mirror. Inside the mode-locked laser cavity, phase-conjugation reflection should be produced with greater fidelity [10] because the pump beams and the effective probe beams experience the same distortion in passing through the laser medium. In the arrangement of Fig.4 this was not necessarily the case since the pump beams will have made an extra transit through the laser dye solution. Also, the temporal structure of the pump beam will have been changed [1] by this extra transit through the amplifying medium. As with the extra-cavity arrangement coincidence of the pump and probe pulses to within the coherence time was needed to achieve phase conjugation reflectivity. Thus intra-cavity phase-conjugation will both correct for thermal distortion and tend to produce bandwidth-limited pulses, since bandwidth-limited structures from the initial intensity fluctuations [1] will be preferentially reflected in multiple passes through the saturable absorber.



<u>Fig.5</u> Demonstration of wavefront conjugation (see text



<u>Fig.6(a)</u> <u>Upper</u> Pump mode-locked pulse train (605 nm) <u>Lower</u> Reflected pulse train.

<u>Fig.6(b)</u> As for Fig.6(a) but laser tuned to operate at 617 nm. Note restructuring of mode-locked pulse train envelope due to generation of DODCI photoisomer and subsequent phase-conjugation reflection.

The effect of photoisomer generation in DODCI [7] was clearly seen by a reflected pulse delay of ~500 ns, compared with the pump pulse, for a DODCI (5 x 10[illegible]M) cell pumped by a 617 nm unmode-locked laser pulse. At 586 nm the efficiency reached 150% with zero delay. That the delay at longer wavelengths arises from photoisomer generation is confirmed by Fig.6. Fig.6(a) shows the pump and reflected pulses for a 605 nm mode-locked pulse train, while Fig.6(b) shows a drastic restructuring and delay (time scale 200 ns major division) when the laser was tuned to 617 nm. At this longer wavelength phase conjugation would be more efficient for the DODCI photoisomer created at the beginning of the train, than for the normal form [1]. The delay in the build-up of the photoisomer concentration could be manipulated for the production of wavelength-dependent multiplexing for applications in real-time holography, data and picture processing and wavelength filtering, all with picosecond response times.

Thus with a tunable dye laser it is possible to produce phase conjugation with gain and with picosecond time response. By exploiting photoisomer effec wavelength-dependent variable delays and pulse envelope restructuring is obtained. Since phase conjugation requires coincidence within the coherence time, bandwidth-limited pulses will be preferentially produced by a retro-reflecting absorber dye cell. Amplification and phase conjugation in BDN saturable absorber by 13 ns pulses of a ND:YAG Q-switched laser and simultaneous intra-cavity Q-switching and phase-conjugation reflection has also recently been reported [11].

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