THERMAL EFFECTS IN PHASE-CONJUGATION IN SATURABLE ABSORBERS WITH PICOSECOND PULSES

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Phase conjugation reflection with efficiencies of 400% for microsecond pulses and ~50% for picosecond pulses has been obtained in saturable absorber dye solutions. The effect of different solvents on the generation of the thermal phase grating involved was investigated.

We have recently reported [1] phase-conjugation [2] with conversion efficiences of up to 50% in DODCI and other saturable absorbers obtained with picosecond pulses from a mode-locked dye laser. Phase-conjugation reflection with lower efficiency, has also been measured [3] in an intra-cavity saturable absorber dye cell in the experimental arrangement commonly used for passive mode-locking of pulsed and CW dye lasers [4,5]. Photoisomer effects [6] in the saturable absorber dye were found [3] to be important in the phase-conjugation process, particularly when the laser was tuned to longer wavelengths. Martin and Hellwarth [7] have shown that thermally induced refractive index changes [8–11] were the dominant mechanism in a variety of liquids employed in a four-wave mixing process for image conversion from infrared to visible and more recently the influence of the thermal grating in phase conjugation was discussed by Heilweil et al. [12]. We have investigated the role played by thermal effects in phase conjugation reflection of picosecond pulses from saturable absorbers. In particular, the influence of solvents with very different thermal properties have been investigated together with the time behaviour of the conjugated wave.

The experimental set up (fig. 1) is similar to that reported previously [1]. The essential improvement is the use of a fast electro-optic Pockels' cell shutter between the laser and the sample cell to select out a well defined portion of the dye laser pulse train. A fast photo diode (ITL type FD 125), PD1, was used to trigger the Pockels' cell and similar photodiodes (PD2 and PD3) monitored the probe beam intensity (I_2) and the conjugate wave intensity (I_4). A flash-lamp pumped passively mode-locked dye laser [4] generated ~1.2 μs pulses of ~100 mJ energy. In both cases, the laser was tuned by an intra-cavity etalon. The laser beam was focussed into the dye cell by lens L (f = 40 cm) and divided by a glass beam splitter

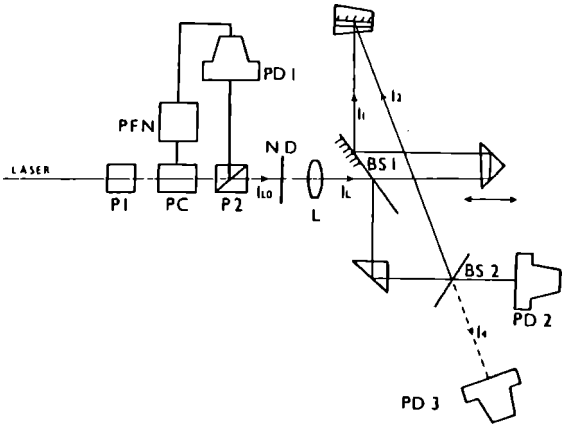

Fig. 1. Experimental arrangement.

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(BS1) to generate the high intensity (I_1) pumping beam and the low intensity (I_2) probe beam. The times of arrival of the two beams at the nonlinear medium were adjusted by varying the position of the prism optical delay line. The dye solution was circulated through a 150 μm width adjustable dye cell of the type used in passively mode-locked dye lasers with a 100% reflectivity mirror immersed in the dye [4]. The retro-reflection of I_1 produced the counterpropagating pump beam (I_3) for phase conjugation.

Solutions of DODCI (3,3'-diethyl-oxadicarbocyanine iodide) and DQOCI (1,3'-diethyl-4,2'-quinolyoxacarbocyanide iodide) in ethanol, a mixture of 50% ethanol and 50% water (by volume) and a similar mixture of ethanol and cyclohexane were investigated. The dye concentration (in the range $10^{-4}-10^{-3}$ M) was adjusted to optimize the phase conjugate reflectivity in terms of the laser wavelength. The laser was tuned between 582 and 620 nm when not mode-locked and between 605 and 617 nm when mode-locked.

Initial experiments were carried out as before [1,3] without using the Pockels' cell switch. A 15 mJ unmode-locked pumping pulse focussed down to an area of 10^{-2} cm^2 produced a reflectivity (I_4/I_2) of ~400% for DODCI in ethanol at wavelengths around the peak of the dye absorption band; this reflectivity is considerably higher than the value of 150% reported earlier [3]. 50% reflection was obtained for the 6 mJ pumping energy in a mode-locked pulse train tuned to 605 nm. The overall temporal behaviour of the conjugate wave was the same in each case being independent of the wavelength, of the dye used and of the concentration (fig. 2a). However, some anomalies were observed. In particular, for dye concentrations much higher than the optimum, the conjugate wave pulse was significantly shorter (fig. 2b) and for DODCI at wavelengths longer than 615 nm there was a noticeably increased delay between the peak of the pump and the peak of the conjugate wave (fig. 2c). At these wavelenghts phase conjugation would be more efficient for the DODCI photoisomer created at the beginning of the train than for the normal form. At 617 nm the photoisomer equilibrium concentration [12] increases the absorption by a factor of ~5.

To obtain phase conjugate reflection it was necessary to ensure that the difference in arrival times between the pump and probe beams at the absorber cell

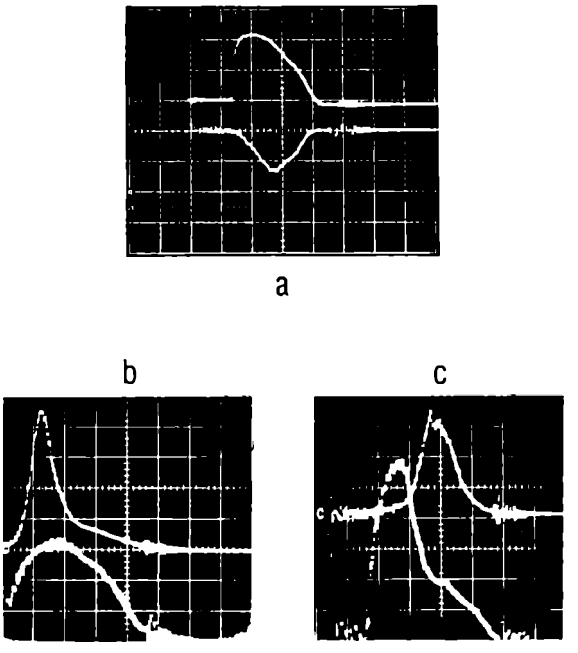

Fig. 2. Temporal profile of probe and conjugate UML pulses (500 ns per major division). a) Typical probe pulse (upper trace) and conjugate pulse (lower trace, inverted). b) Conjugated pulse (upper) where $\alpha \sim 3\alpha$ optimum for DODCI at 592 nm. c) Identical to (b) but optimum concentration and 617 nm.

was kept less than the coherence time of the laser. The temporal coherence function of the laser can be measured effectively by this method [13]. Fig. 3 shows the normalised reflected intensity I_4, as a function of the time delay between the probe and pump beams. The laser was operated at 605 nm and the spectral bandwidths for the conditions of the mode-locked and unmode-locked laser operation were recorded by a spectrograph. The experimental points of I_4 are fitted by gaussians $I_4(t)/I_4(0) = \exp(-4 \ln 2t^2/t_c^2)$ with FWHM of 0.5 ps (ML) and 1.6 ps (UML). In both cases the experimental spectral bandwidths were about twice the theoretical limit ($\Delta\nu\,\Delta t = 0.43$) for a gaussian spectrum. A similar result has been reported for a Nd:YAG mode-locked [14] laser. The discrepancy can be explained for the mode-locked pulse train in terms of spectral broadening due to self-phase modulation when the laser is pumped well above the threshold [6].

In the second part of the experiment, a 100 ns vol-

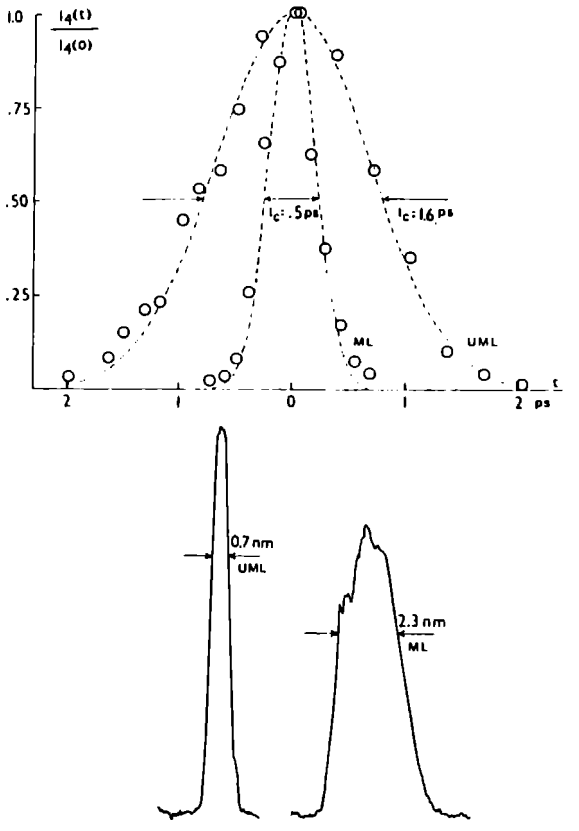

Fig. 3. Normalised reflected intensity as a function of the time delay between the probe and pump beams for ML and UML operation fitted by gaussians (upper) and the corresponding bandwidths for ML and UML (lower).

tage pulse from a Blumlein pulse-forming network (PFN) was applied to the Pockels' cell when the laser was near maximum amplitude. For DODCI in ethanol and the laser tuned to 605 nm, the reflectivity reached 40%, at the end of the 100 ns unmode-locked pulse with a pump energy of 1.9 mJ. For 0.7 mJ pump energy in the picosecond pulses the reflectivity was 6% for the end pulses. Fig. 4 shows the oscillograms for the probe beam and the corresponding phase conjugated beam. With DQOCI, for which no significant photoisomer effect at this wavelength has been reported, a similar build-up in the phase conjugate intensity was obtained. The reflectivity was also measured in different solvents showing a direct relation between the thermal properties of the solvent and the reflectivity at the end of the pumping pulses. In table 1 we summarise the results for the 100 ns (1.9 mJ) pulses excitation, together with the relevant thermal properties of the solvent used.

Several mechanisms seem to be involved in the nonlinear process that produces the diffracted beam. It is clear from fig. 3 that the first picosecond pulses are diffracted with relatively high efficiency (0.6%). At this stage, the mechanism of diffraction can arise either from resonance with the transition $S_0 \rightarrow S_1$ in the dye molecule, or to the thermal grating due to the optical leakage through the Pockels' cell switch (attenuation ratio ~150 : 1) or a combination of both effects. In subsequent pulses it is more likely that the thermal grating is the dominant diffraction mechanism in the saturable absorbers.

In a dye with quantum yield of fluorescence ϕ, a fraction ($f \sim 1 - \phi$) of the energy absorbed is thermalized very quickly by the solvent molecules and the resulting temperature rise produces a phase grating [10,11]. In the geometry of the present experiment, the relevant diffraction grating is created by the interaction of I_1 and I_2 with a thermal decay time of 5 μs (calculated for an angle of 8° between I_1 and I_2 in ethanol). Therefore, thermal diffusion does not erase the grating during the duration of the pulse I_3 which is diffracted to generate the phase conjugated wave I_4. The grating associated with I_2 and I_3 has a periodicity of 0.2 μm and is erased quickly, so it does not reflect appreciably. The diffracted intensity I_4 can be calculated on the basis of this model using the Kogelnik's equation for thick lossy gratings [15]. If we assume that the modulation is uniform inside the cell, in the limit $I_4 \ll I_3$ the reflectivity is given for the following expression:

$$R(t) = I_4(t)/I_2$$

$$= f^2 D^2 (\alpha D)^2 \exp(-3\alpha d) t^2 I_1^2 \quad (t < \tau)$$

$$R(t) = 0 \quad (t > \tau)$$

where D contains the thermal properties of the solvent summarised in table 1, α is the absorption in cm^{-1}, d is the cell length in cm, I_1 is the constant intensity of the pump beam in W/cm^2 and τ is the duration of the probe-pump pulses.

For a 1.9 mJ, 100 ns pump pulse focussed to 10^{-2} cm^2 and using $\phi = 0.42$ for DODCI in ethanol the reflectivity should reach 32% at the end of the pulse. This result is in good agreement with the measured

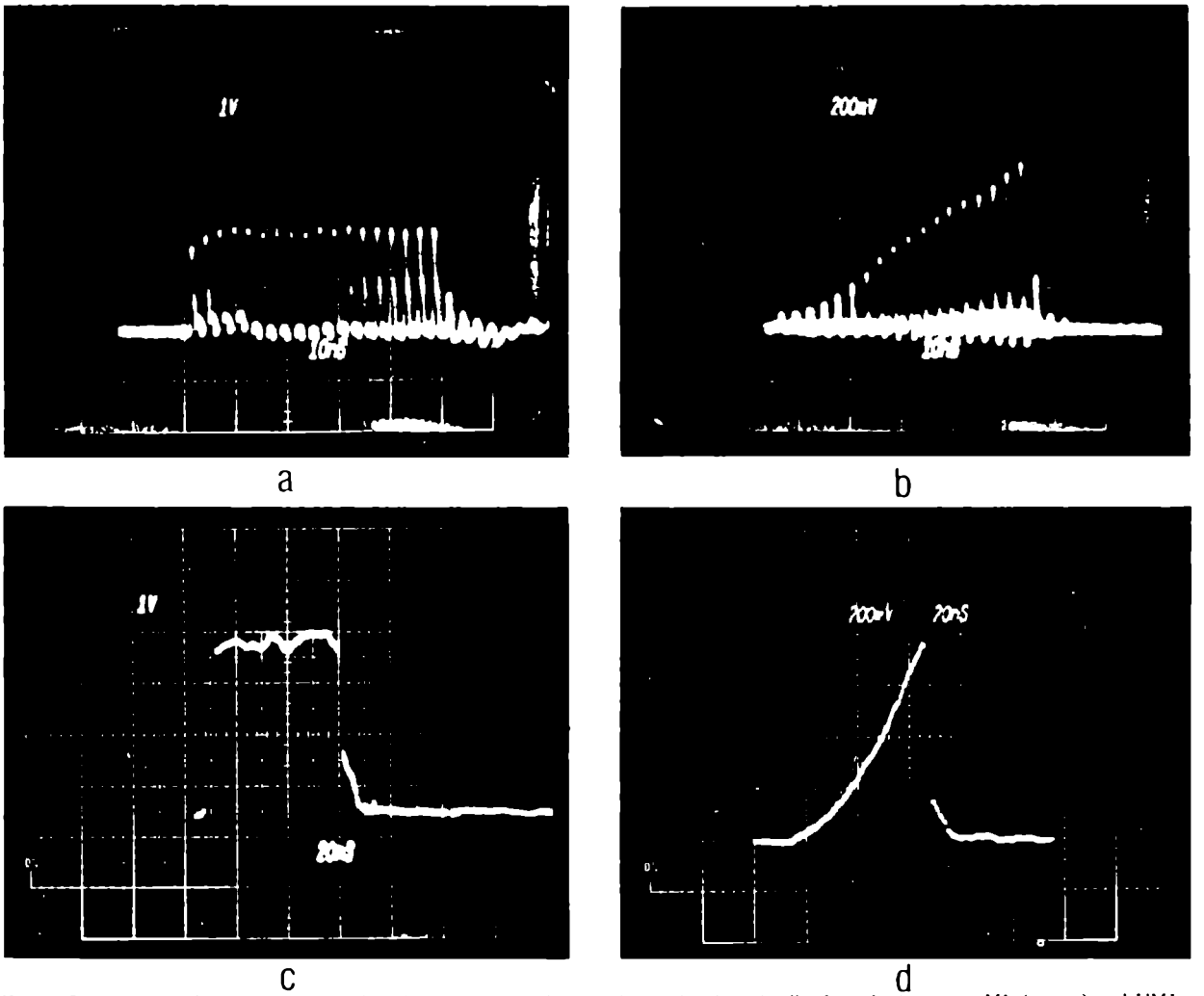

Fig. 4. Oscillogram of probe pulses (a, c) and the corresponding conjugated pulses (b, d) when the laser was ML (upper) and UML (lower). In (b) the reflectivity each 6% at the end of the train. In (a) the maximum reflection is 40%.

Table 1

Solvent	$-dn/dT$ $\{^0C^{-1}\}$	D (605 nm) [a] $\{Jg^{-1}\,^0C^{-1}\}$	R (exp)	R (theor)
H_2O	0.8×10^{-4}	1.95	–	–
ETOH	4×10^{-4}	21.1	0.40	0.32
C_6H_{12}	5.4×10^{-4} [b]	39.1	–	–
H_2O–ETOH	2.65×10^{-4}	8.5	0.08	0.05
ETOH–C_6H_{12}	–	–	0.60	–

a) $D = (dn/dT)/c\rho C_p$, where dn/dT is the temperature coefficient of the refractive index, ρ is the density, and C_p is the specific heat at constant pressure.

b) American Petroleum Institute. Selected values of properties of hydrocarbons and related compounds. Table 23a-E (part 1).

value of 40%. For the ethanol–water mixture, the calculated reflectivity is 5%.

In summary, we have observed efficient generation of phase conjugated waves using a saturable absorber dye solution and a broad band dye laser in the microsecond and picosecond temporal regimes. The spatially periodic change in the absorption relation to the $S_0 \rightarrow S_1$ transition in the dye molecule and the thermalization by collisions with the solvent seem to be the principal mechanisms involved. The same technique could be used to produce high efficiency frequency up-conversion [7] by appropriate selection of the dye.

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