Optical phase conjugation reflectivity and fidelity in CS_2 by picosecond pulse four wave mixing

B. Van Wonterghem, T. E. Dutton, S. M. Saltiel, and P. M. Rentzepis
Department of Chemistry, University of California, Irvine, California 92717

(Received 2 May 1988; accepted for publication 12 July 1988)

We have performed experimental studies designed to elucidate the parameters which limit the efficiency and fidelity of optical phase conjugate reflection of picosecond laser pulses in transparent Kerr media. The experimental data show that at low pulse energies, the reflectivity follows the expected square dependence on the pulse energies. However, it falls below the square at high energies even though, at these high energies, a phase conjugation efficiency of 300% was observed. We observed a rather strong dependence of the reflectivity on the ratio of pump to probe pulse energy. The quality of phase conjugation decreased at very high pump energies, but could under some conditions be improved by focusing the probe beam into the interaction region. Of the several mechanisms responsible for the decrease in efficiency and fidelity, we find that the predominant ones are pump depletion, small scale self-focusing, and dephasing owing to a difference in energy of the two pump pulses.

I. INTRODUCTION

Since Hellwarth's pioneering work on optical phase conjugation (OPC) by degenerate four wave mixing (DFWM),[1] intensive research has been conducted in this new field of coherent optics. One of the main reasons for this activity being the numerous exciting applications in image processing and adaptive optics which this field promises to make possible (see for example Ref. 2). During this decade, many new mechanisms causing a nonlinear optical behavior have been discovered and some of them, e.g., the photorefractive effect,[3] have been applied successfully in image amplifier devices[4] and phase conjugate resonators.[5] The nonlinear optical Kerr effect (OKE),[6] however, has been known already for a very long time and has been studied extensively in connection to self-focusing[7] and optical Kerr shutters.[8] The optical Kerr effect is characterized by an intensity dependent refractive index. The microscopic mechanism for the optical Kerr effect in simple liquids is the orientation of induced dipoles in molecules with an anisotropic polarizability by the electric field of a strong electromagnetic wave. OKE may also be caused by the hyperpolarizability of the molecules (electronic contribution), which of course, effects liquids to a lesser degree.[9]

The most important characteristic of the optical Kerr effect is the very short response time, which amounts to picoseconds for the rotational mechanism and is, in principle, instantaneous for the electronic contribution ($t_r < 10^{-12}$ s). Among organic liquids, CS_2 shows the highest nonlinear optical coefficient [$n_2 = 2.0 \times 10^{-10}$ cm^2/kW (MKS)] and is characterized by a response time shorter than 2 ps. In contrast to, for example photorefractive materials, CS_2 has been used only very few times as PC-producing medium in image processing applications. Different configurations have been reported to perform optical phase conjugation by FWM in CS_2 with reflectivities ranging from 0.001 to 2.[10–16] Amplified reflection in CS_2 ($R \approx 2$) has been achieved by Pepper *et al.*, in a collinear geometry of pump and probe beams with perpendicular polarization in a 40-cm cell, using a Q-switched Ruby laser. Blashchuk *et al.* used a multiple pass configuration for the probe beam to increase the interaction length. They report a reflectivity $R = 0.2$ using a pump intensity of 10 MW/cm^2. Only recently the use of picosecond pulses for DFWM for in CS_2 has been reported[17] where it was found that at a pump fluence of 15 mJ/cm^2, the reflectivity equaled 0.6. The same authors developed a model, which relates the phase conjugate reflectivity to the fluence of the pump pulses in the case that the overlapping region of the pulses is well within the nonlinear medium. Depletion of the pump beams was however not taken into account. Other media have been used to obtain high reflectivities from picosecond pulses.[18,19] In these cases, the grating was found to be mainly thermal in character. Previous theoretical work on transient response FWM in Kerr media was performed mainly on four wave mixing using cw pump beams and a pulse probe beam.[20,21]

The most predominant processes cited which limit the efficiency and the fidelity of the phase conjugate reflection of nanosecond pulses in Kerr media are stimulated Brillouin scattering (SBS) and self-focusing of the high power pump beams. The magnitude of these perturbations are difficult to evaluate numerically because neither of these factors are yet integrated into the present theoretical description of four-wave mixing. We expect, however, that the use of picosecond pulses would decrease the influence of both these nonlinear processes. A reason for this is that the threshold for SBS increases sharply when the pulse width becomes smaller than the relaxation time of the hydrodynamic modes and the length of the pulse becomes comparable to the characteristic gain length.[22,23] Depletion as a limiting factor has been described theoretically in the case of collinear pump and probe beams.[24] A more important effect, which is unavoidable in a four-wave mixing geometry using retroreflection is a difference in energy of the two pump pulses, caused by scattering losses on the sample cell walls and imperfect reflection by the retroreflecting mirror. This results in a slight phase mismatch, caused by the intensity dependence of the wave vectors in the Kerr medium.[24,25] Effects due to the Gaussian beam profile of the applied laser pulses, which cause a spatial dependence of the phase conjugated reflectivity, have been treated theoretically by Trebino[27] and Bochove.[28]

In this paper, we report on high efficiency phase conjugate reflection using picosecond pulses in CS_2. The influence of pump pulses energy ratio, pump to probe pulse energy ratio, and cell length were determined. The effect of focusing on the efficiency and fidelity was studied and is discussed in this paper.

II. EXPERIMENTAL APPARATUS

Phase conjugation in CS_2 was studied experimentally utilizing the four wave mixing experimental system shown in Fig. 1. A passively mode locked Nd:YAG oscillator produced a train of 8–10 pulses, with a pulse duration of 25 ps in a TEM_{00} transverse mode. A single pulse was selected out of the pulse train and amplified through two single pass amplification stages. Between the two amplifiers, the beam was expanded by a factor of 3 by means of an inverted telescope, to increase the amplification and to reduce the divergence of the beam. The amplified pulse was frequency doubled to 532 nm, resulting in a single picosecond pulse with a maximum energy of 15 mJ, which corresponds to a peak power of approximately 600 MW. Vertical polarization of the frequency doubled pulses was insured by a polarizing beamsplitter. Beamsplitter BS_1 ($R = 0.15$) was used to create the probe beam while beamsplitter BS_2 ($R = 0.50$) reflects a small portion of the probe pulse into a reference pulse I_r. The reference pulse is reflected back to BS_2 by mirror M_8, whose reflectivity was chosen to meet experimental requirements. The probe beam traverses the sample at an angle of 6° with the pump beam I_f. The temporal overlap was adjusted by the variable delay line consisting of mirrors M_3–M_5. The backward pump beam I_b was created by retroreflection of I_f by mirror M_1 in close contact with the sample cell to insure a maximum temporal overlap of the pulses within the nonlinear medium. The 0.55-mm thickness of the cell wall added to the retroreflection setup an inherent delay of 3 ps between the two pump pulses. This delay is much smaller than the pulse width and the experimentally determined coherence length of the pulses, which was 14 ps. In some experiments a special cell was used, the rear end of which was a mirror. This cell was however more difficult to handle and did not improve the maximum attainable phase-conjugate reflectivity. CS_2 was HPLC grade (glass distilled and filtered) and was contained in quartz spectroscopic cells with 1-, 2-, or 10-mm optical path lengths. The diameter of the pump beams in the sample cell was 3 mm. Some of the experiments were performed focusing the probe-beam into the sample cell. The phase conjugated beam I_c retraced the path of the probe and was detected by a fast photodiode (ITT F4000). The intensity was attenuated by three glass slides (G) which could be replaced by mirrors when necessary. The reference pulse is detected by the same photodiode and precedes the conjugate pulse by 7 ns. Both pulses were measured using a Tektronix 500 MHz storage oscilloscope. The detection system was calibrated by placing mirror M_R in the path of the probe beam. Pulse energies were measured by a pyroelectric energymeter (Laser Precision RJ7200).

III. EXPERIMENTAL RESULTS AND DISCUSSION

Using the experimental system described in Sec. II, phase conjugate reflectivity was measured as a function of pulse energy for different values of probe to pump pulse fluence F_{pr}/F_f, for different ratios of the two pump pulse energies E_f/E_b and for different cell lengths L. We also checked the presence of stimulated Brillouin scattering and whole beam self-focusing of the pump pulses. Both of these nonlinear effects were found to be negligible. In an attempt to detect two-photon absorption in CS_2, the transmission of picosecond pulses was measured as a function of their energy. However no significant energy dependence of the transmission could be detected up to an energy of 5 mJ/pulse.

In the retroreflection setup, the maximum interaction length of the two pump beams is limited to $\frac{1}{2}$ of the pulse length, so it is advantageous to minimize wasted overlap.

In the first experiment we used a specially designed cell, with length 2 mm, in which the rear surface consisted of a

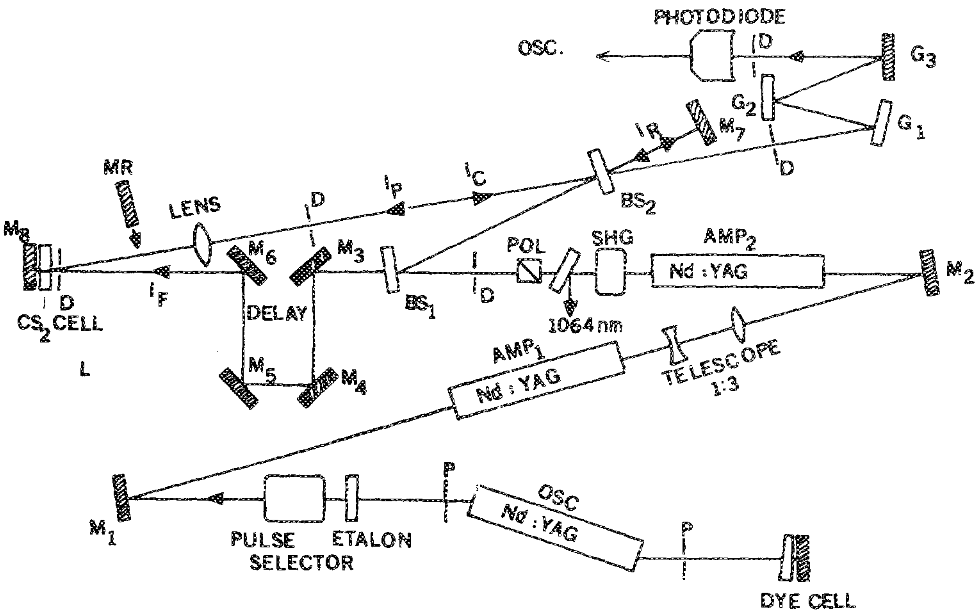

FIG. 1. Experimental setup for the production of phase conjugation by degenerate four wave mixing of picosecond pulses. The backward pump pulse is produced by retroreflection of the forward pump beam. M: mirror; D: diaphragm; SHG: second harmonic generator; POL: polarizer; BS: beamsplitter; G: glass plate; P: pinhole; I_f: forward pump beam; I_p: probe beam; I_c: phase conjugated beam; I_r: reference beam.

dielectric mirror. Using this cell, we were able to increase the region of temporal overlapping of the two pump beams in the CS_2. Figure 2 shows a typical result for reflectivity versus pump pulse energy, using the special designed cell. The ratio between pump and probe pulse energy was 14. The decrease of the slope of the reflectivity versus energy was accompanied by a decrease in quality of the phase conjugated beam, which we attributed to small-scale self-focusing; the pump pulse seems to break up in bright and dark regions, and this same distorted profile is transferred to the phase conjugate beam. The power density at which saturation takes place is about 1 GW/cm^2, although theory predicts a critical power threshold for macroscopic self-focusing in CS_2 at 10^5 W. The short optical path length of the sample cell causes the self-focusing to occur outside the cell. In this case there is no change into the amplitude profile of the beam inside the cell, only a change in the phase profile, i.e., the shape of the wave fronts. According to Akhmanov a 1-mm cell containing a Kerr-like medium acts as a thin lens with focal length z_f, given by[32]

$$z_f = w_O^2/4I_O n_2 L \, , \tag{1}$$

where w_O is the $1/e$ beam radius at the cell entrance, I_O is the intensity at the beam center, n_2 the nonlinear refractive index, and L the cell length. At a power level $I_O = 1$ GW/cm^2, $w_O = 3$ mm and $L = 1$ mm, $z_f = 5$ m in the case of CS_2. This shows that the short interaction region in picosecond FWM, permitting the use of a short sample cell, reduces the effects of macroscopic or whole beam self-focusing.

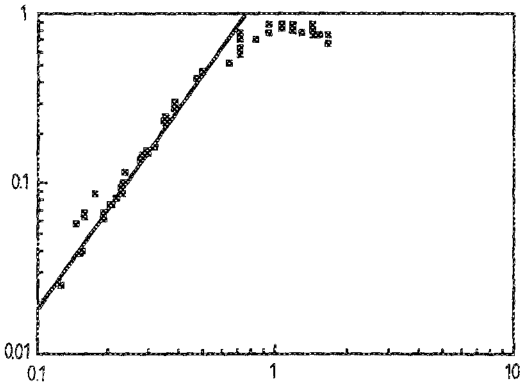

FIG. 2. Phase-conjugate reflectivity of CS_2 in a 2-mm cell vs the pump energy in the forward pump beam, using a sample cell with retroreflection mirror within the sample. The pump:probe pulse energy ratio is 14:1. The curve clearly indicates the negative deviation from a square dependence of the reflectivity on the pump energy at high energies and is typical for most experiments we performed.

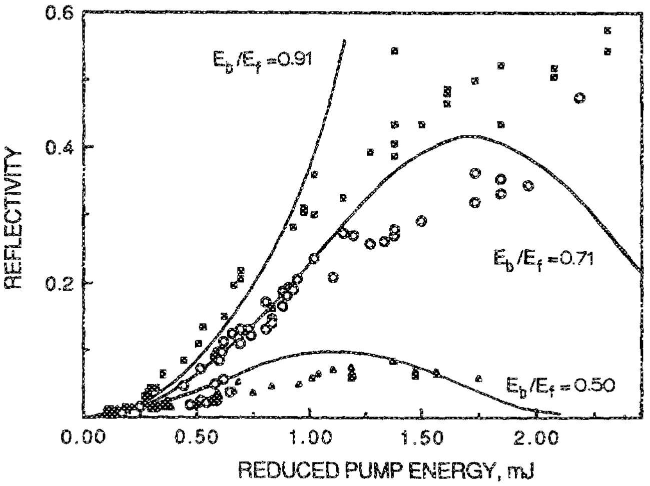

FIG. 3. Phase conjugate reflectivity of CS_2 in a 2-mm sample cell vs the reduced pump energy $E_r = (E_f E_b)^{1/2}$, for three different values of the ratio of backward to forward pump pulse energy E_b/E_f: (▩) 0.91; (○) 0.71; (▲) 0.50. The solid lines are calculated curves, assuming a $1/e$ beam radius of 1.5 mm. The ratio of forward pump pulse energy to probe pulse energy was 14:1.

In order to determine the influence of a difference in energy of the forward and backward pump pulses, the conjugate reflectivity versus pump pulse energy was measured using a retroreflector with a reflectivity less than unity. Figure 3 shows reflectivity versus reduced energy curves for three different values of the backward to forward pump pulse energy ratio E_b/E_f. Taking into account the Fresnel losses at the sample cell window, we performed measurements for the following values of E_b/E_f: 0.91, 0.71, and 0.50. Reduced pump energy E_r is defined as the geometric mean of the forward and backward pump pulse energies E_f and E_b: $E_r = \sqrt{E_f E_b}$. Examination of the plot on Fig. 3 makes evident the sharp decrease in efficiency with increasing the energy difference between forward and backward pump pulses. The solid lines represent the theoretical curves obtained using the model described in Ref. 23. The wave vector $\mathbf{k}$ of an electromagnetic field depends on the intensity in a nonlinear medium since the refractive index is dependent on the electromagnetic field strength. Therefore, for two counterpropagating fields with identical vacuum wave vectors $\mathbf{k}_f$, $\mathbf{k}_b$, within a nonlinear medium, $\mathbf{k}_f + \mathbf{k}_b \neq 0$, resulting in a phase mismatch of the four wave mixing process. If, however, the vacuum intensities are equal, then $\mathbf{k}_f + \mathbf{k}_b = 0$, and there should be no dephasing. The mismatch can be calculated as a function of the energy ratio r and the forward pump pulse intensity I_f, analogous to Ref. 31,

$$|\Delta \mathbf{k}| = |\mathbf{k}_f + \mathbf{k}_b| = \tfrac{9}{2}\omega\sqrt{\mu/\epsilon}\,\chi^{(3)}(1-r)I_f \, . \tag{2}$$

Using the nonlinear coupling coefficient κ,

$$\kappa = (\omega/2)\sqrt{\mu/\epsilon}\,\chi^{(3)}\sqrt{I_f I_b} \, , \tag{3}$$

with I_f and I_b being the intensities of forward and backward pump beam, expression (2) can be written as follows:

$$|\Delta \mathbf{k}| = 9\kappa[(1-r)/\sqrt{r}]I_f \, . \tag{4}$$

Using expression (4), the phase conjugate reflectivity R with asymmetric pump beams can be calculated as follows:

$$R = \left(\frac{\sin^2(\kappa L\sqrt{1+\rho^2})}{\cos^2(\kappa L\sqrt{1+\rho^2}) + \rho^2}\right), \tag{5}$$

where $\rho = \tfrac{9}{2}[(1-r)/\sqrt{r}]$ and L is the interaction length of the pump and probe pulses inside the nonlinear medium. The theoretical curves in Fig. 3 were calculated using the value of $\chi^{(3)} = 2 \times 10^{-31}$ (SI units), leading to the following expression for the coupling constant in CS_2:

$$\kappa = 0.21 I_f \ (\text{MW/cm}^2\,\text{m}) \, . \tag{6}$$

The curves were calculated assuming a beam diameter of 3 mm. We see that for the ratio $E_b/E_f = 0.55$ the theory predicts very well the maximum reflectivity. The difference of maximum position of theoretical and experimental curve is attributed to the fact that we only made an estimate of the beam diameter. These results show that for ratios E_b/E_f of 0.71 and 0.91 and at pump energies higher than 0.75 mJ, the experimental data for the phase-conjugate reflectivity fall below the theoretically expected values, even when the dephasing due to pump pulse energy asymmetry is taken into account. The experimental data shown in Fig. 1 lead to the same conclusion, because in this experiment the pump pulse difference was minimized. From this we conclude that pump pulse asymmetry is not the limiting factor for the phase conjugate efficiency in our experiments.

A second important factor which limits the efficiency of phase conjugation is the depletion of the pump beams. It is obvious that the four-wave mixing process can never produce a conjugate pulse which has a higher energy density or fluence than that of the pump pulses. However, this effect has only seldom been taken into account when solving the coupled-wave equations for the OPC by DFWM.[24,29] Numerical solutions to these equations, incorporating pump depletion, indicate no significant depletion effects when the pump to probe intensity ratio is higher than 10.[29] In Fig. 4 we compiled the results of six different experiments, each having a different ratio of probe to pump *fluence* F_{pr}/F_f. The maximum reflectivity we could obtain in each experiment is plotted as a function of the F_{pr}/F_f. In all experiments, except the one with $F_{pr}/F_f = 5$, this maximum was the saturation level of the reflectivity occurring at pump pulse energy levels above 2 mJ. Note that the power dependence in each of these experiments was measured by varying the intensity of the source beam so as to obtain reflectivity data at a constant pump to probe fluence ratio. In the normal setup, $F_{pr}/F_f = 0.05$. In order to investigate the influence of variations in F_{pr}/F_f, this ratio was decreased by neutral density filters in the probe beam. At the lowest value of F_{pr}/F_f we obtained a reflectivity of more than 300%, which means phase conjugated amplification of the probe pulse. The reflectivity versus energy curve for this experiment is shown in Fig. 5. Note that this curve shows very little saturation effect, as compared to Fig. 2 for example. The cell length was 2 mm in this case. In the other extreme, where the ratio F_{pr}/F_f was increased to 5, by focusing the probe beam into the sample cell, a reflectivity of only 0.1 could be obtained. The probe beam was focused into the center of the collimated pump beams, reducing its radius by a factor of 10, causing the fluence of the probe beam to increase by a factor of 100. However, focusing of the probe beam has been shown to increase the quality of the phase conjugation.[27,28] Under certain conditions a lens transforms the probe beam to its spatial Fourier transform in the focal plane, located in the interaction region of the pump beams. In this case, the Gaussian reflectivity profile determined by the product of the pump pulse transverse intensity profiles, acts as a spatial filter, because the reflectivity is highest in the center of the Fourier plane, where the low spatial frequency components of the probe beam are concentrated. We observed that in this case the phase conjugated pump beam had the same diameter as the probe beam, while the phase conjugated reflection of a collimated probe beam reduces its beam radius. Our results however indicate that focusing of the probe beam leads to a stronger dependence of the PC efficiency on the energy of the probe pulse, reducing the fidelity of the phase conjugation.

Self-focusing of the pump beams is very important as a limiting factor for the fidelity. Although whole beam self-focusing was never observed, the phase-conjugated beam at high pump pulse energy was degraded by small scale self-focusing, leading to beam breakup. Experiments using a 10-mm sample cell length, indicate a lower maximum reflectivity than 2-mm-long cells. Normally one would expect a slight increase due to the increased overlap region. We attribute this effect to an increase of the wave front distortions induced by small scale self-focusing in a longer sample cell.

In this discussion, we have generally used pump energy rather than pump intensity as our independent parameter. It was not possible to measure accurately the beam diameters used in these experiments. We estimate variation of the beam

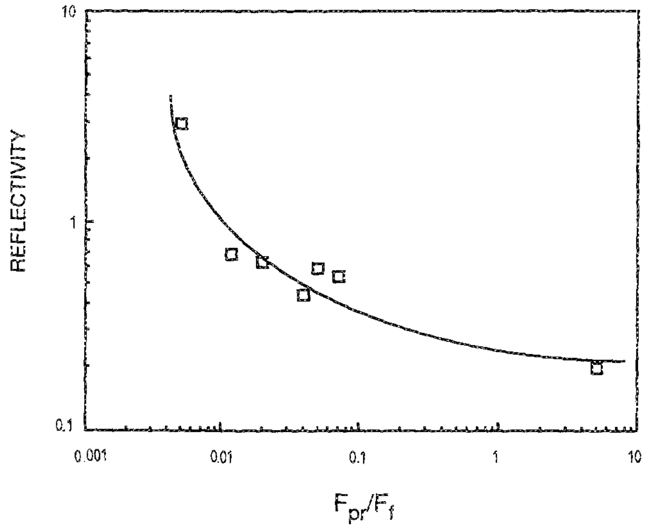

FIG. 4. Maximum value of the phase conjugate reflectivity, as obtained from different experiments, plotted vs the ratio of probe pulse fluence to forward pump beam fluence F_p/F_f.

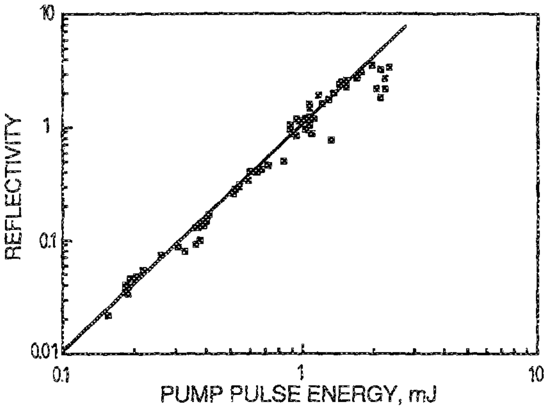

FIG. 5. Phase conjugate reflectivity of CS_2 in a 2-mm cell, at a pump to probe energy ratio of 500:1, plotted vs forward pump pulse energy. A maximum reflectivity of 3 has been observed, i.e., an amplification of the phase conjugated reflection of the probe beam.

diameters to be 20% from experiment to experiment since realignment was involved. In addition to this, slight variations of the pulse width could occur due to changing concentration of the passive modelocker dye.

IV. CONCLUSION

We have measured the efficiency of the phase conjugate reflection of picosecond pulses in CS$_2$, a transparent optical Kerr medium. We obtained very high reflection efficiencies although some saturation of the reflectivity at high fluences was observed. We attribute this to "small scale" self-focusing, which is a typical problem in high energy optical systems. The increased efficiency for the phase conjugation of picosecond pulses in Kerr media is explained by a reduction of the influence of processes such as SBS and whole beam self-focusing, which limit the phase conjugate efficiency of nanosecond pulses. In addition, we observed a strong dependence of the phase conjugation efficiency on the ratio of the pump to probe fluences.

ACKNOWLEDGMENTS

This work was supported in part by USAFOSR Grant No. F29601-87-K-0057 and Rockwell International Corporation Grant No. B8X197858.

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