Time-dependent multiphoton absorption and optical Kerr effect in liquid CS$_2$

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Multiphoton absorption processes in liquid CS$_2$ were observed when the power density of 1.05 μm ps laser pulse exceeded 4 GW/cm^2. Laser pulse distortion was achieved involving the combination of multiphoton absorption and optical Kerr effect. These effects can affect the overall response kinetics of Kerr shutters for extracting temporal information on chemical systems.

Knowledge of multiphoton absorption processes[1,2] is important to the propagation of high power laser pulses[3,4] through transparent materials. Liquid CS$_2$ is transparent in the visible and near infrared region; however, multiphoton processes are possible due to the strong electronic absorption band of the CS$_2$ molecules at about 3500 Å.[5] In this paper, we report on the first measurement of the time dependent two-photon and three-photon absorptions in CS$_2$ liquid using very intense picosecond laser pulses. We have used the combination of multiphoton absorption and optical Kerr effect to distort the envelope of an optical pulse. This has important consequences on the operation of the Kerr shutter for extracting kinetic information on chemical systems.

The experimental setup for measuring the time-dependent, nondegenerate, two-photon (0.53 plus 1.06 μm pulses) absorption is shown in Fig. 1(a). The setup is similar to the single shot Kerr technique.[6-9] A single 1.054 μm (1.05) pulse with ~ 10 ps duration emitted from an amplified mode-locked Nd:phosphate glass laser system[10] was used to produce the transitory multiphoton absorption in a 1 cm CS$_2$ cell (C$_2$). The pulse was selected near the peak of the pulse train. A second harmonic 0.527 μm (0.53) pulse was used to probe the nonlinear absorption change. The 1.05 μm pulse was incident on the sample (C$_2$) at an angle of approximately 3° to the propagation direction of 0.53 μm beam. The 1.05 μm beam was focused to a diameter of about 3 mm at the sample cell. The beam diameter of 0.53 μm was expanded and passed through an echelon. In this manner, the adjacent segments of the interrogating beam (30 pulses) were delayed with a step progression of 2.5 ps per step. This oblique wave front probed the transitory absorption induced by 1.05 μm pulse in the sample. The area of the segmented 0.53 μm beam profile was focused to a size of 1 mm at the sample site to insure complete overlap with the 1.05 μm beam profile inside the sample. The 0.53 μm step signals transmitted through the CS$_2$ cell were imaged by a camera lens onto the target face of a GBC SIT video camera. The video signal was processed by a Hamamatsu temporal analyzer. The time axis of the oblique front was calibrated by moving a delay prism in the 0.53 μm laser beam path. The energy of the 0.53 and 1.05 μm

pulses were measured by a fast photodiode and oscilloscope on each laser shot. The energy was calibrated using a Hadron thermopile. The maximum power density used in this experiment for the 1.05 μm pulse was ≈ 20 GW/cm^2 and for the 0.53 μm pulse was ≈ 100 kW/cm^2.

A typical normalized intensity profile of the time dependent absorption of the 0.53 μm beam induced by an intense 1.05 μm pulse is shown in Fig. 2. The dotted curve is the transmitted signal of the 30 segmented probe 0.53 μm pulses when the 1.05 μm pulse is blocked. When an intense (≈ 10 GW/cm^2) 1.05 μm pulse (≈ 10 ps) was propagated into the CS$_2$ sample during the echelon
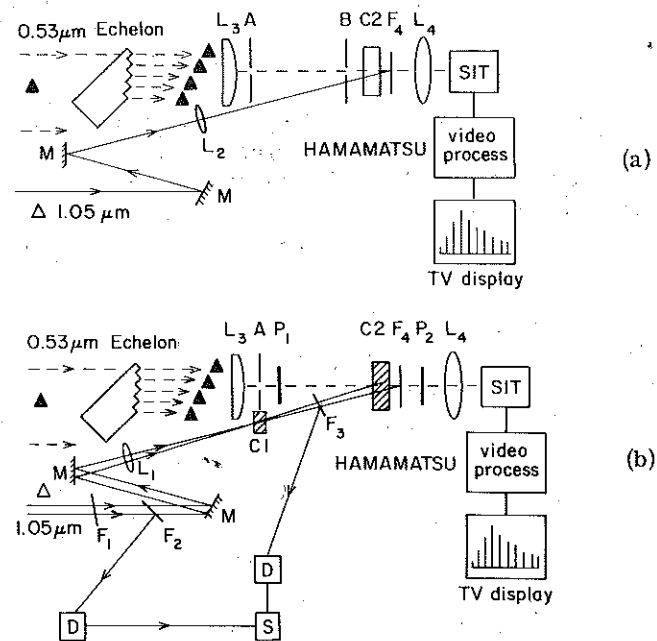

FIG. 1. A schematic diagram of the experimental setup for (a) two-photon absorption and (b) three-photon absorption. A: 1 mm slot, B: small aperture, C$_1$: 1 cm long CS$_2$ cell, C$_2$: 1 cm long CS$_2$ cell, D: photodiodes, F$_1$: Hoya neutral density filter, F$_2$: Corning 7-99 filter, F$_3$: Hoya neutral density filter, F$_4$: Corning 4-96 filter and two Ditric 3C narrow band filters, L$_1$: $f=28$ cm lens, L$_2$: $f=40$ cm lens, L$_3$: $f=30$ cm lens, L$_4$: 50 mm /f1.8 camera lens, M: dielectric coated mirrors, S: Tektronix 519 oscilloscope, P$_1$ and P$_2$: Polaroid HN22 polarizers; polarized along the directions 45° and −45°, respectively, TV: Hamamatsu temporal analyzer video system, echelon: 30 pieces of 1 mm thickness glass oriented at Brewster's angle.

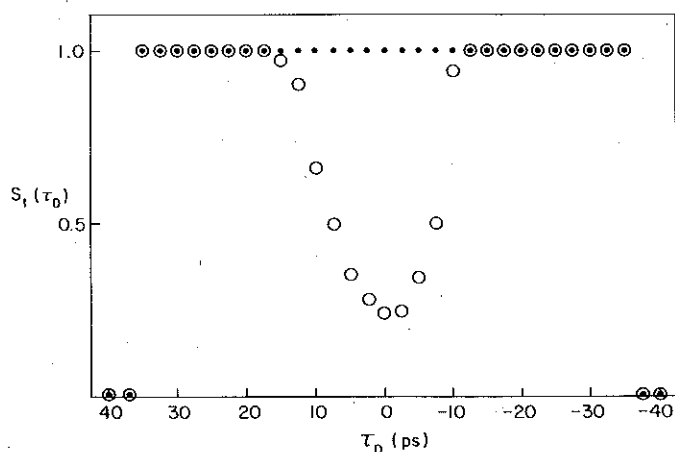[a] Present address: NCR Corporation, P.O. Box 728, Cambridge, Ohio 43725.

FIG. 2. The normalized transmitted signal of 0.53 μm pulses at various delay times τ_D with respect to the arrival time of the 1.05 μm pulse into the CS_2 sample. A dot ● represents the measured transmitted signal of the 0.53 μm pulse without the 1.05 μm pulse being present. The power density at CS_2 cell C_2 is about 10 GW/cm^2. A circle o represents the measured signal of the 0.53 μm pulse with an intense 1.05 μm pulse incident on the CS_2. The FWHM of the hole is $\simeq 15 \pm 3$ ps.

time domain (≈ 75 ps), an absorption hole was created in the 0.53 μm profile (circles) around the zero time delay. The peak of the absorption hole for the 0.53 μm pulse was measured to be roughly proportional to the intensity of the 1.05 μm pulse and the relative change was found to be independent of the intensity of the 0.53 μm pulse. This is consistent with the intensity change of the 0.53 μm pulse caused by two-photon absorption of 1.05 and 0.53 photons.[11,12] The induced absorption was found to be independent of the polarization of both beams. Neglecting the effect of higher order absorptions, the two-photon absorption coefficient is estimated to be 10^{-10} cm/W.
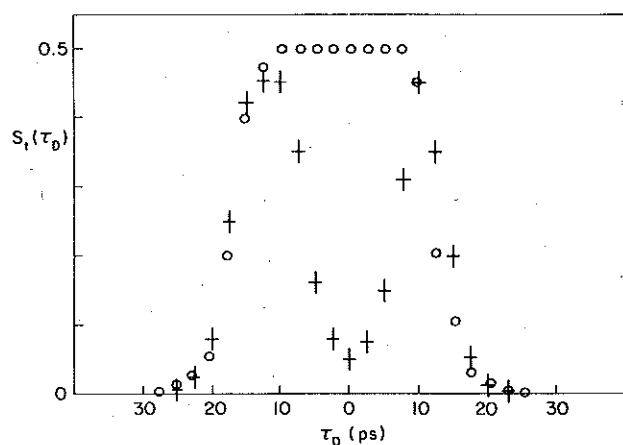

FIG. 3. The normalized Kerr transmitted signal of 0.53 μm pulses at various delay times τ_D with respect to the arrival time of the 1.05 μm pulse into the CS_2. The power density of the 1.05 μm pulse at CS_2 cell C_2 was about 10 GW/cm^2. The circles o are the calculated transmission signals through the Kerr gate[7] without taking into account the induced two-photon absorption process. The crosses + are the measured normalized 0.53 μm Kerr signals.
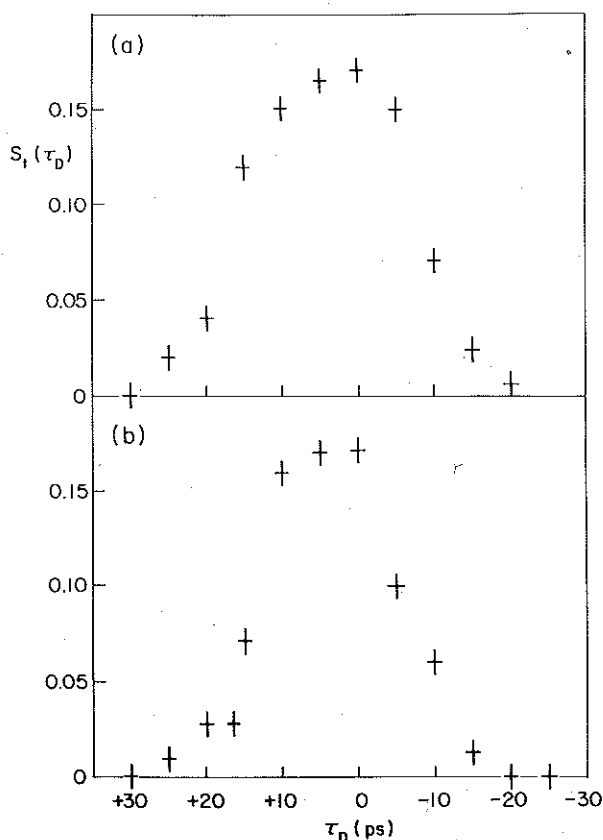

FIG. 4. The transmitted Kerr signal of 0.53 μm pulses at various delay times τ_D with respect to the arrival time of the 1.05 μm pulse into the CS_2 cell C_2: (a) The 1.05 μm pulse traveled through the CS_2 cell C_1 where three-photon absorption occurred. The power density of the 1.05 μm pulse in C_1 was about 20 GW/cm^2. (b) The CS_2 cell C_1 was removed. The power density of the 1.05 μm at the CS_2 cell C_2 was maintained in both cases at about 100 MW/cm^2 by inserting different neutral density filters F_3. The FWHM are 25 and 17 ps for the profiles (a) and (b), respectively.

When a pair of crossed polarizers P_1 and P_2 were placed into the 0.53 μm beam path before and after the CS_2 cell (C_2) in Fig. 1(a), a combined intensity profile of two-photon absorption and the optical Kerr effect was measured. The combined transmitted intensity profile is displayed in Fig. 3. The power density of the 1.05 μm pulse used in this measurement is about 10 GW/cm^2. The intensity dependence of the Kerr effect has been discussed in detail by Ho and Alfano.[7] The transmitted Kerr intensity at $\tau = 0$ at ultrahigh intensities saturates. Without the two-photon absorption mechanism, a broadened optical Kerr gate intensity profile is expected.[7,13] The circles in Fig. 3 are the calculated transmission Kerr signals[7] without taking into account the induced two-photon absorption processes. At such a high power density of the pumped 1.05 μm pulse ≈ 10 GW/cm^2, multiphoton induced absorption for the 0.53 μm beam is expected. The crosses are the measured normalized Kerr signals for a 10 GW/cm^2, 1.05 μm pulse. The induced absorption causes the segmented 0.53 μm pulse to be reshaped into two pulses after passing through the Kerr gate.

The experimental setup to determine the effect of three-photon absorption on the Kerr kinetics in CS$_2$ liquids is shown in Fig. 1(b). Three-photon absorption of the 1.05 μm pulse occurs in the first CS$_2$ cell (C$_1$). The transmitted signals of the 0.53 μm probe pulses are displayed versus the delay time with respect to the arrival of the 1.05 μm pulse for two different cases: (a) The 1.05 μm pulse was focused into a CS$_2$ cell (C$_1$) to a peak power density of about 20 GW/cm^2 before propagating through the Kerr CS$_2$ cell (C$_2$); and (b) the 1.05 μm pulse was still focused at the site of the cell (C$_1$) and passed through the Kerr cell (C$_2$) but the sample cell (C$_1$) was removed. The intensity of the 1.05 μm pulse at the Kerr cell site was maintained at about 100 MW/cm^2 for both cases. There is a noticeable difference between the two intensity profiles displayed in Figs. 4(a) and 4(b). The intensity profile of the transmitted 0.53 μm pulses in Fig. 4(a) is wider than the curve shown in Fig. 4(b). The FWHM are 25 and 17 ps for curves (a) and (b) in Fig. 4, respectively. This indicated that the pulse shape of the 1.05 μm laser was changed via a three-photon absorption process that occurred in the CS$_2$ sample (C$_1$) cell. To cause the wider profile, the peak of the 1.05 μm pulse was reduced by three-photon absorption proportionally more than its wings. The transmitted Kerr signal in this case (a) fluctuated much more than a normal CS$_2$ Kerr gate (b) from laser shot to shot profiles under similar conditions.

In conclusion, the effects of multiphoton absorption on the optical Kerr effect were observed under very intense picosecond laser pulse excitation in liquid CS$_2$.

Shaping of laser pulses and altering the response kinetics of a Kerr shutter can be accomplished using these effects.

ACKNOWLEDGMENTS

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