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

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(Received 27 August 1980; accepted 10 October 1980)

Multiphoton absorption processes in liquid CS$_2$ were observed when the power density of 1.05 $\mu$m 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 is important to the propagation of high power laser pulses 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. 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 $\mu$m pulses) absorption is shown in Fig. 1(a). The setup is similar to the single shot Kerr technique. A single 1.054 $\mu$m (1.05) pulse with ~10 ps duration emitted from an amplified mode-locked Nd:phosphate glass laser system 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 $\mu$m (0.53) pulse was used to probe the nonlinear absorption change. The 1.05 $\mu$m pulse was incident on the sample (C$_2$) at an angle of approximately 3° to the propagation direction of 0.53 $\mu$m beam. The 1.05 $\mu$m beam was focused to a diameter of about 3 mm at the sample cell. The beam diameter of 0.53 $\mu$m was expanded and passed through an echelon. In this manner, the adjacent segments of the interrogating beam (30 pulses) were delayed by a step progression of 2.5 ps per step. This oblique wave front probed the transitory absorption induced by 1.05 $\mu$m pulse in the sample. The area of the segmented 0.53 $\mu$m profile was focused to a size of 1 mm at the sample site to insure complete overlap with the 1.05 $\mu$m beam profile inside the sample. The 0.53 $\mu$m step signals transmitted through the CS$_2$ cell were imaged by a camera lens onto the target face of a GBC SIT vidicon 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 $\mu$m laser beam path. The energy of the 0.53 and 1.05 $\mu$m pulses were measured by a fast photodiode and oscilloscope on each laser shot. The energy was calibrated using a Fladron thermopile. The maximum power density used in this experiment for the 1.05 $\mu$m pulse was ~20 GW/cm$^2$ and for the 0.53 $\mu$m pulse was ~100 kW/cm$^2$.

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

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FIG. 2. The normalized transmitted signal of 0.53 μm pulses at various delay times τ₀ with respect to the arrival time of the 1.05 μm pulse into the CS₂ 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 CS₂ cell C₁ is about 10 GW/cm². A circle ○ represents the measured signal of the 0.53 μm pulse with an intense 1.05 μm pulse incident on the CS₂. The FWHM of the hole is ±15 ± 3 ps.

The 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. 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 10⁻¹⁵ cm/W.

FIG. 3. The normalized Kerr transmitted signal of 0.53 μm pulses at various delay times τ₀ with respect to the arrival time of the 1.05 μm pulse into the CS₂. The power density of the 1.05 μm pulse at CS₂ cell C₁ was about 10 GW/cm². The circles ○ are the calculated transmission signals through the Kerr gate 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 τ₀ with respect to the arrival time of the 1.05 μm pulse into the CS₂ cell C₂: (a) The 1.05 μm pulse traveled through the CS₂ cell C₁ where three-photon absorption occurred. The power density of the 1.05 μm pulse in C₁ was about 20 GW/cm². (b) The CS₂ cell C₁ was removed. The power density of the 1.05 μm at the CS₂ cell C₂ was maintained in both cases at about 100 MW/cm² by inserting different neutral density filters P₀. The FWHM are 25 and 17 ps for the profiles (a) and (b), respectively.

When a pair of crossed polarizers P₁ and P₂ were placed into the 0.53 μm beam path before and after the CS₂ cell (C₂) 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/cm². The intensity dependence of the Kerr effect has been discussed in detail by Ho and Alfano. The transmitted Kerr intensity at τ = 0 at ultrahigh intensities saturates. Without the two-photon absorption mechanism, a broadened optical Kerr gate intensity profile is expected. The circles in Fig. 3 are the calculated transmission Kerr signals without taking into account the induced two-photon absorption processes. At such high power density of the pumped 1.05 μm pulse ≈ 10 GW/cm², multiphoton induced absorption for the 0.53 μm beam is expected. The crosses are the measured normalized Kerr signals for a 10 GW/cm², 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 \textit{CS}_2 liquids is shown in Fig. 1(b). Three-photon absorption of the 1.05 \( \mu \text{m} \) pulse occurs in the first \textit{CS}_2 cell (C_1). The transmitted signals of the 0.53 \( \mu \text{m} \) probe pulses are displayed versus the delay time with respect to the arrival of the 1.05 \( \mu \text{m} \) pulse for two different cases: (a) The 1.05 \( \mu \text{m} \) pulse was focused into a \textit{CS}_2 cell (C_1) to a peak power density of about 20 GW/cm\(^2\) before propagating through the Kerr \textit{CS}_2 cell (C_2); and (b) the 1.05 \( \mu \text{m} \) 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 \( \mu \text{m} \) 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 \( \mu \text{m} \) 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 \( \mu \text{m} \) laser was changed via a three-photon absorption process that occurred in the \textit{CS}_2 sample (C_1) cell. To cause the wider profile, the peak of the 1.05 \( \mu \text{m} \) 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 \textit{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 \textit{CS}_2. Shaping of laser pulses and altering the response kinetics of a Kerr shutter can be accomplished using these effects.

**ACKNOWLEDGMENTS**

This research was supported in part by NSF grants CPE 7920192, PCM 7714966, AFOSR-80-0079, and a grant PSC-BHE from CUNY. R. R. Alfano is an Alfred P. Sloan Fellow.