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TITLE

Optical Limiting Studies in a Carbon-Black Suspension for Subnanosecond and Subpicosecond Laser Pulses

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Optical limiting studies in a carbon-black suspension for subnanosecond and subpicosecond laser pulses

Denis Vincent, Stéphane Petit, and See Leang Chin

This paper presents results of measurements of short (0.3 ps, 0.2 ns, and 10 ns) laser pulse transmissions through a liquid suspension of fine carbon particles (named CBS for "carbon-black suspension") at input-pulse energies up to 10 mJ. The 10-ns pulses came from a Nd:YAG laser at 1064 nm, and the shorter pulses came from a Ti:sapphire laser at 800 nm. Limiting was observed with the 10-ns and the 0.2-ns laser pulses, but the 0.3-ps pulses produced white light and underwent the same level of attenuation in the solvent and in the CBS. © 2002 Optical Society of America

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1. Introduction

The behavior of carbon-black suspension (CBS) against nanosecond pulses at visible and near-infrared wavelengths has been studied extensively in various laboratories.[1-6] Some research teams have also reported the CBS behavior against laser pulses that are a few tens of ps long. In particular, it has been noted that the transmittances of 30–42 ps and 14-ns pulses become nonlinear at the same threshold energy within a factor of two[3,7] and that the nonlinearity begins approximately 50–100 ps after the excitation and becomes maximum after about 0.4–0.5 ns.[8,9] These latter measurements were made with carbon suspensions in either water, water and ethylene glycol, or ethanol. It has also been observed that fine carbon particles suspended in chloroform with an appropriate surfactant have a lower threshold for nonlinear transmission than CBS does in ethanol or water.[10] A series of experiments was carried out to measure the nonlinear transmission of this high-performance CBS for pulse lengths of 0.3 ps and 0.2 ns at 800 nm and to compare it to the values obtained with 10 ns pulses at 1064 nm. The choice of the wavelengths was dictated by the availability of the sources at these pulse lengths.

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2. Measurement System

A schematic diagram of the optical system used for the measurements appears in Fig. 1. The laser sources were either a Ti:sapphire laser at 800 nm that could deliver 0.3-ps or 0.2-ns pulses at a rate of 980 pulses/s and with pulse energy up to a few hundreds of microjoules, or a Nd:YAG laser at 1064 nm that could deliver 10-ns pulses in single-pulse mode at a pulse energy of up to 10 mJ at the cell. Both laser beams passed through a beam expander and a variable attenuator before reaching lens L1. The aperture in front of this lens was largely overfilled with the beams. When the Ti:sapphire laser was used, the input energy was measured with a power meter at 980 pulses/s before the beam expander and the pulse selector, whereas the input energy for the Nd:YAG laser was measured with an energy meter after the beam expander. In both cases, the output energy was measured with an energy meter (pyroelectric detector for the subnanosecond pulses and silicon detector for the 10-ns pulses). All the energy meters were triggered externally by a pick-up photodiode.

Four Ti:sapphire laser pulses could pass through the pulse selector during its opening time (4 ms). The 1-ms time interval between pulses allows the suspension to recover from the precedent pulse.[2] However, at high-input energy, some carbon particles disappear at each laser pulse, and consequently the measured transmission was probably slightly larger than in the single-pulse case.

Lens L1 was a simple biconvex lens with a 100-mm focal length. Its focal spot had a nice central lobe (FWHM diameter of 8 μm at 1064 nm) containing

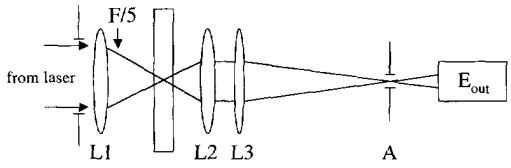

Fig 1 Optical arrangement to measure the nonlinear transmission

about 25% of the total energy. The remaining energy was spread over more than 2 mrad in the focal plane when L1 was apertured to f/5. When this spot was imaged in the analysis plane containing the aperture A (Fig. 1) by lenses L2 and L3, 62% of the energy of the 10-ns pulses at 1064 nm passed through an aperture subtending an angle of 2 mrad, whereas 84% of the energy of the 0.2-ns pulses at 800 nm could pass through it. Lens L2 was a 50-mm-focal-length achromat, and lens L3 was a 500-mm-focal-length achromat for the measurements at 800 nm and a 1000-mm focal length meniscus at 1064 nm. At both wavelengths, aperture A was adjusted to subtend an angle of 2 mrad.

For these measurements a clear cell, containing only the solvent and the surfactant, and a cell with the CBS were prepared. Both cells were standard 2-cm diameter and 1-cm pathlength fused-silica spectrophotometer cells with a stopper. Their clear sides allow the observation of the phenomena occurring in the cell The suspension named DREV CBS-100 (prepared in DREV's laboratories) has been described elsewhere.[1] It contains fine carbon particles with diameters of approximately 50 nm that floculate together to form loose flocs that have an average diameter of ~1 μm. These flocs are more efficient than fine particles against long pulses and do not degrade significantly the performance of the suspension against short pulses. The cells were positioned such that the focal plane of lens L1 was at the center of the cell With 10-ns pulses, the scattering from CBS at energy just slightly higher than threshold was used for this purpose whereas for subnanosecond pulses, the white light generated in the cell by the 0.3-ps pulses was used to center the cell.[11] The shortest pulse length that could be achieved with our system was 0 3 ps (measured with an autocorrelation technique in previous experiments), and this was indicated by the intense white light generated by the laser beam focused in a water cell as a simple verification technique.

3. Results

Figures 2–4 display the measured nonlinear transmission as a function of the input energy per pulse. The nonlinear transmission is defined as E_{out}/T_0E_{in}, where E_{out} and E_{in} represent the output energy through the analysis aperture and the input energy at the cell, respectively, and T_0 represents the linear transmission of the cell. In the arrangement at 800 nm with the pulse selector, E_{in} and E_{out} that were used to evaluate the nonlinear transmission correspond to the total energy during the selector opening time. The measurements at 1064 nm used single-pulse irradiation.

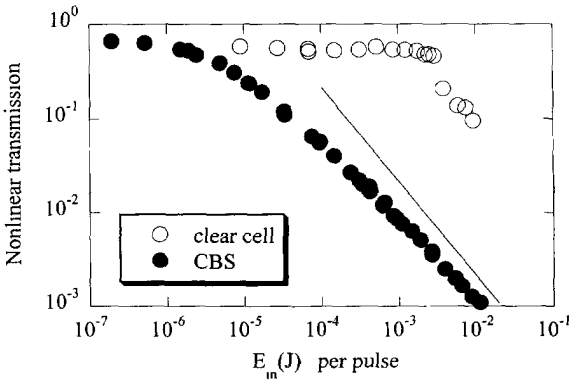

Fig 2 Nonlinear transmission of 10-ns pulses at 1064 nm

In Fig. 2, the transmission of the CBS cell for 10-ns pulses begins to decrease at an input energy near 1 μJ, and its slope tends towards a value of −1 at high input energy; the straight line has a slope of −1. At an input energy of a few millijoules, the CBS cell attenuates the laser pulses by more than 2 orders of magnitude with respect to the clear cell. Laser-induced breakdown in the clear cell at an input energy larger than 3 mJ also attenuates the laser pulses. The low-energy transmission in this clear cell corresponds to the transmission of the analysis aperture.

The transmission of the cells for 0.2-ns pulses appears in Fig. 3. The transmission of the CBS cell begins also to decrease at an input energy near 1 μJ, and its slope tends towards −1 at high-input energy. Breakdown occurs in the clear cell at an energy approximately 24 times lower than with 10-ns pulses, which is consequent with the higher peak intensity in the shorter pulses. The nonlinear transmission is approximately 3 times less in the CBS cell than in the clear cell, whereas it is 2 orders of magnitude lower in the 10-ns case. Finally, the low-energy transmission (0.5) of the clear cell is lower than the transmission of the analysis aperture (0.84) for an unknown reason.

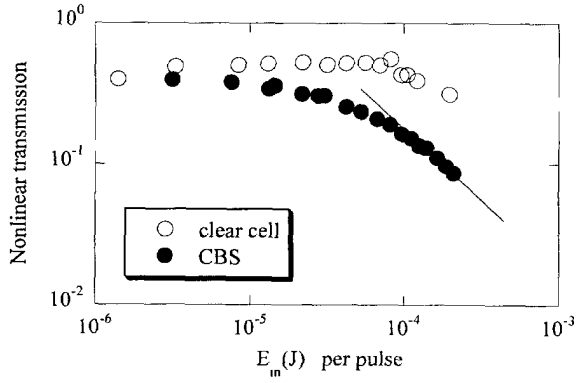

Fig 3 Nonlinear transmission of 0 2-ns pulses at 800 nm

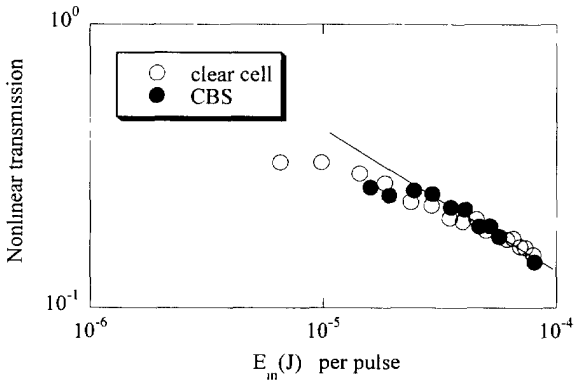

Fig 4 Nonlinear transmission of 0.3-ps pulses at 800 nm

In the subpicosecond regime, white light was generated in both cells, and the nonlinear transmission changed the same way in the clear cell and the CBS cell, as shown in Fig. 4. The decrease in transmission comes possibly from the spreading of the laser spot in the analysis plane, arising from the laser-induced perturbation of the refractive index of the liquid in the cell at high intensity. The slope of this plot at high input energy is -0.5 and means that $E_{out} \propto E_{in}^{0.5}$.

4. Discussion and Conclusion

From the results presented in Figs. 2–4, it appears clearly that the limiting mechanism in the CBS-100 can be initiated in a time shorter than 0.2 ns at an energy comparable with the energy needed to begin blocking much longer pulses. The authors in Refs. 3 and 7 have noted that the threshold energy is the same for nanosecond and subnanosecond pulses. However, the less-efficient limitation for 0.2 ns pulses indicates that this mechanism requires a time larger than 0.2 ns to operate efficiently. The authors in Refs. 8 and 9 have noted that the transmission becomes nonlinear after a time delay between 50 and 100 ps and that the attenuation is maximum after a delay of about 500 ps at high-input energy. Since the heating of the carbon particle requires the same amount of energy whatever the pulse length, and since it takes at least 1 ns at a growth rate of about 200 nm/ns[6] for a bubble to increase the size of the scattering center from 50 nm to 200 nm at which scattering is more noticeable, these observations are compatible with an explanation of optical limitation in CBS that is based on the generation of bubbles. However, it is evident that CBS does not limit efficiently at pulse lengths smaller than 1 ns. In particular, a clear cell of chloroform shows the same level of attenuation of 0.3-ps pulses as a CBS cell does.

The values of threshold energy found in Figs. 2 and 3 depend on the laser spot size at focus. Since the lens L1 presents a large blur at its focal point, the spot sizes at 800 nm and 1064 nm are approximately the same Accordingly, the calculated *apparent* threshold fluence is given by 25% of 1 μJ in a paraxial focal spot having a diameter of about 8 μm at half-maximum. That leads to a value of 0.5 J/cm^2, which is much larger than the value given in Ref. 10, but the aperture used here is much larger, leading to a considerable averaging effect.

In conclusion, this paper has reported the behavior of the DREV CBS-100 optical limiter over a range of pulse lengths covering more than 4 orders of magnitude. It appeared clearly that optical limitation in this suspension begins at a pulse length smaller than 0.2 ns.

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