Optical limiting properties of carbon disulfide at 2.05 μm wavelength

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ABSTRACT

Several types of infrared sensors are based on sensitive focal plane arrays. In such sensors, the intensity will typically increase by a factor $\sim 10^7$ at the focal plane, compared to the intensity of the incoming radiation. Such arrays are thus vulnerable when illuminated with high-intensity laser pulses. One solution for protecting the array against such pulses is to use an optical limiter. We here present results where carbon disulfide (CS_2) has been tested as an optical limiting material against high-energy laser pulses at 2.05 μm wavelength.

Keywords: Optical limiting, z-scan, carbon disulfide, focal plane arrays

1. INTRODUCTION

Several types of electro optic sensors utilize sensitive detectors based on semiconductor materials. Often a focal plane array is used to image the scene. In such sensors, the intensity will typically increase by a factor of the order of 10^7 at the focal plane, compared to the intensity of the incoming radiation. This means that the detector elements can be easily damaged if the sensor is illuminated by a laser. The damage threshold of semiconductor materials, which are illuminated with laser pulses with a duration of some tens of nanoseconds, is typically of the order of 1 J/cm^2.[1] If the sensor is focusing the radiation down to a diameter of 10 μm, damage will occur if the pulse energy exceeds about 1 μJ, which is easily available in pulsed lasers.

One way of protecting a sensor against damage from laser pulses is to use an optical filter that blocks the most common laser wavelengths. This is however not a safe method, due to the development of laser sources with new wavelengths. Another solution is to use an optical limiter, which is a passive device that blocks radiation with high intensity and transmits radiation with low intensity.[2,3] This is achieved by focusing the radiation down to a small spot in the optical limiter material, which has a nonlinear optical response. The nonlinear mechanisms, typically self-focusing and dielectric breakdown,[4] provide high attenuation for radiation with high power, while radiation with low power is transmitted with low attenuation. For defense applications, there are several sensors operating at the wavelengths 1.5–2.5 μm, 3–5 μm, and 8–12 μm, where the atmosphere has high transmission. It is thus of interest to characterize the properties of optical limiters in these wavelength regions.

An optical limiter material should not be damaged by laser pulses with high energy. This makes liquids attractive candidates as optical limiter materials, because liquids are not locally damaged, in contrast to solids. We here present results where carbon disulfide (CS_2) has been tested as an optical limiting material when illuminated with 25 ns pulses with up to 150 mJ energy at 2.05 μm wavelength. The laser beam had a beam quality $M^2 = 1.5$, and the beam was focused inside a CS_2 cell using a lens with an effective f-number of 10 and a focal length of 50 mm. The focus position in the CS_2 cell was reimaged, with magnification 20, onto an aperture with 1.4 mm diameter. Pulse energies of up to 150 mJ were incident on the cell, while at most 0.6 mJ was transmitted through the aperture, due to dielectric breakdown and laser beam breakup in the CS_2 cell at high pulse energies. To our knowledge, these are the first optical limiting experiments performed at 2 μm wavelength using CS_2.

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2. PROPERTIES OF CS_2

An important property of liquids used as optical limiters is the intensity dependent refractive index, $n = n_0 + \gamma I$, where n_0 is the refractive index at low intensities, I is the optical intensity, and γ is the third-order nonlinear refractive index. One of the Kerr active liquids with largest γ is carbon disulfide.[5] Optical limiting in CS_2 has previously been demonstrated at visible wavelengths, in the 0.7–1.1 μm range, and at 10.6 μm.[2,3,6,7]

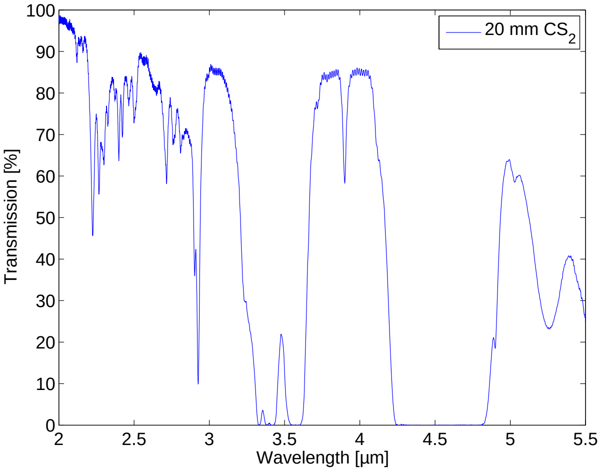

Figure 1. Measured transmission through 20 mm CS_2.

Figure 1 shows measured transmission through a 20 mm thick CS_2 cell using an FTIR spectrometer, where we have corrected for the losses due to the cell windows. The transmission at 2.05 μm through the cell is 96.9%, resulting in an attenuation coefficient of 1.6/m, in good correspondence with previous measurements.[8] We observe from Fig. 1 that there are regions of low transmission, especially near 3.5 μm and 4.5 μm, showing that CS_2 is not suitable for use as an optical limiting material in the entire region from 2–5 μm. Using a standard setup for closed aperture z-scan measurements,[9] we measured a nonlinear refractive index of $\gamma = (2.8 \pm 1.0) \cdot 10^{-18}$ m^2/W in CS_2, for 25 ns long pulses at 2.05 μm, in reasonable agreement with previous reports.[10,11] No transmission changes were observed, within experimental uncertainties, in an open aperture z-scan measurement, implying a nonlinear absorption coefficient of less than $4 \cdot 10^{-13}$ m/W. The linear refractive index at $\lambda = 2.05$ μm wavelength is $n_0 = 1.59$.[11]

3. EXPERIMENTAL SETUP FOR OPTICAL LIMITING

Figure 2 shows a schematic of the optical limiting experiment. The optical limiter itself consisted of the CS_2 cell together with the lenses L1 and L2. The lenses L2 and L3 image the beam in the focus of the CS_2 cell onto the aperture AP, with magnification M equal to the ratio between the focal lengths of L3 and L2, giving $M = 20$. The FWe^{-2}M diameter of the beam incident on L_1 was 5.2 mm, giving an effective f-number $f_\# \approx 10$ for the optical limiter. This resulted in a measured beam diameter of 32 μm in the focus position. The lens L3 and the aperture AP can be thought of as a sensor unit, and the task of the optical limiter is to give an upper limit of the fluence (pulse energy per unit area) that hits the sensor plane, here represented by AP. In some of the limiter experiments, the aperture AP was replaced by a pyroelectric camera, such that the beam in the position of AP could be recorded.

To characterize the optical limiter, we measured the encircled energy after the CS_2 cell.[2] The encircled energy is here defined as the energy transmitted through the aperture AP, which had a diameter of 1.4 mm. This was the smallest aperture diameter that gave good transmission at low pulse energies. The transmission was measured by recording the encircled pulse energy using the energy meter EN2, while simultaneously measuring the incident energy using a reference energy meter (EN1). Reference measurements were necessary due to fluctuations in the pulse energy of the laser. The setup was made for choosing between two alternative reference beam paths. At low input energies, the transmitted beam through a wedge W was used as reference (low energy regime), while

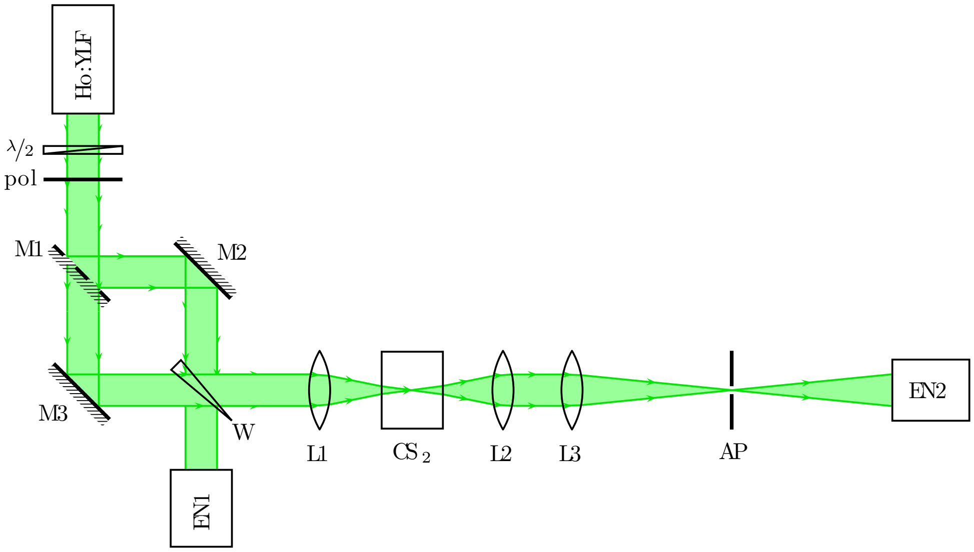

Figure 2. Schematic of the optical limiting setup. Ho:YLF: Holmium laser, $\lambda/2$: half-wave plate at 2.05 μm, pol: polarizer, M1: removable mirror, M2–M3: fixed mirrors, W: wedge, EN1 and EN2: energy meters, CS_2: 20 mm long CS_2 cell, L1–L3: lenses with focal lengths 50 mm, 50 mm, and 1 m, respectively, AP: aperture with a diameter of 1.4 mm.

at high input energies, the reflected beam from the wedge was used as reference (high energy regime). A mirror M1 was inserted into the beam path in the low energy regime, while it was removed in the high energy regime.

The laser used in the experiments was a Q-switched cryogenic Ho:YLF oscillator, which provided 25 ns long pulses at 2.05 μm wavelength, with a beam quality $M^2 = 1.5$, and FWHM line width 0.4 nm.[12] A pulse energy of up to 150 mJ, incident on the CS_2 liquid, was used in these experiments, and the laser was operated with a repetition rate of 1 Hz.

The CS_2 cell was 20 mm long. It was made of aluminum and had uncoated sapphire windows. The estimated peak fluence on the cell windows is $\sim$100 J/cm^2, which was why uncoated sapphire windows was used, because sapphire has a damage threshold of $\sim$200 J/cm^2.[13] In addition, the refractive index of sapphire ($n = 1.7$) matches the refractive index of CS_2 reasonably well.

4. RESULTS

4.1 Observation of dielectric breakdown

When the pulse energy into the cell exceeded about 0.8 mJ, a visible spark was observed in the focus position, due to plasma generated by dielectric breakdown. Well above the threshold for dielectric breakdown, an elongation of the spark was observed, in agreement with previous observations.[3] Images of the breakdown at different input energies are shown in Fig. 3. According to the moving-focus model,[14] the focus position of a nanosecond pulse experiencing self-focusing will extend over an interval from the original focus position and towards the input window, due to the time-varying pulse power. We observed that the elongation of the breakdown region extended towards the input window, as expected from the moving-focus model. Estimating the threshold for self-focusing using the formula[15,16]

$$P_{\mathrm{cr}} = 0.146\frac{\lambda^2}{n_0\gamma}, \tag{1}$$

gives $P_{\mathrm{cr}} = 128$ kW, corresponding to a pulse energy of 3.2 mJ for a pulse duration of 25 ns. The observed dielectric breakdown at 0.8 mJ is significantly lower than the estimated threshold for self-focusing. This indicates that dielectric breakdown happens independent of self-focusing, but could also be due to high-power spikes in the pulses, because the Ho:YLF laser operates on many ($\sim$100) longitudinal modes.

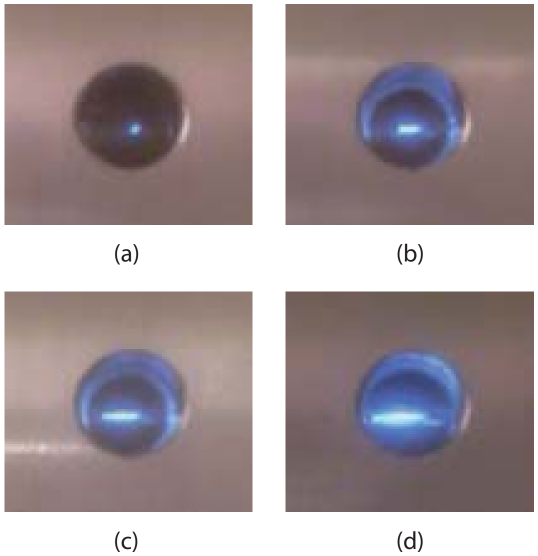

Figure 3. Images of dielectric breakdown in the CS_2 cell at varying input pulse energies. (a) 0.9 mJ, (b) 3.6 mJ, (c) 9.9 mJ, and (d) 17.7 mJ. The images were taken through a hole at the top of the cell.

4.2 Imaging the beam at focus

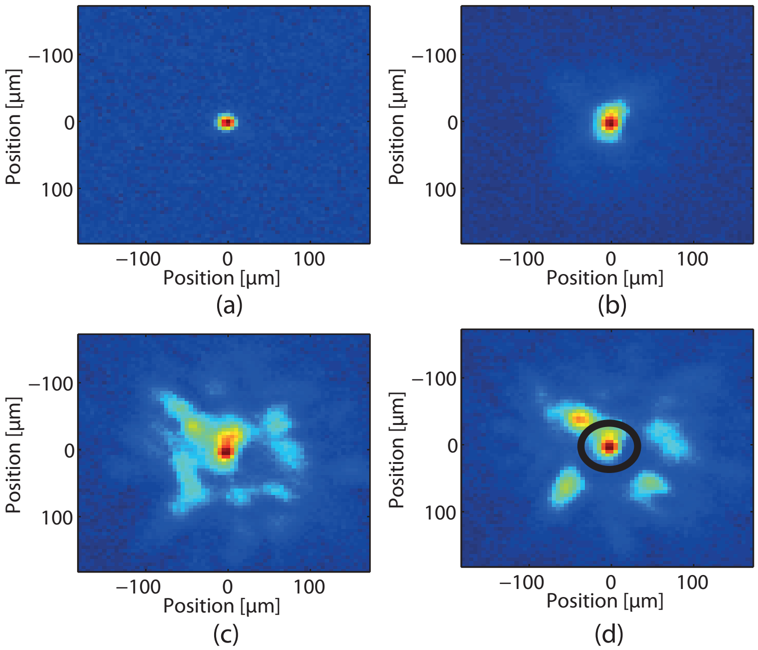

Figure 4. Images of the beam at the focus position in the CS_2 cell. (a) low pulse energy, (b) 7 mJ, (c) 23 mJ, (d) 32 mJ. The black circle in (d) corresponds to how an aperture with diameter 1.4 mm in the position of AP would block parts of the beam.

The beam profile at the focus position in the CS_2 cell was imaged with a magnification $M = 20$ by placing a pyroelectric camera in the position of the aperture AP. Figure 4 shows the beam profile at different pulse energies. Laser beam breakup is clearly apparent in Fig. 4(c) and (d). The images in Fig. 4 represent single pulses, and the filament pattern varied significantly from pulse to pulse, even though the input energy was kept constant. As explained in Sec. 3, we measured the encircled energy by using an aperture AP with diameter 1.4 mm. In Fig. 4(d) it is indicated how such an aperture would block parts of the beam. We can estimate the peak fluence in the image as

$$F_{\max} \approx \frac{2E_{\mathrm{en}}}{\pi(M\omega_0)^2}, \tag{2}$$

where E_{en} is the encircled energy and ω_0 is the beam waist radius in the CS_2 cell. We measured a maximum encircled energy of 0.6 mJ. This gives $F_{\max} = 149M^{-2}$ J/cm^2. Here we have $M = 20$, giving $F_{\max} \approx 0.4$ J/cm^2. Equation (2) can be used to estimate the peak fluence for other focal lengths of the lens L3. The minimum practical focal length of L3 is about 10 mm, which gives $M = 0.2$ and $F_{\max} \approx 4$ kJ/cm^2. In this case further attenuation of the beam is needed, and a possible solution would be a tandem setup, where CS_2 is used in the first limiter stage.[2]

No laser beam breakup was observed in an air-filled cell, proving that the filament pattern was indeed due to self-focusing, because air has a nonlinear coefficient, which is a factor $\sim 10^5$ lower that CS_2.[11]

4.3 Measuring the encircled energy

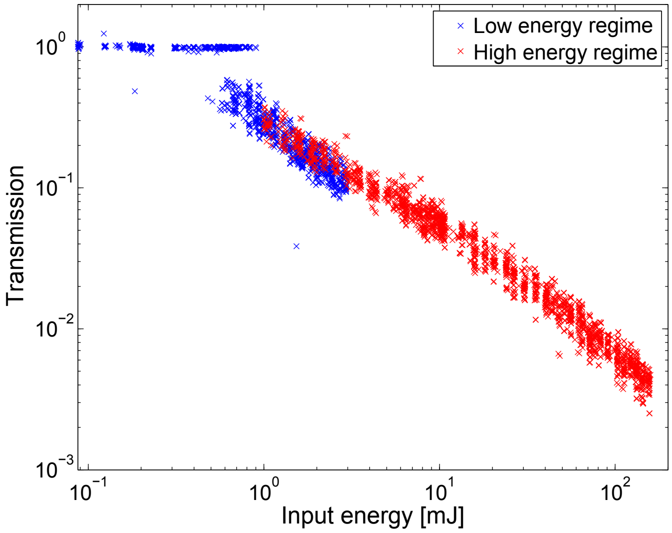

Figure 5. Measured transmission through the limiting aperture as a function of input energy on the CS_2 cell. We have corrected for losses in the uncoated cell windows.

Figure 5 shows measured transmission through the CS_2 cell and the limiting aperture as a function of pulse energy incident on the CS_2 cell, where we have corrected for losses in the uncoated cell windows. The figure shows results from measurements in both low energy and high energy regime. We observe from the low energy measurements that transmission starts to decrease for an incident pulse energy of ∼0.8 mJ. As noted in Sec. 4.1, the decrease in transmission was caused by dielectric breakdown, as in Fig. 3(a). We observed that there was an input energy range where only some of the pulses led to dielectric breakdown. This may be due to the fact that the laser operates on multiple longitudinal modes, such that the peak power varied from pulse to pulse, even though the pulse energy was the same. Figure 5 shows that the transmission is approximately inversely proportional to the input pulse energy above the threshold for dielectric breakdown. This is equivalent to a constant transmitted pulse energy at high input energies. An analysis of the raw data showed that the transmitted energy through the limiting aperture never exceeded 0.6 mJ, even though the input energy was as high as 150 mJ. The constant transmitted pulse energies at high input energies is supported by Fig. 4, which shows that the energy of the central part of the beam is approximately constant when the input energy increases.

Based on Figs. 3–5, we obtain the following picture of the optical limiting mechanisms: Consider a high-energy pulse incident on the CS_2 cell. The start of the pulse, where the power is less than the limit for dielectric breakdown and self-focusing, will be transmitted linearly through the cell, with little attenuation. At some point in the pulse, the power will exceed the threshold for dielectric breakdown, which will generate a plasma that attenuates later parts of the pulse. At still later times, self-focusing will move the focus position towards the input window. At even later times, the beam will break up into several filaments. As shown in Fig. 4, the filaments will spread the pulse energy over a larger area. The total effect is that only the first part of the pulse is transmitted through the limiting aperture AP, regardless of input pulse energy.

5. CONCLUSION

Carbon disulfide (CS_2) has been tested as an optical limiting material at 2.05 μm wavelength. Dielectric breakdown and laser beam breakup occurred in the focus in the CS_2 cell at high pulse energies, which lead to a strong reduction of the energy passing through a subsequent aperture. Pulse energies of up to 150 mJ were incident on the cell, while at most 0.6 mJ was transmitted through the aperture. If further attenuation of the beam is needed, a possible solution is a tandem setup, where CS_2 is used in the first limiter stage.

REFERENCES

1. Wood, R. M., [*Laser-induced damage of optical materials*], Institute of Physics, Bristol, UK (2003).
2. Hernández, F. E., Yang, S., Van Stryland, E. W., and Hagan, D. J., "High-dynamic-range cascaded-focus optical limiter," *Optics Letters* **25**, 1180–1182 (Aug. 2000).
3. Soileau, M. J., Williams, W. E., and Van Stryland, E. W., "Optical power limiter with picosecond response time," *IEEE Journal of Quantum Electronics* **QE-19**, 731–735 (Apr. 1983).
4. Tutt, L. W. and Boggess, T. F., "A review of optical limiting mechanisms and devices using organics, fullerenes, semiconductors and other materials," *Prog. Quant. Electr.* **17**, 299–338 (1993).
5. Ho, P. P. and Alfano, R. R., "Optical Kerr effect in liquids," *Physical Review A* **20**, 2170–2187 (1979).
6. Liu, Z.-B., Liu, Y.-L., Zhang, B., Zhou, W.-Y., Tian, J.-G., Zang, W.-P., and Zhang, C.-P., "Nonlinear absorption and optical limiting properties of carbon disulfide in a short-wavelength region," *J. Opt. Soc. Am. B* **24**, 1101–1104 (May 2007).
7. Mohebi, M., Aiello, P. F., Reali, G., Soileau, M. J., and Van Stryland, E. W., "Self-focusing in CS_2 at 10.6 μm," *Opt. Lett.* **10**, 396–398 (Aug 1985).
8. Plyler, E. K. and Humphreys, C. J., "Infrared absorption spectrum of carbon disulfide," *Journal of Research of the National Bureau of Standards* **39**, 59–65 (1947).
9. Sheik-Bahae, M., Said, A. A., Wei, T. H., Hagan, D. J., and Van Stryland, E. W., "Sensitive measurement of optical nonlinearities using a single beam," *IEEE Journal of Quantum Electronics* **26**(4), 760–769 (1990).
10. Ganeev, R. A., Ryasnyansky, A. I., Baba, M., Suzuki, M., Ishizawa, N., Turu, M., Sakakibara, S., and Kuroda, H., "Nonlinear refraction in CS_2," *Appl. Phys. B* **78**, 433–438 (2004).
11. Nikogosyan, D. N., [*Properties of optical and laser-related materials*], Wiley, West Sussex, England (1997).
12. Fonnum, H., Lippert, E., and Haakestad, M. W., "550 mJ Q-switched cryogenic Ho:YLF oscillator pumped with a 100 W Tm:fiber laser.," *Optics Letters* **38**, 1884–6 (June 2013).
13. Uteza, O., Bussière, B., Canova, F., Chambaret, J.-P., Delaporte, P., Itina, T., and Sentis, M., "Laser-induced damage threshold of sapphire in nanosecond, picosecond and femtosecond regimes," *Applied Surface Science* **254**, 799–803 (2007).
14. Couairon, A. and Mysyrowicz, A., "Femtosecond filamentation in transparent media," *Physics Reports* **441**, 47–189 (2007).
15. Boyd, R. W., [*Nonlinear Optics*], Academic Press, San Diego (2003).
16. Marburger, J. H., "Self-focusing: Theory," *Prog. Quant. Electr.* **4**, 35–110 (1975).