100X Enhancement of the Nonlinear Refractive Index of Sulfur-Doped CS_2 over Pure CS_2

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Abstract
Preliminary Z-scan measurements of variable concentration sulfur-doped CS_2 indicate a two-order of magnitude enhancement of the nonlinear index (n_2) over CS_2. The laser repetition rate will be varied to determine any thermal contribution to n_2.

Introduction
The search for new materials with greater third-order nonlinear susceptibility for high-speed optical switching and limiting applications has become a driving force within the field of nonlinear optics. Carbon disulfide (CS_2) is mostly used as a standard reference nonlinear material due to its high nonlinear index of refraction caused predominantly by the reorientational Kerr effect [1]. Carbon disulfide has also been known as an excellent solvent for crystalline forms of sulfur. These eight-membered rings of sulfur atoms are quite soluble in CS_2 with the solubility increasing logarithmically with temperature [2]. The motivation behind this study is to investigate the enhanced nonlinear refraction of such "sulfur-rich" CS_2 molecules which may serve as a potential candidate for optical switching and sensor applications.

Experimental Procedure and Results
We employed the Z-scan technique, which has become a standard method for measuring the nonlinear optical properties of materials; particularly nonlinear refraction and nonlinear absorption with high sensitivity [1]. Our laser source is a Time-Bandwidth 76-MHz, 1064-nm, 10-ps Nd:Vanadate system which is modelocked using a Semiconductor Saturable Absorber Mirror (SESAM) technology. The sulfur-doped CS_2 was prepared by dissolving a known mass of pure sulfur in 20ml of CS_2. A 3-mm thick fused silica glass cell was filled with pure CS_2 and a similar cell contained the doped CS_2. In order to make the interpretation of our results easier, both samples were simultaneously studied under the same conditions. We measured the n_2 for pure CS_2 to be $(1.3 \pm 0.2)x10^{-11}$ esu while the accepted value is $(1.3 \pm 0.3)x10^{-11}$ esu [1]. The experiment was repeated with increasing concentrations of sulfur. Figure 1 shows a Z-scan plot and the fit for sulfur-doped CS_2 with 1.05 % wt of sulfur (i.e. 0.2681g of sulfur dissolved in 20ml of CS_2) for which $n_2 = (1.2 \pm 0.2)x10^{-9}$ esu, which represents two orders of magnitude enhancement over pure CS_2 ($n_2 = (1.3 \pm 0.3)x10^{-11}$ esu) [1] .The next stage of this work will involve a detailed study of the n_2 dependence on the sulfur concentration, particularly the determination of the point at which the nonlinearity peaks with concentration. Similarly, studies are in progress to measure the variation in the linear and nonlinear absorption of doped CS_2 as a function of sulfur concentration – this may require frequency doubling to 532-nm. We will also measure the expected changes in switching times as a function of sulfur concentration utilizing a polarization spectroscopy technique based on optical Kerr gating [3]. Any possible thermal contribution to the measured nonlinearities will also be investigated by using a Pockels cell

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based pulse picker with which the repetition rate of the laser will be varied from 76MHz to a value as low as 7.6kHz.

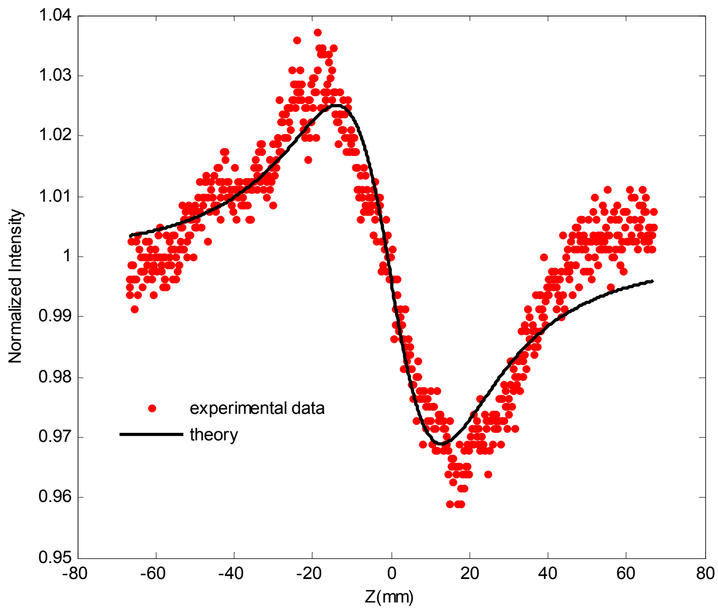

Fig. 1 Closed aperture Z-scan measurement of sulfur-doped CS2 with 1.05 % wt sulfur using a 76-MHz, 1064nm, and 10-ps laser

Conclusion

We measured n_2 for pure and sulfur-doped CS_2 under the same experimental conditions. Our values for pure CS_2 are in excellent agreement with the accepted values [1, 4]. On the other hand, the sulfur-doped CS_2 showed more than two orders of magnitude increase in n_2 when compared with pure CS_2. Although further studies are underway to fully understand the mechanism(s) responsible for these interesting results, we believe additional reorientational effects caused by the presence of the extra sulfur molecules may be the major contribution. Optical switching time studies, nonlinear refraction, linear and nonlinear absorption measurements will be carried out as a function of sulfur doping of CS_2.These results will be presented together with a quantitative assessment of any evidence of thermo-optical contributions to n_2.

References

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