Resolution of discrepancies in measured values of n_2 of CS_2 at 10 μm

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We explain the recent discrepancies between reported values of the nonlinear refractive index of CS_2 at 10 μm in terms of the relative importance of electrostriction and molecular reorientation under different experimental conditions.

The n_2 of CS_2 at 10 μm has been the subject of several experimental investigations, the results of which have differed by 3 orders of magnitude.[1-3] The recent study by Golub *et al.*[4] is another measurement at 10.6 μm. They report $n_2 = (2.1 \pm 0.7) \times 10^{-11}$ esu for CS_2, which is 10 times smaller than the results of Ref. 3 [$(2.2 \pm 0.7) \times 10^{-10}$ esu], 1000 times smaller than the results of Ref. 2 (3.5×10^{-8} esu), and in relatively good agreement with the results of Ref. 1 (1.1×10^{-11} esu) and other[5,6] measurements of n_2 performed in the visible and the near IR [$(1.5 \pm 0.3) \times 10^{-11}$ esu].

In all these studies the dominant nonlinearity is considered to be the reorientational Kerr effect, and the presence of electrostriction has been ignored. Shen[7] performed a comparative study of the Kerr and electrostrictive nonlinearities. The relative influence of these nonlinearities depends on the beam size, the pulse duration, and some material parameters. Under the approximations outlined in Ref. 7, Shen calculates the ratio of the index change due to electrostriction Δn_ρ to the index change due to the reorientational Kerr effect Δn_α to be

$$\frac{\Delta n_\rho}{\Delta n_\alpha} \simeq \frac{K_\rho}{K_\alpha}\frac{v^2}{{\omega_0}^2}{t_p}^2, \quad (1)$$

where the ratio of the electrostrictive coefficient K_ρ to the Kerr coefficient K_α is calculated to be 0.78 for CS_2,[7] and we have evaluated this ratio at the radial position $r = \omega_0$ (the spot radius) and at the time $t = t_p$ (the laser pulse width). The velocity of sound v in CS_2 is $\simeq 1.5 \times 10^5$ cm/sec.

The origin of the disagreement in the values of n_2 lies in the various experimental conditions used in the works mentioned above. In Ref. 3, long pulses (130 nsec) and small spot sizes (27 μm) were used. Under these conditions, electrostriction becomes the dominant nonlinearity, with the ratio in relation (1) being $\simeq 40$. For the experimental conditions of Ref. 4, where $\omega_0 = 1.5$ mm, this ratio is $\simeq 7 \times 10^{-6}$ for the 3-nsec pulses and 0.03 for the 200-nsec pulses. In the case of research done in the visible and near IR, 40-psec pulses and 100-μm spots were used.[5] Under these conditions, the ratio is $\simeq 10^{-7}$. These small ratios are obtained because the variations of the indices of refraction owing to electrostriction must propagate with the speed of sound a distance approximately equal to the beam size in order to contribute to self-focusing.

In Ref. 1, the rotation of polarization of a He–Ne laser beam propagating through the CS_2 cell by a CO_2 laser pulse was detected. Unlike the reorientational Kerr effect, electrostriction does not induce birefringence in CS_2, thus the experimental method is insensitive to electrostriction. This explains the good agreement between the results of Ref. 1 and Refs. 4–6.

The large value of n_2 reported in Ref. 3 is due to the presence of large electrostriction. Experimental evidence of the dominant presence of electrostriction in CS_2 with long pulses and small spot sizes is reported in Ref. 8. Under experimental conditions identical to those reported in Ref. 3, we have observed a 10% increase in the strength of the nonlinearity when the temperature of CS_2 was increased from 22 to 50°C. This is consistent with electrostriction, since the sound velocity increases with temperature. The reverse is expected from the Kerr effect, because thermal agitation makes it more difficult to align the molecules. Also, only a 6% increase in the threshold power for self-focusing was observed when the polarization of the CO_2 beam was changed from linear to circular. An increase of a factor of 2 is expected for the Kerr effect,[5] and no change is expected for electrostriction. These results at 10 μm confirm that electrostriction was the dominant nonlinearity in Ref. 3 and that the reorientational Kerr effect was dominant in Ref. 4. Since these conclusions show that little or no dispersion in the reorientational Kerr effect is observed from the visible to 10 μm, we conclude that the nearby vibrational resonances in the IR are not contributing to Δn_α, as was suggested in Ref. 3. However, linear ab-

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sorption in the IR may give rise to a small negative contribution to n_2 under experimental conditions in which electrostriction is dominant.[3,8] The extremely large value of n_2 reported in 1971 in Ref. 2 is most likely due to an erroneous assumption that the observation of nonlinear transmission indicated that the input power was near the critical power for self-focusing.

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