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Studies of the Nonlinear Switching Properties of Liquid Crystals with Picosecond Pulses

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A comparative study of self-focusing in seven liquid crystals using picosecond 0.53 and 1.06 μm pulses is presented. MEBBA was found to have the highest nonlinearity at 0.53 μm as determined in an optical power limiting experiment. This limiting appears to be due to nonlinear refraction enhanced by two-photon absorption.

We report here the results of a sytematic study of self-focusing in several liquid crystals in the isotropic phase using picosecond pulses at 0.53 μm and 1.06 μm. The samples studied and their chemical compositions are listed in figure 1.

Large nonlinearities have been previously measured in liquid crystals using nanosecond pulses[1] and such materials have been used in cw four-wave mixing experiments.[2] The large nonlinear refractive indices n_2 observed in the earlier work are believed to be due to light induced reorientation of the anisotropic liquid crystal molecules which is a relatively slow process. In order to estimate the value of n_2 in the liquid crystals, a power limiting configuration was used.[3] For highly nonlinear self-focusing materials such as CS_2, it was established in Ref. 3 that the limiting power obtained in such a configura-

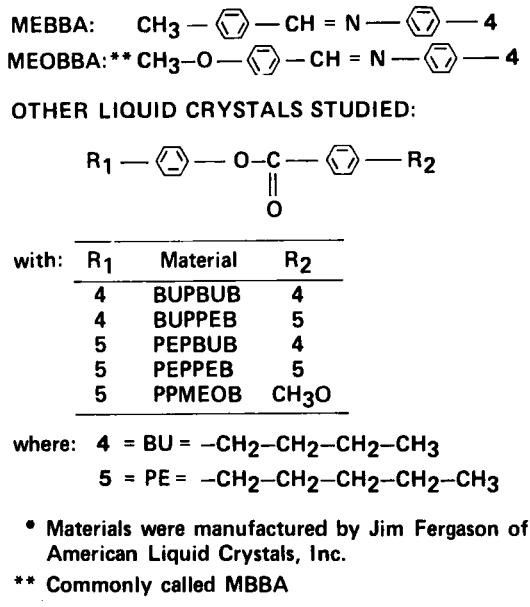

FIGURE 1 Chemical structures and nomenclature for the liquid crystals studied in this work.

tion is P_2, the second critical power for self-focusing. P_2 is related to n_2 by the relation.

$$P_2 = \frac{3.77c\lambda^2}{32\pi^2 n_2} \tag{1}$$

where c is the speed of light in vacuum, λ is the wavelength and 3.77 comes from a numerical solution to the nonlinear wave equation.[4]

The experimental technique is described in detail in Ref. 3 and is summarized here in Fig. 2. A lens (L_1) focuses light from a frequency-doubled mode-locked picosecond Nd : YAG laser operated in the TEM_{00} spatial mode. A second lens (L_2) reimages the transmitted light through an aperture onto the detector D_4 which measures the transmitted energy. As the incident power reaches P_2, the beam undergoes severe phase distortion and the reading on D_4 stops increasing for further increase of input power. This switching is due to self-lensing and subsequent optical breakdown. Figure 3 shows an example of such laser induced switching for CS_2 and MEBBA at 0.53 μm. The input field was linearly polarized. It is to be noted that the switching power for MEBBA is less than that of CS_2.

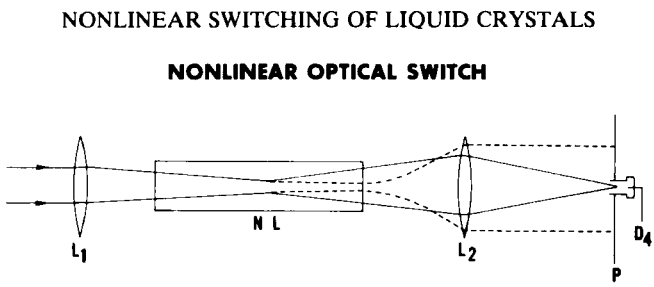

FIGURE 2 Technique for measuring the onset of self-focusing. The solid lines schematically trace the input beam for low input power. The beam is focused into the nonlinear medium by lens L_1 and then imaged by L_2 through an aperture onto detector D_4. The transmission for low input powers can be near unity. As the input power is increased to approximately P_2, the critical power for self-focusing,[3] the beam undergoes severe phase aberrations (i.e., nonlinear refraction) and the transmission through the aperture decreases. The high power situation is shown schematically by the dotted lines.

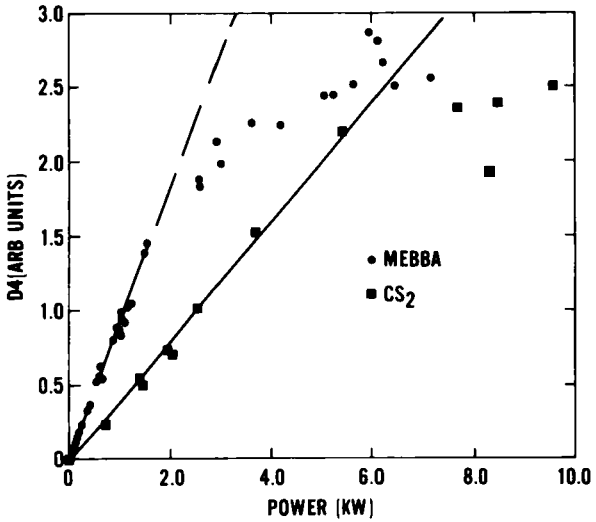

FIGURE 3 Nonlinear optical switching in CS_2 and MEBBA (4-methyl benzylidene 4′-n-butylaniline). The detector reading D_4 monitors the aperture transmission after the sample. D_4 remains unclamped with input power until P_2 is reached. The data shown are for 42 psec (FWHM), linearly polarized 0.53 μm pulses.

The ratio of the critical powers for the circularly polarized field to the linearly polarized field is approximately two for each of the samples studied, as shown in Figs. 4 and 5. Figure 4 shows the switching powers for the liquid crystals MEBBA, MEOBBA (commonly known as MBBA) and PPMEOB for linearly polarized input field and Fig. 5 shows the corresponding results for circularly polarized light. Figures 6 and 7 show the results for linear and circularly

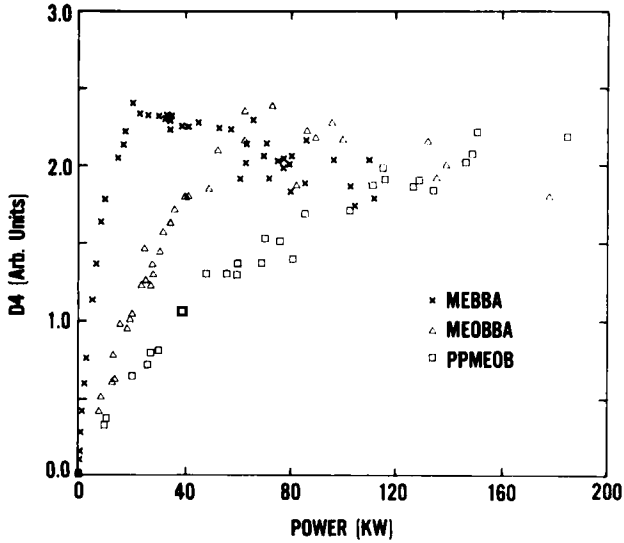

FIGURE 4 Comparison of nonlinear optical switching for the liquid crystals MEBBA, MEOBBA and PPMEOB. Linearly polarized light at 0.53 μm with 42 ps (FWHM) pulsewidth was used.

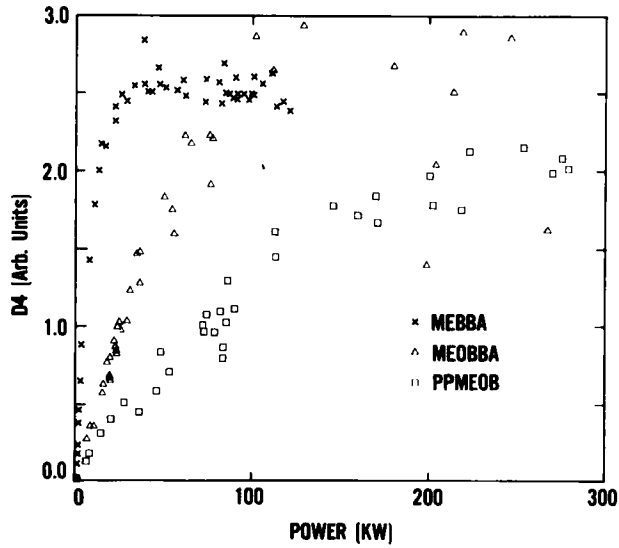

FIGURE 5 Comparison of nonlinear optical switching for the liquid crystals MEBBA, MEOBBA and PPMEOB. Circularly polarized light at 0.53 μm with 42 ps (FWHM) pulsewidth was used.

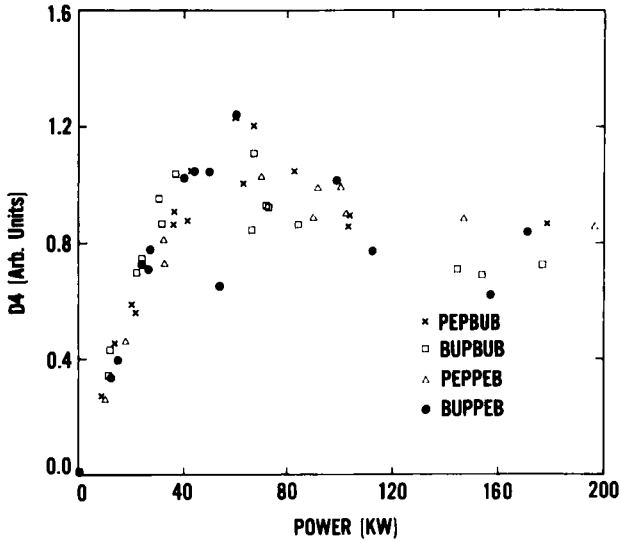

FIGURE 6 Comparison of nonlinear optical switching for the liquid crystals PEPBUB, BUPBUB, PEPPEB and BUPPEB. Linearly polarized light at 0.53 μm with 42 ps (FWHM) pulsewidth was used.

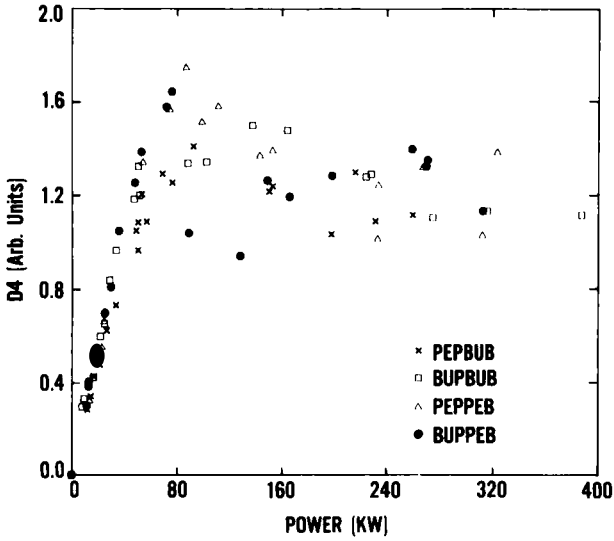

FIGURE 7 Comparison of nonlinear optical switching for the liquid crystals PEPBUB, BUPBUB, PEPPEB and BUPPEB. Circularly polarized light at 0.53 μm with 42 ps (FWHM) pulsewidth was used.

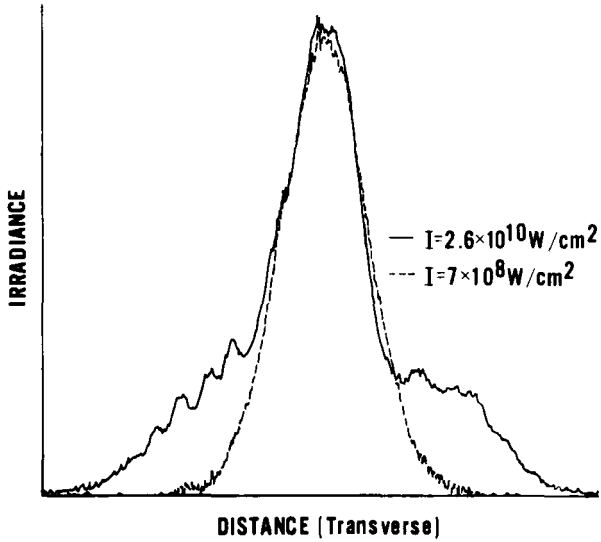

FIGURE 8 Vidicon scans showing the far field spatial profile of a 0.53 μm beam after transmission through an optically thin sample of MEBBA (thickness less than a Rayleigh range). The dashed curve is for low input irradiance and shows no beam distortion. The solid trace is for high irradiance and shows the effects of phase distortion in the sample.

polarized light, respectively, using the liquid crystals PEPBUB, BUPBUB, PEPPEB and BUPPEB. P_2 is seen to have the same value for for each of these four samples and for the sample PPMEOB. The fact that this ratio of switching powers for circular and linear polarizations is two is a clear indication that self-focusing is the dominant mechanism as discussed in Ref. 3. In fact, this is exactly the ratio observed for CS_2 where the dominant nonlinearity is molecular reorientation. Therefore, the observed polarization dependence suggests that the dominant nonlinearity, even for the short pulses, is molecular reorientation for these materials. However, for these large molecules the decay time constant would be expected to be considerably longer than the 42 psec (FWHM) pulses used.

Figure 8 shows the distortion in the far field spatial pattern of the beam after transmission through an optically thin sample of MEBBA (thickness of the sample less than the Rayleigh range). The effect of the optically induced phase distortion is observed by comparing the profiles at high input irradiance (solid curve) to that at low input irradiance (dashed curve). That this phase distortion arises from self-focusing rather than self-defocusing is confirmed by the results

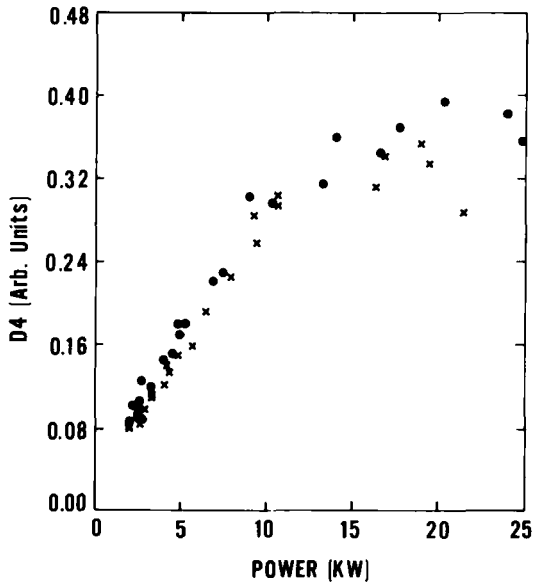

FIGURE 9 Comparison of nonlinear optical switching powers in MEBBA for different input irradiances. The focal length of the lens L_1 was 37.5 mm for the circles and 75 mm for the crosses so that the spot size in the sample changed by a factor of 2 (irradiance changes by 4 for a given power). Linearly polarized light at 0.53 μm with 42 ps (FWHM) was used.

shown in Fig. 9 in which the power dependence of the switching is shown for different input irradiances. The two curves shown correspond to experiments using lenses of focal lengths differing by a factor of two yielding spot sizes differing by a factor of two. If the limiting were due to self-defocusing, a dependence on the input irradiance would be expected, which is seen to be absent in Fig. 9. Also the fact that the switching power remains constant when the irradiance changes by a factor of four shows the dominance of nonlinear refraction as opposed to, for example, nonlinear absorption. An additional indication of self-focusing was the observation that at and above the critical input power there was optical breakdown in the liquid crystal media.

In Fig. 10 a comparison of critical powers in MEBBA at 1.06 μm and 0.53 μm is shown. The ratio of the values of P_2 at the two wavelengths is approximately 20 whereas self-focusing theory only predicts a ratio of 4 between the two wavelengths (see equation 1).[3,4] This large dispersion implies that the mechanism responsible for nonlinear refraction in MEBBA is not simply molecular reorientation.

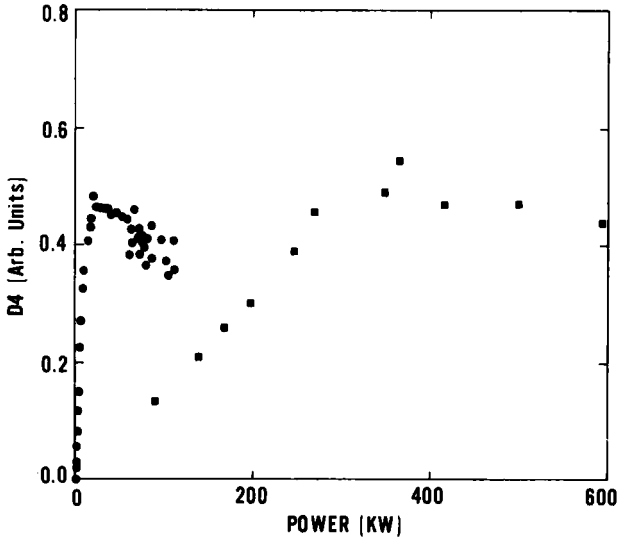

FIGURE 10 Comparison of nonlinear optical switching power in MEBBA between 0.53 μm and 1.06 μm radiation. The pulsewidths used were 42 ps and 67 ps (FWHM) respectively. Linearly polarized light was used.

Figure 11 shows optical switching in PEPBUB, BUPBUB, PEPPEB and BUPPEB at 1.06 μm. Comparing with Fig. 6 we find a ratio of 10 in the critical powers for this set of samples at the two wavelengths. The dispersion is still larger than the prediction of 4 from self-focusing theory, but smaller than that observed in MEBBA.

The larger nonlinearities at 0.53 μm may be due to an enhancement of the nonlinear refraction due to nonlinear absorption. Nonlinear absorption in MEBBA was observed at 0.53 μm and the results are plotted in Fig. 12 where the inverse transmission is displayed as a function of the incident irradiance. The nearly linear dependence on irradiance is consistent with a two-photon absorption process.[5] The dotted line shows the theoretical predictions for a two-photon absorption coefficient of 0.6 ± 0.2 cm/GW. If we now go back to the limiting configuration and calculate, using linear optics, the expected irradiance at focus we find a maximum change of transmission due to two-photon absorption of 15%; not enough by itself to account for the limiting. No nonlinear absorption was observed at 1.06 μm up to irradiances where there was obvious material breakdown. It is worth mentioning that excimer formation by nonlinear absorption of picosecond laser pulses has been observed in liquid benzene[6] and similar processes could play a role in liquid crystals as well.

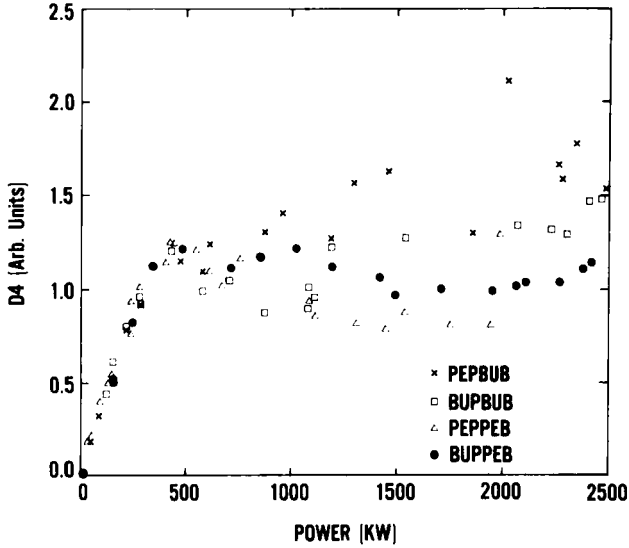

FIGURE 11 Comparison of nonlinear optical switching in PEPBUB, BUPBUB, PEPPEB and BUPPEB at 1.06 μm using linearly polarized light with 67 ps (FWHM) pulsewidth.

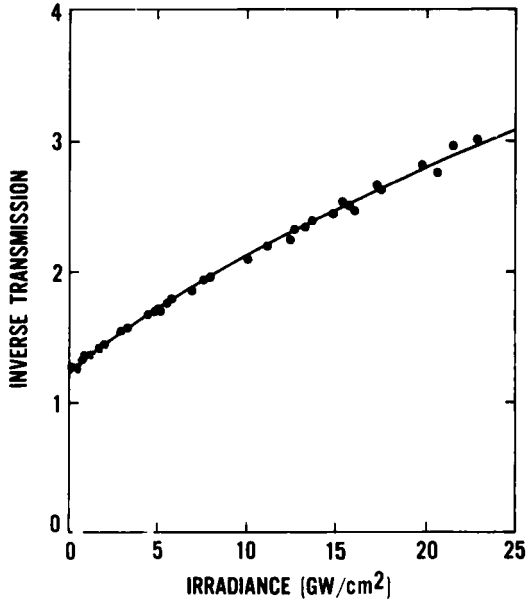

FIGURE 12 The inverse of the transmission of a 0.5 cm pathlength cell filled with MEBBA is plotted as a function of the incident irradiance at 0.53 μm. The solid line is a theoretical fit with a nonlinear absorption coefficient of 0.6 cm/GW.

To summarize, P_2 was measured for various liquid crystals and was found to vary greatly with minor changes in chemical composition. For example the difference between MEBBA and MEOBBA is the addition of a single oxygen atom. The liquid crystals were found to exhibit large and fast nonlinearities comparable to those of the Kerr liquids and are effective materials for optical power limiter applications. In addition, they have potential for applications in the areas of phase conjugation, optical-bistability and optical switching. Extensive studies of nonlinear hyperpolarizabilities of organic molecules have been performed with nanosecond pulses to determine the ways to maximize the nonlinear susceptibility by synthesizing molecules with different substituents.[7] Similar work using liquid crystals and determination of the origins of the nonlinearities would be of practical interest. Further study, including other materials, a determination of the pulsewidth and temperature dependence of the nonlinearities, and a more refined theoretical modeling is currently in progress.

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