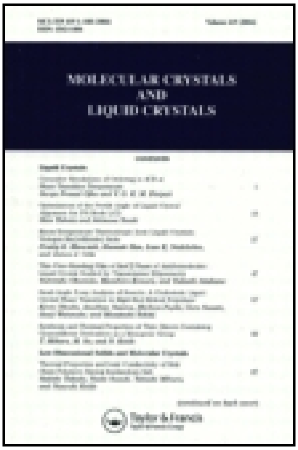

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Nonlinear optical Properties of Liquid Crystals In The Isotropic Phase

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NONLINEAR OPTICAL PROPERTIES OF LIQUID CRYSTALS IN THE ISOTROPIC PHASE

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Abstract Picosecond nonlinear absorption and nonlinear refraction were studied for several classes of isotropic phase liquid crystals, including Schiff base and ester compounds. Materials studied exhibit a large two-photon absorption coefficient (β) at 532 nm. Values of β were found to be ~0.6 cm/GW in several of the compounds studied. Nonlinear refraction was also observed and the nonlinear refractive index, n_2 was measured for each material at 1.06 μm using an external self-focusing arrangement. n_2 ranged from 6 x 10^{-13} esu to 2 x 10^{-12} esu. The combination of nonlinear absorption and nonlinear refraction in these materials result in optical limiting for input energies as low as 0.15 microjoules for 30 psec pulses at 532 nm.

INTRODUCTION

In this paper we report the results of simple direct measurements of nonlinear absorption and nonlinear refraction in selected liquid crystals in their isotropic phase. These measurements are part of an extensive study of nonlinear optical properties of materials in our laboratories.

I. C. Khoo's invited talk at this meeting[1] contained many excellent examples of the so-called giant optical nonlinearities in liquid crystal materials. These giant nonlinearities are associated with the anisotropy in the linear susceptibility of these organic molecules and the large scale ordering and changes in structural phase induced by an impressed optical field. These nonlinearities involve large scale motion of the relatively massive liquid crystal molecules and as a result the nonlinear response is quite slow (with response times as large as seconds). Liquid crystals have the delocalized π-electronic structures that have been identified by many workers as potential sources of fast and large optical nonlinearities.[2] In this work we use single pulses of picosecond duration to study such nonlinearities at 532 and 1064 nm. Thermal effects, electrostriction and long time constant reorientational effects are minimized in these measurements and the nonlinearities reported are an upper bound for the fast electronic effects.

EXPERIMENTAL

The laser source for these measurements was a Nd:YAG operated at 1064 nm. This laser system produced output pulses of 30 to 200 psec duration and operated in a single spatial mode (TEM_{00}) as verified by pinhole scans and whole beam analysis with an optical multichannel analyzer. The energy and pulsewidth was monitored for each laser shot using the procedure described in ref. 3. The laser output was harmonically converted to give output at 532 nm. The maximum repetition rate used was 5 Hz, thus insuring that the duty cycle was very low and that thermal effects were minimized.

The measurements were conducted using two very simple techniques which are described in detail elsewhere. In one case the sample thickness is much larger than the depth of focus of the beam (i.e., Rayleigh range) that is tightly focused into the bulk of the material being tested.[4,5] The nonlinear response is monitored by measuring the ratio of the input to the on-axis fluence in the far field of the beam after it exits the sample. This simple technique allows for the rapid comparison of the nonlinear response of many materials. In the case of simple nonlinearities, such as the electronic Kerr effect, the nonlinear refractive index (n_2) may be extracted from such measurements by monitoring the input power required for the onset of whole beam self-focusing. For more complex nonlinearities we simply determine the input power (P_C) required to limit the far field on-axis fluence. This parameter (P_C) is a rough measure of the combined effects of nonlinear refraction and nonlinear absorption and as such is a good indicator as to whether or not more quantitative measurements should be performed.

In the second technique the Rayleigh range of the beam incident on the sample is larger than the sample thickness and the beam is collimated as it traverses the sample. The two-photon absorption coefficient (β) is determined by measuring the total energy transmitted through the sample as a function of the input irradiance in the manner described in detail in refs. 6 and 7. The nonlinear refractive index (n_2) is determined by measuring the changes in beam shape and/or on-axis fluence in the near or far field of the test cell (see ref. 8 for details).

RESULTS

We have previously reported optical limiting in MBBA (p-methoxy benzylidene p-n-butylanilene).[9] Table I is a summary of P_C, β and n_2 measurements in this material and other liquid crystals of similar structure. All the data shown are for pulsewidths of approximately 30 ps and the liquid crystals were in the isotropic phase. Data for CS_2 are shown for comparison. The data for P_C were taken using the optically thick arrangement described in refs. 4 and 5. P_C is indicative of the total nonlinear response of the materials as previously noted. For simple Kerr nonlinearities P_C is inversely proportional to n_2. Note that P_C at 532 nm is about the same for all the samples tested including CS_2. The P_C (or limiting power) for CS_2 is associated with the onset of whole beam self-focusing whereas the observed limiting in the liquid crystal samples is associated with beam depletion due to two-photon absorption.

The nonlinear refractive index (n_2) was directly measured for these materials using the thin sample arrangement described in ref. 8. Note that in all cases the n_2 values for the liquid crystals are approximately an order of magnitude smaller than that of CS_2.

The relatively low values for the fast nonlinearity (assumed to be due to an optical Kerr effect) measured in these materials is somewhat disappointing considering reports of large, fast nonlinearities in organics reported by other workers. We are in the process of extending our measurements to other organics.

SUMMARY

The two-photon absorption coefficient (β) was measured for five isotropic phase liquid crystals at 532 nm. The values of β are large enough ($\simeq$0.6 cm/GW) for use in various nonlinear optical device applications. The nonlinear refractive index, n_2, was measured at 1064 nm for each sample and was found to be approximately an order of magnitude smaller than that of CS_2. Several other classes of organics were examined at 532 nm and all were found to have a smaller nonlinear response than CS_2.

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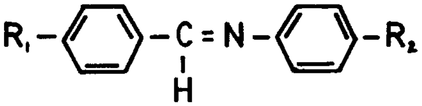
TABLE I

The molecular structure of the samples studied are of the form:



Sample	Color	R_1	R_2	P_C(kW) at 532 nm	(cm/GW) at 532 nm	$n_2 x 10^{13}$(esu) at 1064 nm
MEBBA	Orange	CH_3	C_4H_9	7±1	.55±.05	6±1
ETBBA	Light Yellow	C_2H_5	C_4H_9	8±1	.5 ±.05	17±2
IPBBA	Light Yellow	C_3H_7	C_4H_9	8±1	.5 ±.05	13±2
BBIPA	Orange	C_4H_9	C_3H_7	7±1	.5 ±.05	12±2
PEBBA	Orange	C_5H_{11}	C_4H_9	7±1	.55±.05	14±2
CS_2	Clear			8±1	Small	130