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Nonresonant third order susceptibility measurements in a nematic liquid crystal-PCH-5

John Philip and T. A. Prasada Rao
Laser Laboratory, Department of Physics, Indian Institute of Technology, Madras 600 036, India

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We report for the first time the determination of the real part of the third order nonlinear susceptibility $\chi^{(3)}(-\omega;\omega,0,0)$ above the nematic to isotropic phase transition temperature of a nematic liquid crystal 4 (trans-4′-4′-*n*-pentylcyclohexy)-benzonirtile (PCH-5) from electro-optic Kerr effect experiments. The value of $\chi^{(3)}$ observed at 632.8 nm is found to be 6.2079×10^{-19} m^2 V^{-2} close to the phase transition temperature at 55.1 °C.

INTRODUCTION

The potential application of materials with high $\chi^{(3)}$ values, in optical switching, computing, bistable elements, logic devices, etc., have attracted a number of researchers in the study of nonlinear optical properties of materials. Liquid crystals and polymers are receiving much attention because of their highest third order nonlinearities.

The measurements of $\chi^{(3)}$ have been made using various techniques such as electro-optic Kerr effect,[1–4] optical Kerr effect,[5] and degenerate four wave mixing.[6] Recently, $\chi^{(3)}$ measurements from optical phase conjugation by degenerate four wave mixing (DFWM) experiments in dilute solutions of polyphenyl acetylene (PPA) have been investigated by our group.[7] Highest nonlinearities have also been observed in many artificial Kerr media.[7–14] The largest nonresonant $\chi^{(3)}$ value known to be reported in the diacetylene systems, is of the order of 10^{-17} m^2 V^{-2}.[6]

Among the liquid crystalline mesophases—nematic, smectic, and cholesterics—nematic liquid crystals are well known for their extraordinary large optical nonlinearities. But very little information is available on the nonlinearities in the isotropic phase of the liquid crystals. Recently we have reported the static nonresonant third order nonlinear susceptibility $\chi^{(3)}(-\omega;\omega,0,0)$ measurements in two nematic liquid crystals MBBA and EBBA from our electro-optic Kerr effect studies.[15]

In this paper we report our measurements of the real part of the third order nonlinear susceptibility $\chi^{(3)}(-\omega;\omega,0,0)$ from electro-optic Kerr effect studies in a polar nematic liquid crystal PCH-5. The temperature dependence of $\chi^{(3)}(-\omega;\omega,0,0)$ values above nematic to isotropic transition temperatures (T_{NI}) have also been investigated in this nematic liquid crystal.

THEORY

The application of a static field to an optically isotropic liquid leads to anisotropy in the molecular distribution. This may be due to the intrinsic anisotropy of the molecules, and consequently becomes oriented by the field, or the applied field itself induces some anisotropy. If a beam of light of frequency ω_1 passes through a liquid subjected to electric fields, the refractive index for light polarized parallel to the electric field direction is different from the refractive index for light polarized perpendicular to the field direction. The phase difference (δ) between the parallel and perpendicular components of light after traversing the liquid is

$$\delta=2\pi l(n_1-n_2)/\lambda, \tag{1}$$

where l is the length of the Kerr cell, λ the wavelength of the probe beam, and (n_1-n_2) the change in refractive index given by

$$n_1-n_2=\lambda BE^2, \tag{2}$$

where B is the Kerr constant and E the electric field strength. The phase difference δ can be determined by the relation[16,17]

$$I=I_0\sin^2(\delta/2), \tag{3}$$

where I_0 is the maximum intensity of the probe beam. The dc Kerr constant is given by[18]

$$B=(24\pi/n\lambda_1)[\chi^{(3)}_{1212}(\omega_1,-\omega_1,0,0)+\chi^{(3)}_{1221}(\omega_1,-\omega_1,0,0)]. \tag{4}$$

For an isotropic medium the nonvanishing elements of third order nonlinear susceptibilities are the following:[19]

$$\chi^{(3)}_{1111},\chi^{(3)}_{1122},\chi^{(3)}_{1212}, \quad \text{and} \quad \chi^{(3)}_{1221},$$

where

$$\chi^{(3)}_{1111}=\chi^{(3)}_{1122}+\chi^{(3)}_{1212}+\chi^{(3)}_{1221}.$$

According to Klienman symmetry,[20] far from resonance,

$$\chi^{(3)}_{1122}=\chi^{(3)}_{1212}=\chi^{(3)}_{1221}. \tag{5}$$

Using expressions (4) and (5), $\chi^{(3)}(\omega_1,-\omega_1,0,0)$ values for the liquid crystals under investigations have been determined. Hereafter we use $\chi^{(3)}$ instead of writing $\chi^{(3)}(\omega_1,-\omega_1,0,0)$.

EXPERIMENTAL PROCEDURE

We have used a PCH-5 sample that has been supplied by E. Merck, Germany, in our experiments without further purification. The sample cell has been thoroughly cleaned before use and care has been taken to prevent absorption of moisture by sealing the sample cell. The nematic to isotropic transition temperature of this material has been studied from a polarizing microscope attached with a hot stage.

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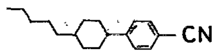

FIG. 1. Structure of PCH-5.

The experimental setup for our investigations has been described elsewhere.[3] A helium-neon laser of 5 mW power has been used as a probe beam. Spectrophotometer cuvette type Kerr cells of 2 cm path length have been constructed for our investigations. The electrodes are made up of stainless steel with 2 mm separation. Polytetrafluroethylene (PTFE) washers have been used to provide insulation between the electrodes. The Kerr cell is sealed during our experiments, in order to prevent evaporation. The inserting type electrode assembly makes the cleaning of the cell very easy and it avoids contamination of the liquid crystals. The temperature of the sample has been maintained within 0.1 °C using a precision thermostat controlled temperature bath. A photomultiplier tube (RCA IP28) is used as a detector. High voltage pulses up to 5 kV have been used to induce birefringence in the medium. The high voltage electric pulses that cause birefringence in the medium have been measured accurately using a Tektronix P6015 high voltage probe (1:1000). Both the Kerr signal and the high voltage pulses have been displayed on an Iwatsu oscilloscope (250 MHz) fitted with a polaroid camera.[21] The polarizer and the analyzer are adjusted such that they are crossed with each other, making an angle 45° with respect to the applied field. Glan–Taylor polarizer (EOD, U.K) with an extinction ratio 10^{-5} has been used as an analyzer. Schott filters have been used to avoid saturation upon the photomultiplier tube during the measurements. These measurements have been separately carried out for different temperatures in our experiment. The Kerr constant B has been calculated from the phase difference δ. The dependence of E^2 with δ has been found to be linear throughout our measurements.

RESULTS AND DISCUSSIONS

The nematic to isotropic transition temperature (T_{NI}) of PCH-5 is found to be 55.1 °C from our polarizing microscopic observations. The structure of this material, PCH-5, is shown in Fig. 1. The general properties of this liquid crystal is given in Table I.[22]

The refractive index of PCH-5 above nematic to isotropic transition temperature is measured at 632.8 nm using an Abbe refractometer, as these values are needed for $\chi^{(3)}$ calculations. Using the experimental values of the electro-optic Kerr constant B, the $\chi^{(3)}$ values above T_{NI} have been estimated for this liquid crystal. The highest value of $\chi^{(3)}$ observed near nematic to isotropic transition temperature, at 632.8 nm, is found to be 6.2079×10^{-19} $m^2\,V^{-2}$. This value is found to be approximately a hundred times greater than the $\chi^{(3)}$ value of CS_2 (7.1×10^{-21} $m^2\,V^{-2}$) (Ref. 5) and ten times greater than the value of MBBA (6.602×10^{-20} $m^2\,V^{-2}$).[15] The variation of $\chi^{(3)}$ with temperature, above T_{NI}, is shown in Fig. 2.

TABLE I. General properties of PCH-5.

1 Phase transitions	C–30 °C-N–55.1 °C-I
2 Melting point	+30 °C
3 Clearing point	+55.1 °C
4 Melting enthalpy	21.35 kJ mol^{-1}
5 Clearing enthalpy	0.96 kJ mol^{-1}
6 Molecular weight	255.4
7 Density, ρ/g cm^{-3}	0.9706 (at 20 °C)
8 Bulk viscosity, ν/mm^2 s^{-1}	22.5 (at 20 °C)
9 Rotational viscosity Pa s	0.1507 (at 20 °C)
10 Optical anisotropy Δn (at 633 nm)	0.1208 (at 20 °C)
11 Dielectric anisotropy $\Delta\epsilon$ (kHz)	12.7 (at 20 °C)

CONCLUSION

The observed value of the nonresonant third order nonlinear susceptibility in PCH-5 is found to be about two orders higher than that of CS_2 and is about one order higher than that of MBBA. These are well known for their highest nonlinearities. MBBA is known[22] for its sensitivity to water by exhibiting nematic to isotropic transition temperature anywhere between 35 and 45 °C. But this new material, PCH-5, is stable and exhibits sufficient stability in response to heat, radiation, and moisture. The nematic to isotropic transition temperature of this liquid crystal is also found to be stable.

The highest value of third order nonlinear susceptibility near nematic to isotropic transition temperature shows the intermolecular orientational correlations, although this phase is isotropic. Just above T_{NI}, long molecules of the

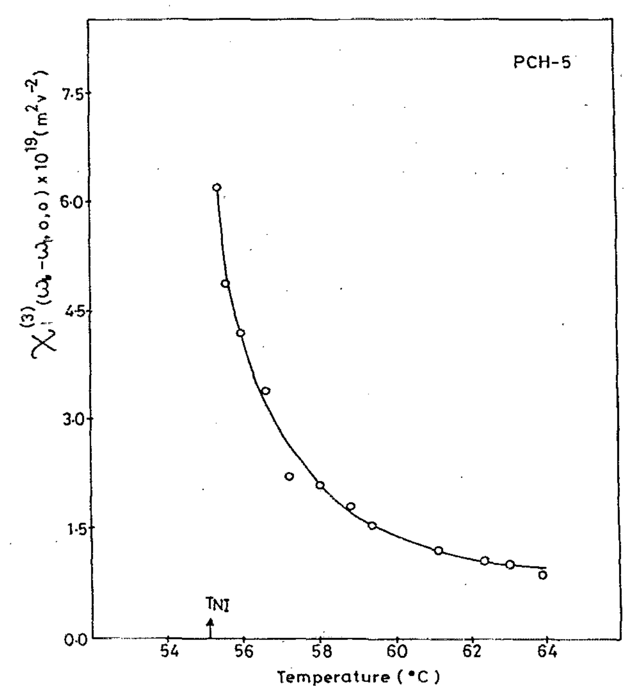

FIG. 2. The variation of $\chi^{(3)}$ with temperature for PCH-5.

liquid crystals will have parallel association to each other resulting in a very high third order nonlinearity. This molecular association increases as the temperature of the medium approaches the transition temperature. The variation of $\chi^{(3)}$ with temperature, above nematic to isotropic transition temperature, is direct evidence of strong fluctuations of the orientational order at these temperatures.

The delocalized π electronic structures of the liquid crystals make them potential sources of fast and large optical nonlinearities. This high third order nonlinearity exhibited by this liquid crystal shows great promise in nonlinear optical and electro-optical devices such as modulators, electro-optic tunable notch filter (EOTF), etc.

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