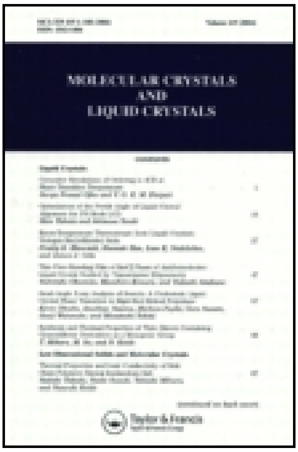

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Observations of Nonlinear Optical Effects in Metallorganic Liquid Crystals

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OBSERVATIONS OF NONLINEAR OPTICAL EFFECTS IN METALLORGANIC LIQUID CRYSTALS

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Abstract . We report the observation of light induced optical nonlinearities of thermal origin in metallorganic liquid crystals. Experimental data about self focusing and self phase modulation are presented and discussed. An estimation of the value of the nonlinear refractive index of the sample is performed.

Keywords: metallomesogens, refractive index

INTRODUCTION

Recently, a remarkable scientific interest has been excited by a new class of thermotropic liquid crystals obtained by complexation of mesogenic molecules with metals[1]. The characterization of these compounds is actually under way. In the literature structural studies have appeared which refer to techniques such as small angle x - ray scattering and x - ray diffraction[2]. Calorimetric, microscopic and conoscopic measurements have also been performed[3]. On the other hand, until now, observations of nonlinear optical effects on these materials have not been reported, while several of such effects have been observed and studied in usual nematic liquid crystals, exhibiting very interesting features[4-7].

In this paper we present the first observations of optical nonlinearities of thermal origin induced in metallorganic liquid crystals.

We have observed the self - focusing and self - phase modulation effects. The interesting feature in this case is represented by the extremely low impinging intensities which are needed to induce the effects. The origin of these phenomena is in the strong light absorption which gives rise to a local temperature increase of the sample. The consequent thermal indexing originates the observed nonlinear effects. The performed measurements enables us to give an estimation of the thermal gradient of the refractive indexes.

EXPERIMENTS AND RESULTS

The experimental setup is shown in Figure 1. It allows both for self - focusing and self - phase modulation experiments. The light from a HeNe laser (λ = 6328 Å, 25 mW fixed output power of Gaussian shape) is linearly polarized by a polarizer and focused by a lens which can be moved in the beam direction by a micrometer movement. A $\lambda/2$ plate before the polarizer can be rotated in order to regulate the power impinging on the sample while a calibrated power meter is systematically introduced after the polarizer for the measurement of this power. At a distance D = 63 cm after the sample, a photomultiplier with a 10 μm pinhole is moved transversally to the beam in order to detect its shape. A computer controls both this movement and the acquisition and elaboration of the signal. A light chopper can be introduced before the lens for the measurement of the response time of the system while a white screen substitutes the receiver when visualizing the typical self phase modulation rings pattern. The sample, 36 or 100 μm thick, has a planar texture and is placed in a thermostatic cell.The liquid crystal molecule, denoted by $A'PdA_2$, contains a planar rigid core - formed by a palladium atom with three aromatic rings - and two aliphatic chains which create an elongated molecular structure. Microscope observations indicate the following values for the melting and clearing temperatures: T_m = 90 °C, T_c = 105 °C.

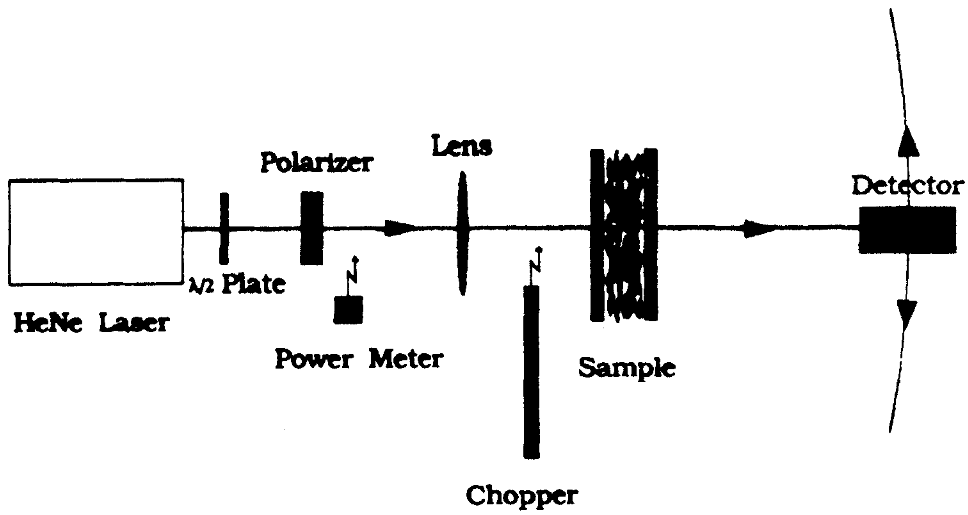

FIGURE 1. Sketch of the experimental setup

SELF - FOCUSING EXPERIMENTS

Measurements of self - focusing effect both for ordinary and extraordinary waves were performed with a focal length f = 200 mm. The samples were in the nematic phase (T = 97 °C) and placed 30 mm before the focal position. In this conditions, a focusing effect for the ordinary wave and a defocusing effect for the pure extraordinary wave have been observed. This effects have been measured by the difference $w - w_0$ between the beam width in D with and without the sample, that gives the inverse f^{-1} of the focal length of the nonlinear thin lens represented by the sample which causes the effect[5]. The measured values of f^{-1} vs the light intensity I are reported in Figure 2 for the ordinary wave in the 36 μm thick sample and in Figures 3 and 4 both for the ordinary and the extraordinary waves in the 100 μm thick sample.

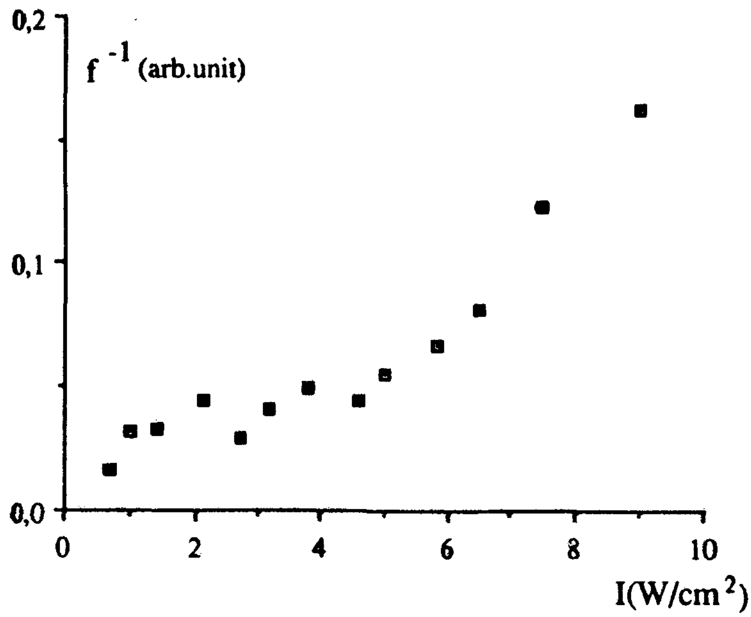

FIGURE 2. f^{-1} for the ordinary wave in the 36 μm thick sample

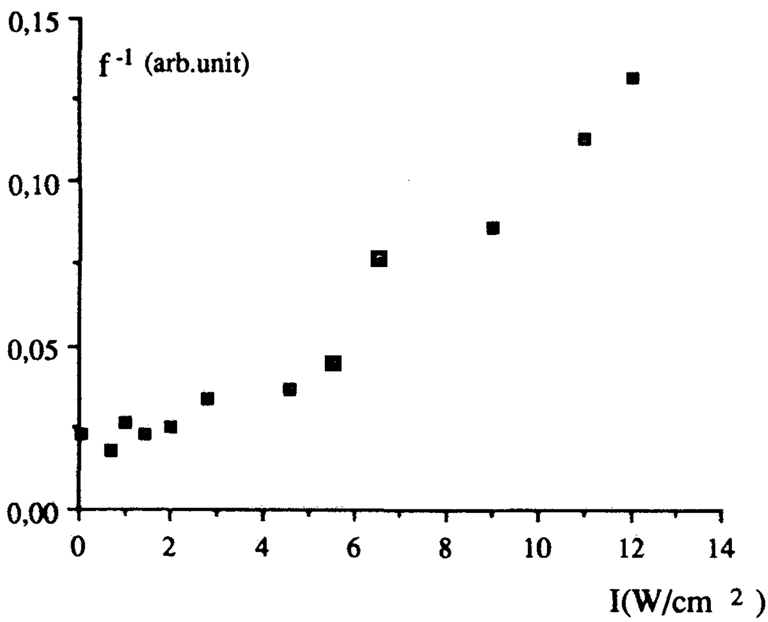

FIGURE 3. f^{-1} vs I for the ordinary wave in the 100 μm thick sample.

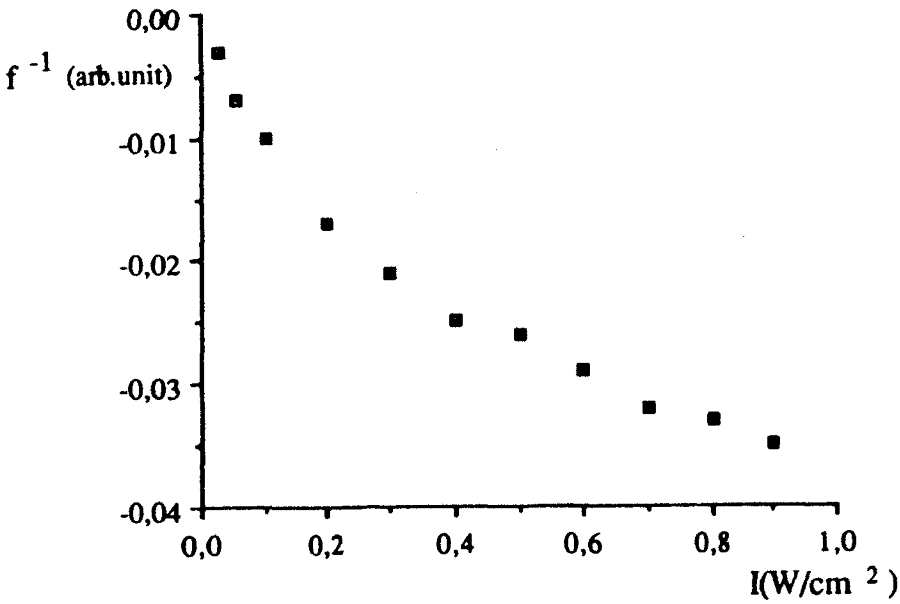

FIGURE 4. f^{-1} vs I for the extraordinary wave in the 100 μm thick sample.

SELF - PHASE MODULATION EFFECT

This effect has been observed in a 100 μm thick sample at 92 °C (nematic phase). The sample is in the focal position of a lens with a focal length f = 100 mm. The number N of rings vs I is reported in Figure 5 for the extraordinary wave.

Response time measurement

The characteristic response time τ of the effect has been measured for the ordinary wave by putting I just above the threshold value and chopping the beam. The output signal S of the detector, recorded vs time, is shown in Figure 6 and gives $\tau \approx 5$ msec.

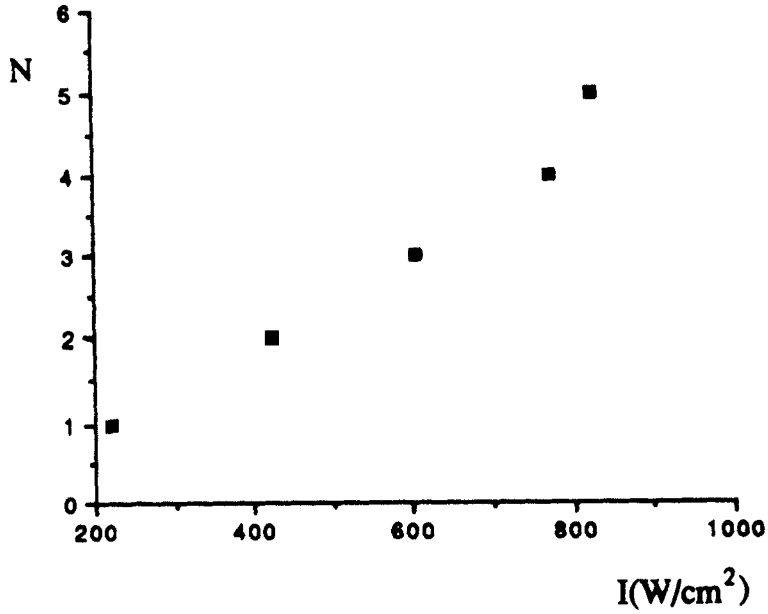

FIGURE 5. Number of rings N vs light intensity I for the extraordinary wave in the 100 μm thick sample

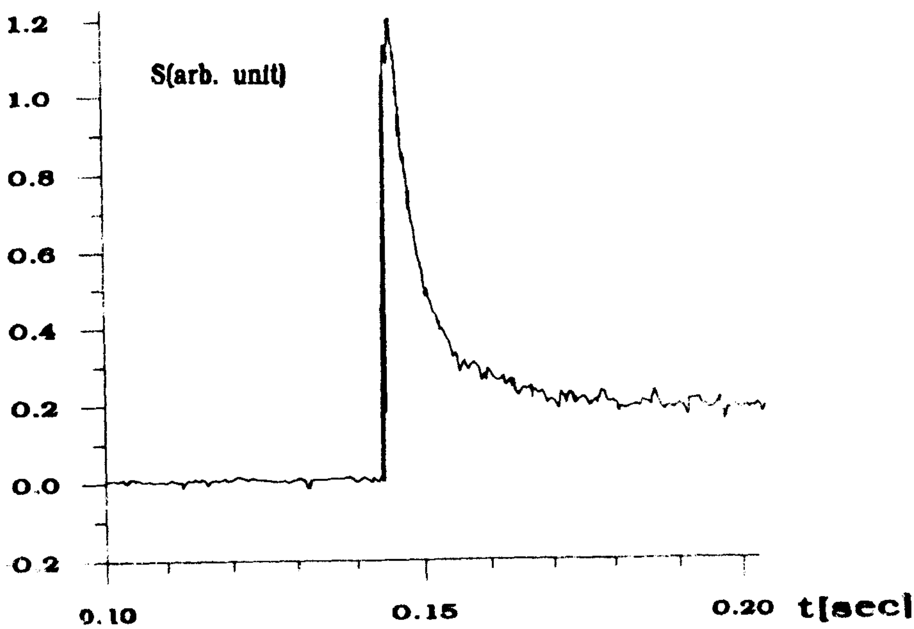

FIGURE 6. The signal S vs time when chopped light is used

DISCUSSION

The opposite sign of f^{-1} for the ordinary and the extraordinary beams and the typical value of τ show that the origin of the observed effects is in a thermal indexing phenomenon due to a strong light absorption. In fact this absorption produces a local temperature increase which can even lead the sample to the phase transition to the isotropic state. This transition, when occurs, is induced in a small droplet in correspondence of the center of the impinging gaussian beam, where the intensity has its maximum, and is demonstrated by the onset of the typical pattern of diffraction rings (quite different from the self - phase modulation ones and easily recognizable) due to light diffraction through the isotropic droplet.

This observation enabled us to evaluate the dependence on the light intensity I of the local thermal increase ΔT induced in the sample. Indeed, for a thermostatic cell temperature $T_0 = 97$ °C we have observed the onset of diffraction rings for an impinging intensity $I = 770$ W/cm^2. In this case we have exactly $\Delta T = T_c - T_0$. By writing for the local temperature increase a linear dependence on the light intensity

$$\Delta T = \alpha I \qquad (1)$$

we obtain $\alpha = 10.4 \times 10^{-3}$ °C cm^2/W

We have used the obtained value of α and the self - focusing and self - phase modulation measurements to estimate in the average the thermal gradient of the refractive indexes dn_o/dT and dn_e/dT in the temperature range of interest, both for the ordinary and the extraordinary beams. Where the self - focusing experiment is concerned, we write[5] the nonlinear refractive index as

$$< \delta n > = f^{-1} w^2/2d \qquad (2)$$

where w is half width of the gaussian beam on the sample and d is the sample thickness. Since we have supposed a thermal origin of the effect, we can write:

$$< \delta n > = (dn/dT)\, \Delta T \qquad (3)$$

Recalling equations (1) and (2) we obtain:

$$f^{-1}/I = (2d\alpha/w^2)(dn/dT) \quad (4)$$

By deducing f^{-1}/I from the experimental data, we find

$$|dn_o/dT| \sim 2 \times 10^{-2}; \qquad |dn_e/dT| \sim 6 \times 10^{-2}$$

A rough estimation of the same physical quantities made by using the self - phase modulation measurements confirm the order of magnitude of these values.

In conclusion we report the first observations of self - focusing and self - phase modulation in metallorganic liquid crystals. The interest of these observations is in the extremely low values of the requested optical intensities in comparison with the values that are needed for usual nematic liquid crystals.

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