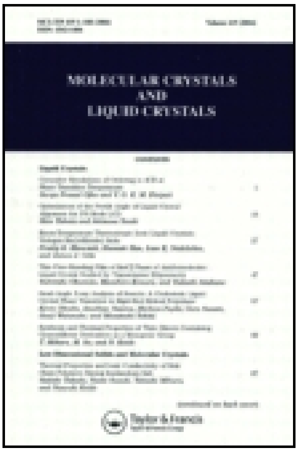

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Nonlinear Diffraction Driven by Low Frequency Electric Field in Polymer Dispersed Liquid Crystals

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NONLINEAR DIFFRACTION DRIVEN BY LOW FREQUENCY ELECTRIC FIELD IN POLYMER DISPERSED LIQUID CRYSTALS

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Abstract We show that in polymer dispersed liquid crystals it is possible to use a bias voltage to control the nonlinear optical interaction of two coherent beams. The orientational and thermal properties of the liquid crystal are coupled in such a way that the former ones drive the thermal nonlinearities. The main features of the observed effect are reported.

Keywords: non-linear, optics, PDLC, field effects

THE CONTROLLED OPTICAL EFFECT

Diffraction due to gratings induced by wave mixing processes in nonlinear materials has been extensively studied in the past few years[1]. This phenomenon has been used to investigate either fundamental molecular properties of the materials or more applied effects such as optical phase conjugation, probe beam amplification and infrared to visible image conversion[2]. In a recent letter[3] we have reported the observation of nonlinear light diffraction with threshold in dye-doped polymer dispersed liquid crystals (PDLC) caused by a combination of the self transparency and the light induced thermal grating effects. In this paper we demonstrate that it is possible to exploit the unique features of PDLC to couple thermal nonlinearities of the liquid crystalline medium with its orientational properties: the nonlinear diffraction in PDLC can be driven by a bias voltage applied to the sample, provided that some conditions are fulfilled.

When two laser beams of the same frequency cross at a small angle in a liquid crystal they can interact through the nonlinear polarization of the medium and thus give rise to new coherent waves of the same wavelength and traveling in different directions, through a four wave mixing process[4]. The new waves can be considered the result of diffraction of the original two beams due to an index grating produced by their interference in the nonlinear medium. In fact, due to the interference of the two beams, the light intensity on the sample, can be written as

$$I = I_o\,[1 + m\cos(2\pi\, x / \Lambda)]$$

where $I_o = I_1 + I_2$, is the sum of the intensities of the two beams, x the transversal coordinate, $m = 2\sqrt{I_1 I_2}/I_o$ is the fringes' modulation and $\Lambda = \frac{\lambda}{n \sin\theta}$ is the grating costant for a crossing angle θ between the waves of wavelength λ/n, where n is the refractive index of the medium. Heat absorption may produce a temperature rise and, as a consequence, an index modulation in the medium. This is the so-called thermal grating studied in liquid crystals[5]. In order to give a self-consistent description of this effect the heat diffusion equation must be supplemented by the wave equation which includes the nonlinear polarization of the medium. Several diffracted beams may arise due to this wave mixing process.

This phenomenon presents a peculiar feature in PDLC: there is a threshold I_{th} for the total light intensity impinging on the sample for the onset of nonlinear diffraction. In fact, since a PDLC sample is usually a strong scatterer, it doesn't allow light transmission until the impinging intensity is above a certain value which produces the self transparency[6]. Therefore only after the onset of self-transparency a thermal grating can be formed in the sample.

The value I_{th} must be obviously dependent on the degree of scattering of the material, since it determines the penetration depth of the radiation into the sample and the consequent efficiency of heating. Even if very low scattering (i.e. high transmission) may be not the best condition for maximum light absorption, nevertheless a lowering of the scattering which allows a deep penetration of the light into the sample will certainly increase the efficiency of heating.

For this reason a suitable low frequency voltage applied to the sample boundaries will be appropriate to control the threshold for the appearance of the nonlinear optical effect, as it can switch a sample from a high scattering state (opaque state) to a low scattering state (transparent state). Then, in order to drive the nonlinear diffraction by an applied voltage, we must fix the impinging intensity below the threshold for the self induced effect ($I_o < I_{th}$) when the voltage is switched off, but high enough to be above threshold when the voltage is switched on.

EXPERIMENT AND RESULTS

The experimental set-up is shown in fig. 1. It is the usual one to study degenerate four wave mixing in liquid crystals. An argon ion laser beam ($\lambda = 514.5$ nm) was divided by a beam splitter into two beams, with intensity

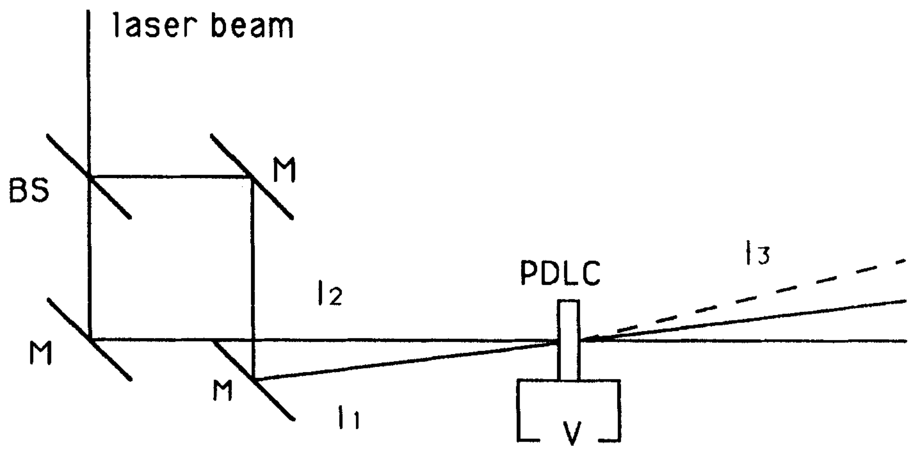

FIGURE 1 Experimental set-up

respectively I_1 and I_2. In order to ensure coherent interaction between them, their optical paths were made approximately equal before they were recombined at a small angle ($\sim$ 0.5 deg) on the PDLC sample with beams' diameter of about 2mm.

In our experiment we had $I_1 / I_2 = 1.3$, due to uneven reflections on the different surfaces.

The sample was obtained using the polymer-induced phase separation method. It was prepared starting from an homogeneous solution containing: (i) E7 liquid crystal from BDH, (ii) the epoxid fluid prepolymer EPON 815 from Shell Chemical and MK107 from Wilmington Chemical, (iii) the polyamminic curing agent Capcure 3-800 and component B from epoxid glue from BOSTIK, (iv) the orange dye D2 from BDH (0.02% in E7). The mixture was sandwiched between two conducting glasses (with ITO coatings) spaced 36 μm apart by appropriate Mylar spacers and was allowed to cure for several days before the measurements were performed.

The laser power was regulated in order to have $I_1 + I_2 < I_{th}$. At this power level very weak transmission of the two impinging beams was observed.

A low frequency (10 Hz) square wave voltage V was applied and observations were made increasing the V value. Up to $V_{th} \simeq 13V$ only a slight increase of transmission of the two original beams was observed, but for $V > V_{th}$ a clear diffracted beam was observed and a further increase gave rise to a fourth beam diffracted on the other side. This observation is well explained by the wavevector geometry of our experiment where $I_1 > I_2$. As shown in fig. 2 the first diffracted beam to appear will be the one with wavevector $\mathbf{k}_3 = 2\mathbf{k}_1 - \mathbf{k}_2 - \Delta\mathbf{k}$ $2\mathbf{k}_1 - \mathbf{k}_2$ ($\Delta\mathbf{k} \simeq 0$ as θ is small) therefore it will be close to the brighter beam; a weaker diffracted beam with wavevector $\mathbf{k}_4 \simeq 2\mathbf{k}_2 - \mathbf{k}_1$ will appear on the other side close to the beam whose wavevector is $\mathbf{k}_2$.

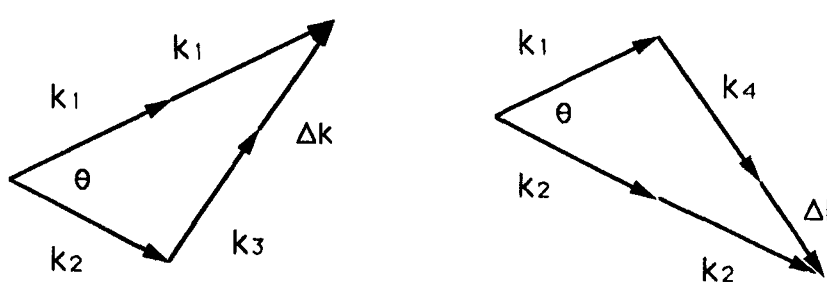

FIGURE 2 Wavevectors' diagram

Measurements of the intensity I_3 of the first diffracted beam were taken vs P_L and vs V, where P_L is the c.w. laser power.

The measurement of I_3 vs P_L allowed us to chek the nonlinear character of the signal which disappeared by blocking either I_1 or I_2, as expected.

The diffracted signal due to a third order effect may be expressed as

$$I_3 = C\, I_2^2\, I_1$$

where C is a constant which includes parameters depending on the material and on the experimental conditions. In particular it includes the square of the refractive index gradient dn/dT which gives the strength of the nonlinear response.

Since $I_1 = \alpha P_L$ and $I_2 = \beta P_L$, where $\alpha < 1$ and $\beta < 1 - \alpha$ are constant quantities determined by the several reflections on mirror and beam splitters, it is straightforward to get

$$I_3 = \text{const.}\ P_L^3$$

The experimental data show a very good agreement with this cubic law. In fig. 3 we report I_3 vs P_L^3 obtained with V = 30 Volt at 10 Hz.

The excellent linear dependence confirms the behaviour expected for a beam diffracted by a thermal grating. We must notice that all the used values of P_L were far below the value necessary to reach the threshold I_{th} (correspondent to 300 mW). At a fixed laser power (P_L = 100 mW) the signal I_3 was recorded varying the applied voltage V at 10 Hz.

A threshold value $V_{th} \simeq$ 13V for the appearance of the nonlinear diffraction was found. Anyway the most interesting observation is the temporal behavior of I_3 which shows, at higher voltages, the growth of an initial fast peak followed by a flat signal which lasts until the voltage is switched off. Moreover they have a different behavior as the voltage is increased, while both components follow a cubic law as function of P_L.

In fig. 4 we report the dependence of the two time components of the signal I_3 vs the applied voltage. It can be seen that the flat component reaches very soon a saturation value, while the peak keeps increasing approaching a saturation which is not reached in our measurements.

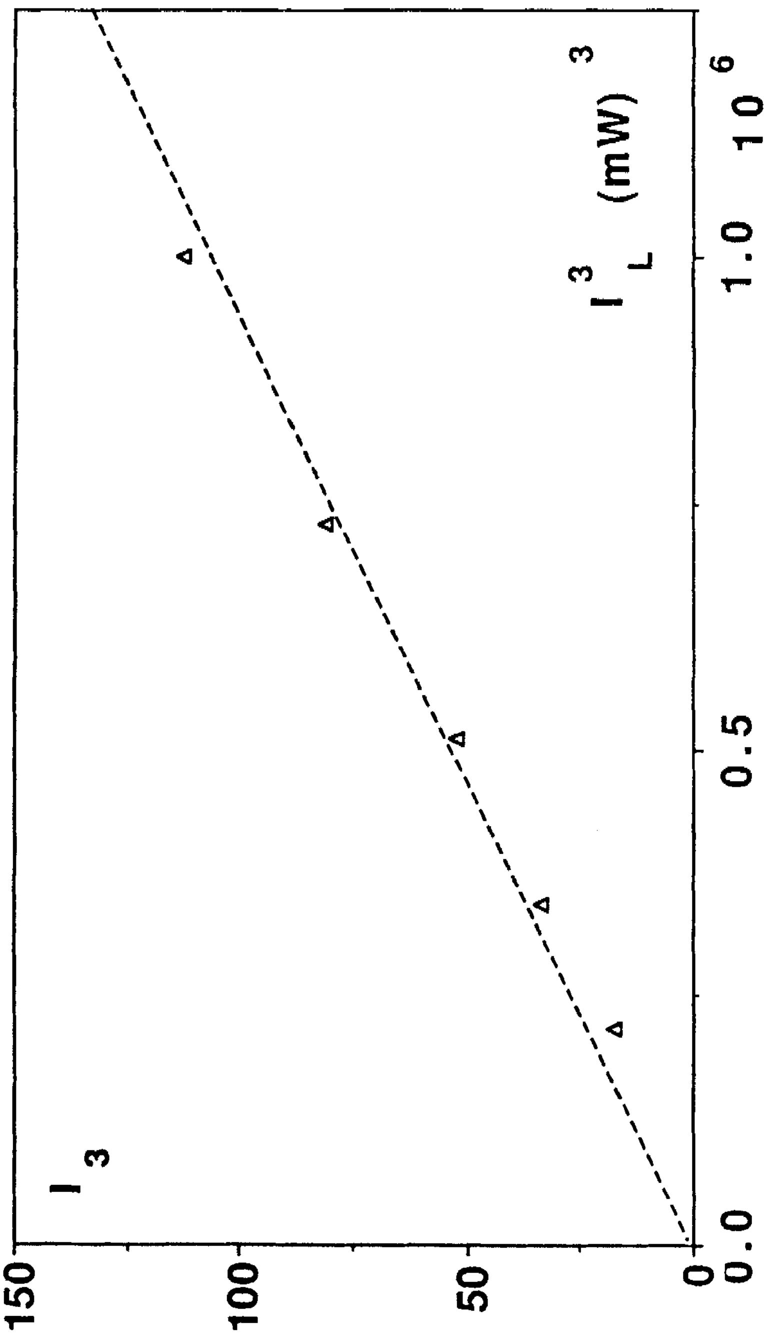

FIGURE 3 The intensity I_3 of the first diffracted beam vs the laser power P_L with an applied square wave voltage V=30 Volt at 10Hz

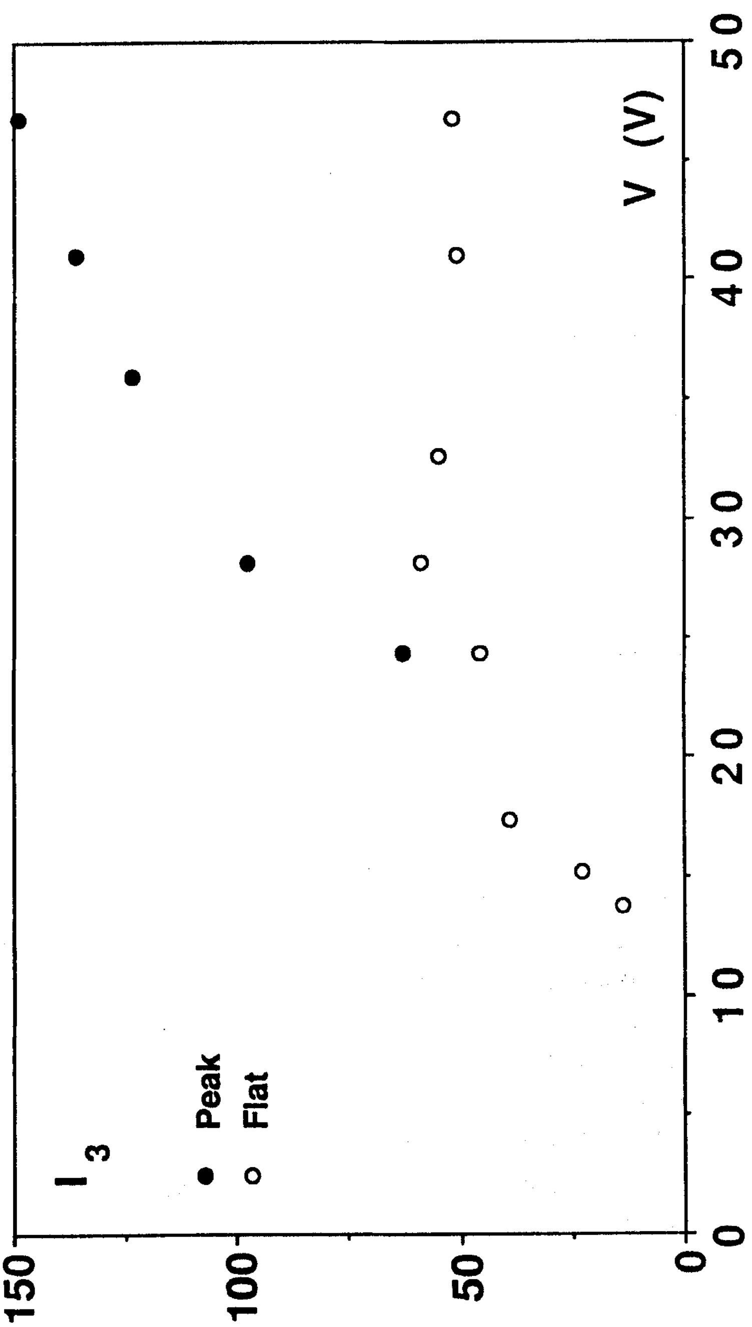

FIGURE 4 The intensity I_3 of the first diffracted beam vs the applied voltage. Laser power $P_L = 100$ mW

The risetime of the peak is about 0.5 msec. while it decays in about 2 msec. The decay time of I_3 as the voltage is switched off is quite long (~ 25 msec.) and correspond to the switching off time of the PDLC transmission under a bias voltage.

The observed time behavior of I_3 and the voltage dependence show that a rather complex nonlinear optical phenomenon takes place.

It is not the aim of this report to investigate the different processes which may affect the observed nonlinear diffraction, anyway it appears that a detailed analysis of them can give more insight into physical mechanisms governing the liquid crystal droplets in a polymeric matrix. In fact the behavior of the nonlinear signal under a square wave voltage must be linked to the correspondent behavior of the beams which cause the process: the switch on and switch off times are limited by the ones of the fundamental beams. Nevertheless the peculiar time behavior of I_3 is not evident in I_1 and I_2 and may be produced by two different mechanisms leading to a nonlinear response namely thermal indexing in liquid crystalline droplets and in isotropic droplets.

Anyway other experimental conditions must be studied in order to give satisfactory answers to the questions arising from this observation, in fact the voltage dependence on either of the two components of I_3 cannot be inferred directly from the one which has been measured for I_1 and I_2, i.e. from the transmission characteristic of a PDLC under a low frequency electric field.

Leaving these problems to future investigations which are already in progress, we must stress the noticeable feature of a nonlinear response of PDLC with a risetime of 0.5 ms, which may be considered interesting in applications such as controlled beam deflection.

In conclusion, we have shown that it is possible to exploit the unique features of a PDLC sample to drive a four wave mixing process using a low frequency voltage.

By switching the transmission of a PDLC sample it is possible to activate the thermal nonlinear response of this material. The effect appears to be an original coupling of thermal and orientational properties of liquid crystals.

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