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TRANSIENT AMPLITUDE GRATING IN POLYMER DISPERSED LIQUID CRYSTALS

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Abstract. We report the time behavior of nonlinear light diffraction driven by a square wave voltage in samples of polymer dispersed liquid crystals. The short peak (2-3 ms) in the leading edge of the signal is shown to be due to the occurrence of a transient amplitude grating in the sample. This effect is explained by the measured dependence of the transmittivity on the light intensity, reported here for the first time. A simple theoretical model gives a satisfactory account of the experiental observations.

INTRODUCTION

Recently we have reported the observation and study of nonlinear light diffraction in dye-doped polymer dispersed liquid crystals (PDLC). We have demonstrated the existence of a threshold for the light self diffraction induced by a phase grating created by two beams crossing on the sample at a small angle¹. We have also shown that this behavior can be exploited to drive the nonlinear diffraction with an applied low frequency voltage, if both beams have intensity much lower than the threshold necessary for the onset of the self induced effect ².

In this paper we report a detailed study of this phenomenon showing that the peculiar temporal behavior of the diffracted signal can be explained by considering the transient superposition of an amplitude and a phase grating. This effect is supported by measurements which show for the first time that the response time of a beam transmitted through a PDLC sample is strongly dependent on the impinging intensity.

EXPERIMENTAL

The experimental set-up used is the usual one to study degenerate four wave mixing in liquid crystals. Two beams from an Argon Ion laser ($\lambda = 514.5$ nm) cross at an angle of about 0.5 deg the PDLC sample, with beams' diameters of 2 mm. In the experiment the intensity ratio of the two beams was $I_{01}/I_{01} \approx 1.3$ because of uneven reflections on the different surfaces. The samples were obtained by phase separation method using a homogeneous solution of E7, EPON 815, Capcure 3-800 and component B of epoxid glue from BOSTIK as curing agents. The orange dye D2 from BDH (0.02% in E7) was mixed to the liquid crystal in order to increase light absorption. The sample, $36\mu m$ thick, was bounded by conductive glasses.

In all the measurements carried out the total laser power was fixed in order to have

 $I_{01} + I_{02} < I_{th}$, where I_{th} is the threshold intensity for self-transparency. A low frequency (10Hz) square wave voltage was applied to the sample. For V > 13 Volts the diffracted signal could be detected and easily driven by the square wave.

This last observation underlines a difference between this effect and the one observed without application of low frequency field and showing an intensity threshold. In the latter case the phase grating occurs in droplets in the isotropic state, and the threshold I_{th} is just above the light intensity necessary to induce the nematicisotropic transition. On the contrary in the present case the droplets keep their nematic state. In fact we always have $I_{01} + I_{02} < I_{th}$, moreover we can drive the effect with a bias voltage, thus showing the role of droplets' reorientation which can occur only in the nematic state. The voltage was then fixed at V = 50V, for which we got two clear and clean diffracted beams.

Under these experimental conditions measurements of the brightest diffracted beam were performed at different laser powers P_L . The impinging beams intensities were proportional to it: $I_{01} = k_1 P_L$, $I_{02} = k_2 P_L$ and as expected the intensity I_3 of the diffracted beam was proportional to P_L^{3} : $I_3 = k P_L^{3}$. In fig. 1 the traces recorded by the scope for $P_L = 40 \text{ mW}$ (a); $P_L = 80 \text{ mW}$ (b) and $P_L = 100 \text{ mW}$ (c) are shown. The correspondent applied square wave voltage is also reported (d).

We want to point out how the shape of the diffracted signal changes by increasing the impinging optical power: from a square-type signal we see, first, the rise of a peak in the leading edge of the signal and, for higher powers, we see also a second lower peak appearing as the voltage is switched off. The shape of the diffracted signal, when both peaks are present, is clearly shown in fig. 2.

Several other measurements were necessary in order to figure out the origin of this behavior. First of all the transmission of the two fundamental beams was recorded in order to compare their time behavior to the one of I_3 and to check if the time dependence of I_3 was due only to a nonlinear enhancement of any effect present in the transmission of the fundamental beams. A typical result of these measurements is reported in fig. 3. Here I_3 (a) is compared to the transmitted intensity I_1 of the more powerful of the two beams (b). No peak is present in the fundamental beam under the same experimental conditions thus demonstrating that the peculiar time behavior of I_3 belongs to the nonlinear effect. A suggestion for its explanation came to us by a new series of measurements which are interesting by themselves and could deserve more detailed future investigations since the reported results may affect devices made by PDLC.

We have recorded the transmitted fundamental beam I_1 at different laser power, measuring the rise and falltime of the signal. The interesting features observed were a relevant decrease of the risetime τ_{on} and a slight increase of the falltime τ_{off} of the signal as the laser power was increased. These data are reported in fig. 4, where τ_{on} and τ_{off} are reported vs P_r .

It is important to underline that the time constants can vary but the final transparency doesn't depend on the impinging light intensity, but only by the applied voltage. This fact is shown in fig. 5 where the steady state value of I_1 is plotted vs P_L . The linear behavior observed demonstrates that transparency (proportional to the slope) is constant.

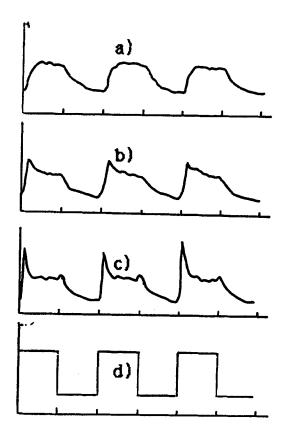


Fig. 1 - Diffracted intensity I3 for different values of the laser power P_L: a) 40mW, b) 80mW, c) 100mW; d) is the applied square wave voltage. Time scale is 50 ms/div.

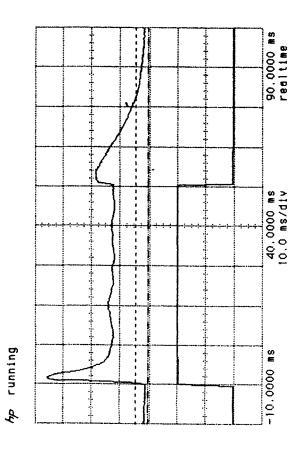


Fig. 2 - Hard copy of the oscilloscope. Channel 1:13, the double peak shape is clear; channel 2: applied voltage.

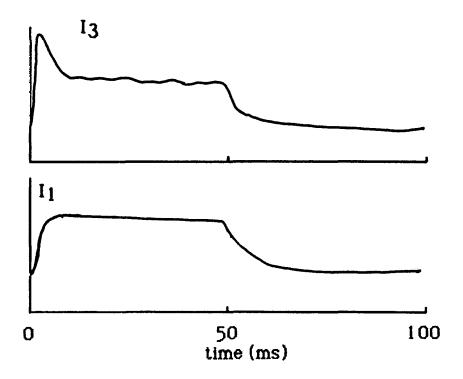
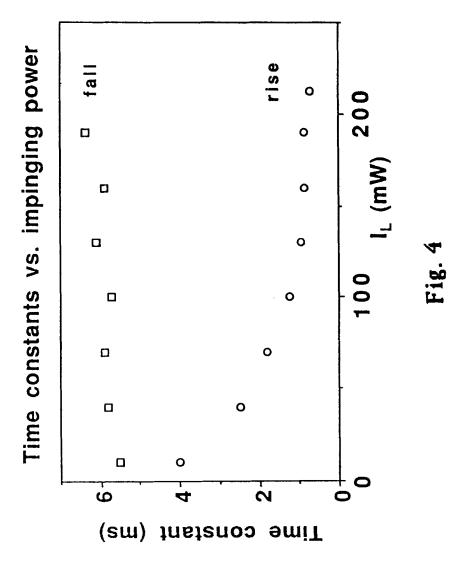
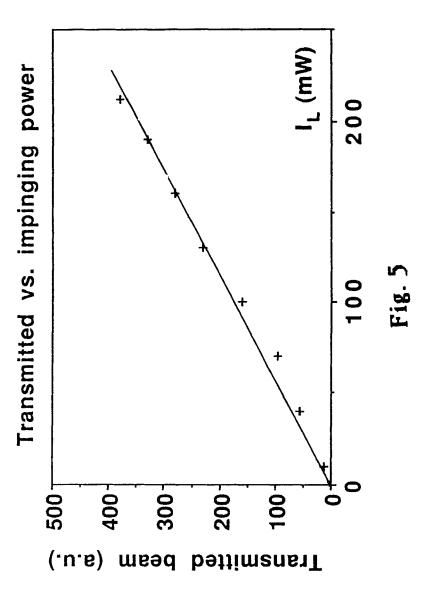


Fig. 3 - Temporal behaviour of I3 and I1





DISCUSSION

The new observations reported here of a risetime strongly dependent on the impinging light intensity is easily understandable. In fact in a PDLC sample the voltage necessary to get transparency is strongly dependent on temperature V = V(T) while it is well known that the risetime depends on the applied voltage: $\tau_{on} = \tau_{on}(V)$. Therefore, since we have a local increase of the temperature in the sample due to light absorption, we can expect the observed dependence $\tau_{on} = \tau_{on}(I)$.

We will show in the following the consequences of this effect on the nonlinear diffraction. When the two beams cross on the sample they produce an interference pattern described by the intensities distribution

$$I = I_0 [1 + m \cos(2\pi x/\Lambda)]$$
 (1)

where $I_0 = I_{10} + I_{20}$, x is the transversal coordinate, $m = 2 - \sqrt{I_{10}I_{20}}/I_0$ is the fringes' modulation and $\Lambda = \lambda/\sin\theta$ is the grating constant for a crossing angle θ .

Then, since according to our observations the risetime of transmission depends on the intensity, we expect that applying a voltage to the sample to switch it to the transparent state, the risetime will be shorter for the maxima of interference pattern as compared to the one for the minima.

By calling $\tau_{on}^{\ \ m}$ the transparency risetime in the maxima and $\tau_{on}^{\ \ m}$ the same quantity for minima, for a time of the order of $\tau_{on}^{\ \ m} - \tau_{on}^{\ \ m}$ we get transmission channels in the sample in the location of the interference maxima thus producing a <u>transient amplitude grating</u>. This grating cannot be stable since after a time $\tau_{on}^{\ \ m}$ also in the location of minima we obtain the same transparency as in the location of maxima, therefore only a phase grating will be possible in the steady state.

A similar effect can be also observed when the voltage is switched off if the falltime $\tau_{\rm off} = \tau_{\rm off}$ (I), but we expect a much weaker effect since we have measured a very weak dependence of $\tau_{\rm off}$ on the light intensity. Some simple calculation can give us a good qualitative agreement with the experimental observations.

The transient amplitude grating is due to a time dependent transmittivity which can be written as

$$T(t) = \overline{T(t)} + 1/2 \Delta T(t) \sin(2\pi x/\Lambda)$$
 (2)

where

$$\overline{T(t)} = T_{high} \frac{T_{high} - T_{ligh}}{2} \left[\exp\left(-t/\tau_{M}\right) + \exp\left(-t/\tau_{m}\right) \right]$$

is the average transmittivity and

$$\Delta T(t) = (T_{\text{high}} - T_{\text{low}}) \left[\exp(-t/\tau_{\text{M}}) - \exp(-t/\tau_{\text{m}}) \right]$$

is the peak-to-peak transmittance difference. Here T_{high} and T_{low} are transmittances in the on and in the off state respectively. According to our experimental observation $\Delta T(t)$ must vanish for $t \rightarrow 0$ and $t \rightarrow \infty$, thus the second term of eq. (2) becomes negligible small for $t >> (\tau_{cm}^{m} - \tau_{cm}^{M})$.

On the other hand thermal indexing will produce in the sample a phase grating, thus the overall transmittance will be given by

$$T(x,t) = [\overline{T(t)} + 1/2 \Delta T(t) \cos(2\pi x/\Lambda)] \exp[i\Phi_0 \cos(2\pi x/\Lambda)]$$

Therefore, in the plane wave approximation, the amplitude of each impinging beam after the sample is

$$A(x, t) = A_0 T(x, t) \exp(i 2\pi/\lambda \sin \theta_{in} x)$$

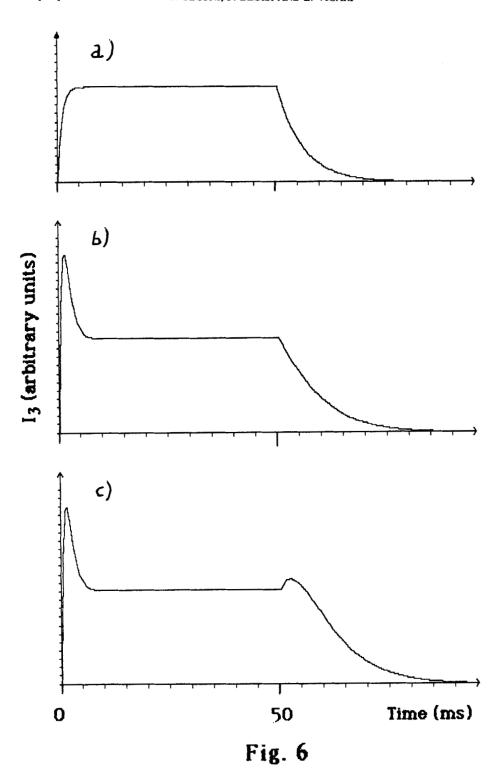
where θ_{in} is the incidence angle of the beam.

In the Fraunhofer approximation the far field amplitude $A(\theta, t)$ is proportional to the Fourier transform of $\Delta(x, t)$

$$\begin{split} A(\theta,t) &= A_0 \sum_{\nu=-\infty}^{\infty} \delta \left(\frac{\partial \cdot \theta_{in}}{\lambda} - \frac{\nu}{\Lambda} \right) \\ &= \{ \overline{T(t)} \ J_{\nu}(\Phi_0) - \frac{i}{4} \ \Delta T(t) \left[J_{\nu-1}(\Phi_0) + J_{\nu+1}(\Phi_0) \right] \} \end{split}$$

where $\delta(\theta)$ is the Dirac delta function and we assumed $\sin \theta_{in} \simeq \theta_{in}$. The amplitude of the first diffracted beam is obtained by placing n=1:

$$A_1 \leftarrow \widehat{T(t)} \ J_1(\Phi_0) - \frac{i}{4} \Delta T(t) \left[J_0(\Phi_0) + J_2(\Phi_0) \right]$$
 (3)



In (3) the first term is due to the phase grating alone while the second term represents the superposition of the amplitude and phase grating, which, as already mentioned, will last for a time duration of the order of $\tau_{om}^{m} - \tau_{om}^{M}$.

From eq. (3) it is possible to calculate the time dependence of the first diffracted beam using $\tau_{on}^{\ m}$, $\tau_{off}^{\ m}$ and $\tau_{off}^{\ m}$ as parameters in order to compare it to the experimental results: $I_{\tau}(t)$ is proportional to:

$$T^{2}(t) J_{1}^{2}(\Phi_{0}) + [J_{0}(\Phi_{0}) + J_{2}(\Phi_{0})]^{2} \Delta T^{2}(t)/16$$
 (4)

In fig. 6 we report the results of these calculations. Curve (a) comes from eq. (4) when we have used the same risetime and falltime for minima and maxima. In this case we have only a phase grating and no peak appear. This situation corresponds to low total impinging intensity.

In curve (b) we have a different risetime $\tau_{on}^{\ m}=0.85\,\text{ms}$ and $\tau_{on}^{\ M}=1.15\,\text{ms}$ and falltime $\tau_{off}^{\ m}=5.85\,\text{ms}$ and $\tau_{off}^{\ M}=6.15\,\text{ms}$.

Now a peak appears in the leading edge of the signal as experimentally observed at intermediate impinging intensities. In the third curve (c) also a slightly different falltime have been used ($\tau_{\rm off}^{\ \ m}=5.55{\rm ms}$, and $\tau_{\rm off}^{\ \ M}=6.45{\rm ms}$) and we get a peak also in the final edge of the signal as experimentally observed at higher intensities.

The results of our calculations are in good qualitative agreement with our measurements. It seems not possible, at this stage, to get a complete quantitave fit of experimental data since it is not known the actual difference between the risetimes and falltimes at different locations of the grating.

In conclusion we have shown that the transient superposition of amplitude and phase grating gives rise to a short (2-3 msec) peak in the leading edge of a diffracted signal driven by a low frequency voltage.

This effect is quite interesting and makes PDLC sample suitable to study some fundamental effects due to light induced gratings; moreover it might be also exploited in some application dealing with light controlled beam deflection.

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