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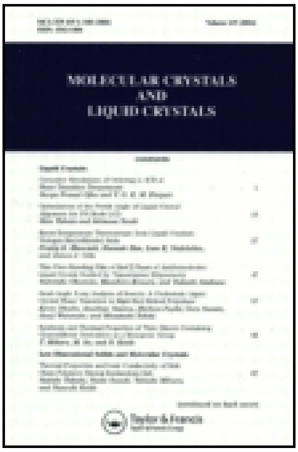

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Self Heterodyning: A Versatile Technique to Investigate Nematic Liquid Crystals

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SELF HETERODYNING: A VERSATILE TECHNIQUE TO INVESTIGATE NEMATIC LIQUID CRYSTALS

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Abstract It is described the "self - heterodyning" technique, based on the detection of the intensity oscillations which, in the far field zone, can be observed, in the initial transient, at the center of a beam which crosses a homeotropically aligned nematic liquid cristal. The simplicity and versatility of the technique is supported by the wide range of its applications.

INTRODUCTION

Nonlinear optical phenomena in liquid crystals have been deeply studied[1-4] and many related techniques have been developed which are successfully used. In particular, the self - phase modulation effect, which in given experimental geometries and conditions occurs in a light beam crossing a nematic liquid cristal (NLC), has been widely investigated[4,5]. The mechanism of formation of the ring system has been discussed in details for the steady state condition[6] and provides an easy method for phase measurements, which is exploited in investigations[7,8]. Only in the last year, however, the time dependence of this mechanism has been studied and exploited as a spectroscopic technique[9], in alternative to the traditional ones which can not be used due to the slowness of orientational relaxation processes in NLC's. The aim was an investigation of general character concerning light interaction with matter, but the interest and possibility of applications of an effect that allowed measurement of relative frequency shift as small as $\delta\omega/\omega \approx 10^{-15}$ was immediately clear.

In this paper, we show that a great quantity of physical informations are contained in the initial transient dynamics of the ring system formation and we present a detailed description of the self - heterodyning technique. This is based on the detection of the intensity oscillations that, after the switch on of the light beam, can be observed, in the far field zone, at the center of the outgoing beam itself. The simplicity and versatility of the technique is supported by the wide range of its applications.

THE TECHNIQUE

When a light beam crosses a NLC with a propagation direction that is parallel to the optical axis of the sample, a nonlinear phase shift is induced only if the light intensity I exceeds a threshold value I_F, called "threshold intensity for the Optical Freedericksz Transition". For $I > I_F$, the expression for $\Phi(t)$ is[5]:

$$\Phi(t) = \Phi_E/[1 + (\Phi_E - \Phi_0)\exp(-2\Gamma_T t)/\Phi_0]. \tag{1}$$

where

$$\Gamma_T = \Gamma(I/I_F - 1) \tag{2}$$

Γ is determined only by the material parameters and the cell thickness, following the expression:

$$\Gamma = (K_3/\gamma)(\pi/L)^2 \tag{3}$$

with γ = viscosity constant, K_3 = elastic constant and L = cell thickness. Φ_E is the value reached by the phase shift at the steady state and, far from saturation, is given by:

$$\Phi_E = \left(\frac{\omega}{2c\sqrt{\varepsilon_\perp}}\right)\left(\frac{\varepsilon_a L}{1 - \frac{9\varepsilon_a}{4\varepsilon_{//}} - \frac{K_3 - K_1}{K_3}}\right)\left(\frac{I}{I_F} - 1\right) \tag{4}$$

where c is the light speed, $\varepsilon_{//}$ and $\varepsilon_\perp$ are the dielectric constant when

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the electric field of light is respectively parallel or perpendicular to the optical axis of the NLC, ε_a is the dielectric anisotropy ($\varepsilon_a = \varepsilon_{//} - \varepsilon_{\perp}$) and K_1, K_3 are elastic constants of the material. We underline that Φ_E can become very large even for small excesses (≈10%) of the impinging intensity over the threshold value and, therefore, saturation can be easily achieved. Φ_0 is the "linear" phase shift which is related to the spontaneous fluctuational angle θ_0 of the optical axis which, initiates the second order, transition - like, light induced reorientation. The relation is given by the equation[5]:

$$\Phi_0 = [\omega L \varepsilon_a / 4c(\varepsilon_{\perp})^{1/2}]\, \theta_0^2 \qquad (5)$$

Following the dependence on (I/I_F-1) given by eq. (4), the nonlinear phase shift Φ_E reached in the steady state is modulated by the NLC sample over the cross - section of a light beam which exhibits a transverse intensity modulation. In particular, a gaussian profile of the impinging beam intensity produces a bell shaped phase front which is equivalent to have two waves propagating in each direction with different phases. In the far field zone, the picture results in a system of bright rings whose interference origin has been thoroughly investigated[5]: The total number of observed rings is $N_{tot} = \Phi_{max}/2\pi$, where Φ_{max} is the maximum of the nonlinear phase shift Φ_E over the beam cross section (i.e. in its centre).

During the initial transient that follows the light switch on, the ring pattern formation coincides with the observation, in the center of the running ring system, of intensity oscillations superimposed to an exponentially lowering baseline (Figure 1). These oscillations and their frequency can be explained by the same mechanism used for the ring pattern formation.

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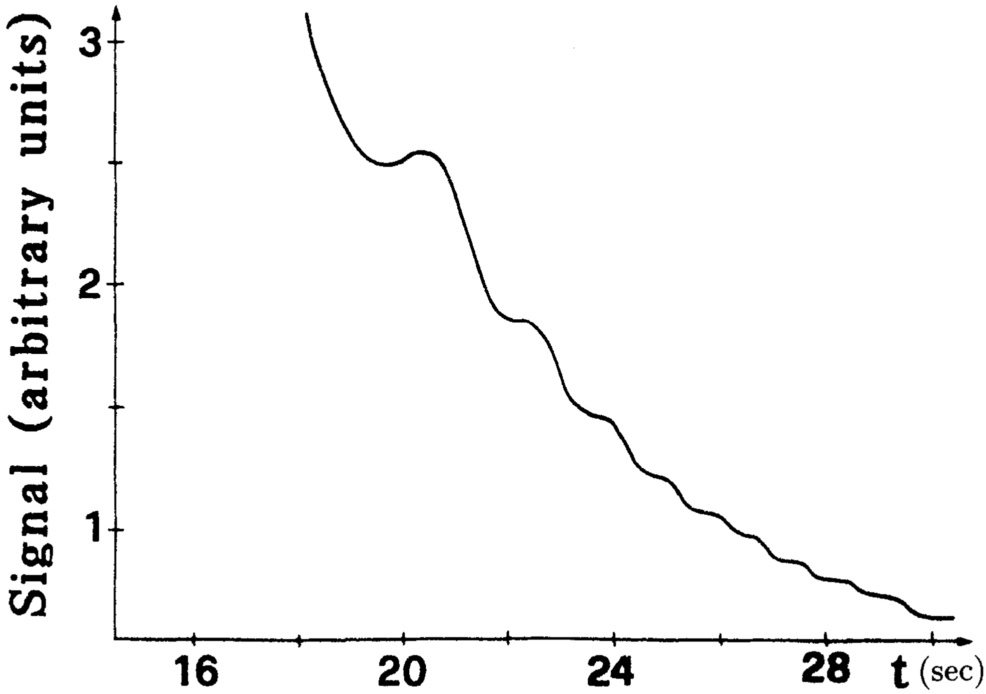


FIGURE 1 Transient intensity oscillations observed in the center of the beam outgoing from the sample.

In fact, in the far field zone, the central part of the picture is built up with the interference of waves from the center and the periphery of the outgoing beam since, in both regions, the normals to the phase front are along the propagation direction of the incident beam (Figure 2). If the optical frequency of one of the two waves changes of the amount $-\delta\omega$, a beating signal of "self heterodyning" is observed as a series of oscillations at the frequency $\delta\omega$. In the initial transient, the instantaneous frequency $-\delta\omega$ is given by the rate $\nu(t) = d[\Phi(t)]/dt$ of the instantaneous nonlinear phase shift induced by the sample in the central wave. Indeed, due to the transverse gaussian intensity modulation of the impinging beam, the nonlinear phase shift induced by the sample in the "peripheral" outgoing wave is negligible if compared with that of the central region and therefore the self heterodyning frequency $-\delta\omega$ coincides with the phase rate $\nu(t) = d[\Phi(t)]/dt$ induced in the central outgoing wave. Into eq. (1), we can thus substitute $\Phi(t)$ with $2\pi N(t)$ and Φ_E with $2\pi N_m$, where $N(t)$ is the number of observed oscillations at time t and N_m its total number:

$$N(t) = N_m/[1 + (2\pi N_m - \Phi_0)\exp(-2\Gamma_T t)/\Phi_0]. \quad (6)$$

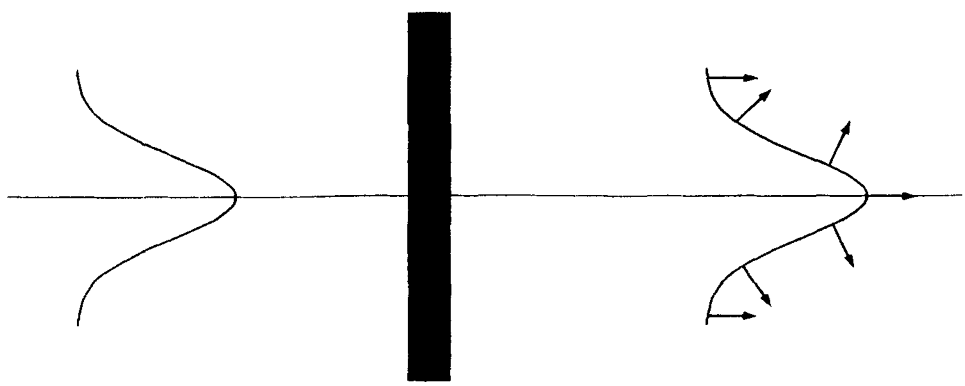
Equation (6) can be approximated to obtain:

$$\ln[2\pi N(t)] = \ln[\Phi_0] + 2\Gamma_T t. \quad (7)$$

For the rate $\nu(t)$ we can write:

$$\nu(t) = d[\Phi(t)]/dt = -4\pi\Gamma[I/I_F - 1][1 - N(t)/N_m]N(t) \quad (8)$$

From the simple detection of the intensity oscillations observed after the switch on of the light in the center of the outcoming beam, we can therefore obtain informations about all the physical quantities involved in the phenomenon and recognized into equations (7) and (8).



a) *b)* *c)*

FIGURE 2 a): Intensity profile of the incident beam; b): NLC cell; c): The phase profile and its normals after the sample.

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The needed experimental set up is very simple and is shown in Figure 3. The light source is an Ar^+ laser and the gaussian shape of its beam has been previously checked. After a shutter and a calibrated system of beam splitter and power meter, the light is linearly polarized and focused (f = 150 mm) on the cell of the homeotropically aligned NLC which, if needed, can be also thermo - stabilized. The outgoing beam is chopped and its central part detected by a photodiode placed behind a 200 μm pinhole. The signal is sent to a lock-in amplifier which improves the Signal to Noise ratio and then to a data acquisition system which drives the shutter.

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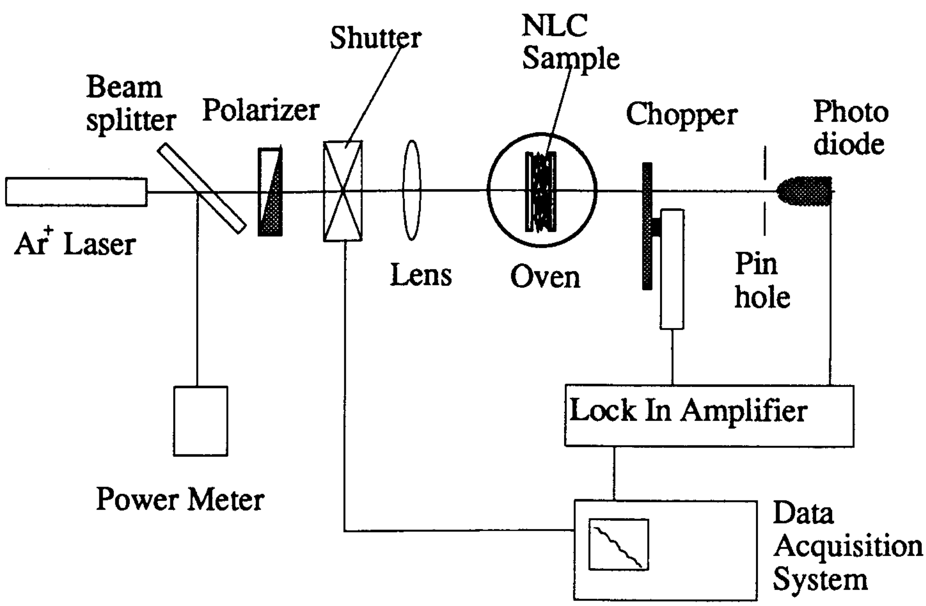

FIGURE 3 Sketch of the experimental setup

APPLICATIONS

The versatility of the "self - heterodyning" technique is here illustrated by some simple applications.

Threshold measurement

Eq. (7) can be written for the two different times t_i and t_j at which the N_i-th and N_j-th rings appear respectively ($N_i, N_j << N_m$):

$$\ln(N_i) - \ln(N_j) = 2\Gamma_{NT}[P/P_F - 1](t_i - t_j) \quad (9)$$

By the experimental determination of the angular coefficient $\mu = 2\Gamma_{NT}[P/P_F - 1]$ for two different values P_1 and P_2 of the impinging power, we find the threshold value P_F:

$$P_F = (P_2\mu_1/\mu_2 - P_1)/(\mu_1/\mu_2 - 1). \quad (10)$$

Direct measurement of the ratio K_3/γ

A first detailed illustration of this measurement performed in a sample of E7 (by BDH) NLC has been given elsewhere[10]. From eq. (7) we see that, by plotting $\ln[2\pi N(t)]$ vs time, we have a straight line (Figure 4) whose angular coefficient $2\Gamma_T$ directly gives K_3/γ if the cell thickness L and the ratio I/I_F are known:

$$(K_3/\gamma) = \Gamma_T/(I/I_F - 1)(\pi/L)^2 \quad (11)$$

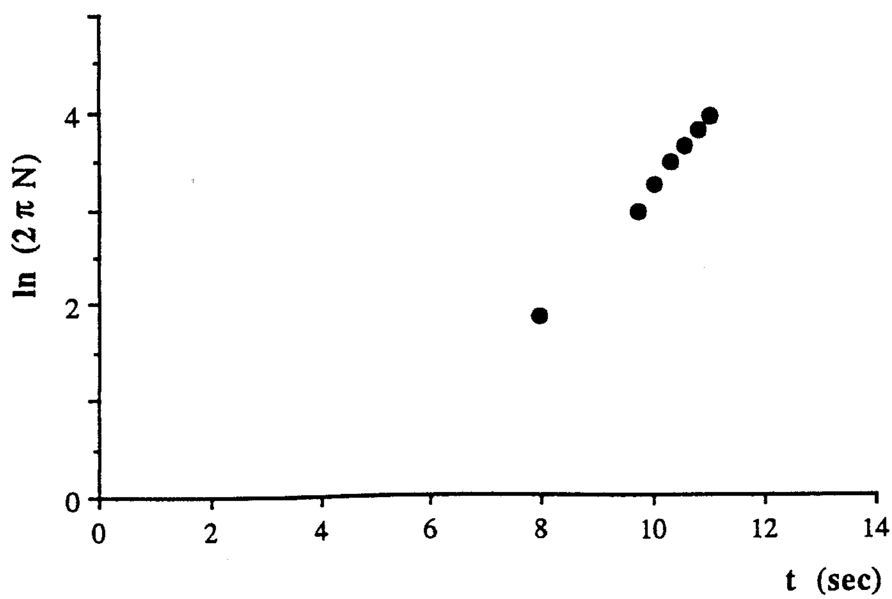

FIGURE 4 Typical trend of $\ln[2\pi N(t)]$ vs time.

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Investigation of threshold relaxation

The traditional approach to the characterization of relaxation phenomena is usually based on the hypothesis of an exponential behavior which allows for the definition of a single temporal parameter: the relaxation constant. The Optical Freedericksz Transition phenomenon produces optical nonlinearities that present a rather complex relaxation behavior which can be described by an exponential function only in the case of non - threshold effects. The threshold case, indeed, exhibits unique peculiarities which demand a deeper characterization, making useful also the measurement of the rate of relaxation $\nu(t)$. In our case, the relaxation can be studied by measuring the transient nonlinear phase shift $\Phi(t)$ induced by the NLC sample in the light which passes through it and, therefore, the rate of relaxation is just given by eq. (8). A detailed study and characterization of the problem is reported elsewhere[11]; here we show only the typical trend of $\nu(t)$ vs time (Figure 5). It is a bell shaped function which exhibits a very interesting feature: it has a maximum at a time moment that is not the initial moment.

Investigation of stochastic fluctuations

Following eq. (7), numerical extrapolation to t = 0 of the straight line of Figure 4, gives $\ln[\Phi_0]$. From eq. (5), we can obtain informations about the fluctuational angle θ_0 which initiates the Optical Freedericksz Transition and make deeper in a stochastic approach to the phenomenon.

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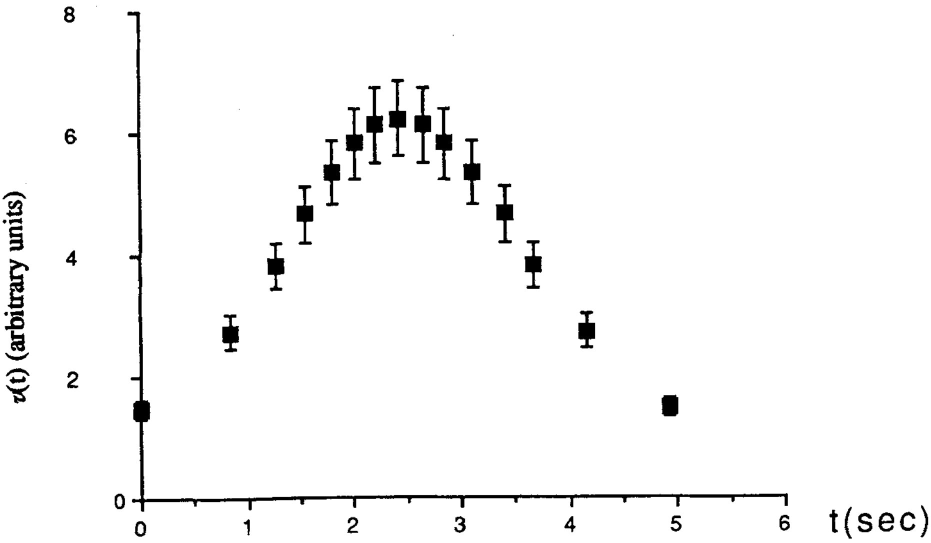

FIGURE 5 Typical trend of ι(t) vs time.

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