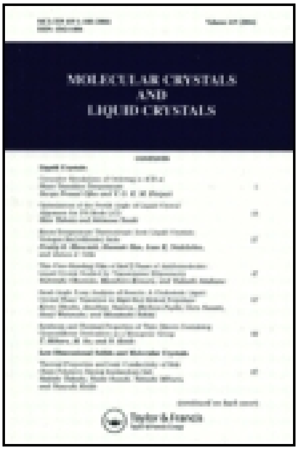

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Optically Induced Reorientation in a Hybrid Aligned Nematic Liquid Crystal Cell

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Optically induced reorientation in a hybrid aligned nematic liquid crystal cell

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Optically induced molecular reorientation in nematic liquid crystals is an interesting phenomenon, suitable of many practical applications, due to the large electric and optical anisotropy of such materials in the nematic phase. Therefore great importance has the study of the dynamics of the molecular reorientation. In the usual configuration the cell contains a homeotropically aligned nematic liquid crystal and a linearly polarized laser beam impinges on it. However, the presence of a Freedericksz transition requires the use of some experimental ingenious contrivances: such as the application of a magnetic field or performing the experiment only for a large enough incidence angle. We show here that the molecular reorientation dynamics of a nematic liquid crystal can be easily studied without any magnetic field, even at normal incidence, using a hybrid aligned nematic liquid crystal cell: i.e. a cell with homeotropic alignment on one boundary and planar alignment on the other.

Keywords: nonlinear optics; liquid crystals

INTRODUCTION

Optically induced molecular reorientation in nematic liquid crystals is an interesting phenomenon, both in static and in dynamic conditions, due to the large electric and optical anisotropy of such materials. Several studies have been carried out in this field[1-6]. Some experiment have been carried out in the so called "free surface" (i.e. glass—liquid-crystal—air) configuration, but more often the liquid crystal is in a confined (glass—

liquid-crystal—glass) configuration; in both cases the required molecular orientation is obtained by inducing some molecular orientation on the surface of the glass plates (surface coupling agents, mechanical rubbing, chemical etching, etc.). The use of substrates inducing homeotropic (nematic director perpendicular to substrate) or planar (nematic director parallel to substrate) surface orientation allows to obtain several cell alignments: *homeotropic* (homeotropic surface alignment on both cell boundaries), *planar* (parallel planar surface alignment on both cell boundaries), *twisted* (non parallel planar surface alignment on both cell boundaries), *hybrid* (homeotropic surface alignment on one cell boundary, planar surface alignment on the other).

Usually, to study optical molecular reorientation in nematic liquid crystals, a linearly polarized laser beam impinges on a homeatropically oriented cell. However, the presence of a Freedericksz transition[7] requires the application of a magnetic field or the execution of the experiment only for a large incidence angle.

Here we show that the molecular reorientation of a nematic liquid crystal can be easily studied without any magnetic field, even at normal incidence, using a hybrid aligned nematic liquid crystal cell.

The absence of a Freedericksz threshold allows the use a relatively low power pump beam, i.e. an unfocused laser beam, and this has the advantage to not introduce either non-local or thermal effects.

HYBRID ALIGNED CELL

Choosing the z axis normal to the cell boundaries and the x axis so that the nematic director $\hat{\mathbf{n}}$ is everywhere in the xz plane, it is possible to write $\hat{\mathbf{n}} \equiv (\sin\vartheta, 0, \cos\vartheta)$ so that the liquid crystal configuration is described by the function $\vartheta(\zeta)$ defined for $\zeta \in [-0.5, +0.5]$, where we have used the dimensionless variable $\zeta = z/h$ (h is the cell thickness). By minimization of the free energy we obtain the usual differential equation

$$\frac{h^2}{K_{33}}\Gamma_{opt} + \left(1 - \kappa \sin^2 \vartheta\right) \frac{d^2\vartheta}{d\zeta^2} - \kappa \sin\vartheta \cos\vartheta \left(\frac{d\vartheta}{d\zeta}\right)^2 = 0 \qquad (1)$$

where $\kappa = 1 - \frac{K_{11}}{K_{33}}$ is the elastic anisotropy, K_{ij} are the liquid crystal Frank elastic constants and the optical torque per unit volume Γ_{opt} is the y component of the torque produced by the optical electric field on the liquid crystal molecules:

$$\Gamma_{opt} = \varepsilon_0 E_0^2 n_e^2 u \left[\left(1 - \eta^2\right) \sin\vartheta \cos\vartheta - \eta \left(\sin^2\vartheta - \cos^2\vartheta\right)\right]$$

since

$$\mathbf{\Gamma}_{opt} = \mathbf{P} \times \mathbf{E} = \varepsilon_0 n_e^2 u \left(\hat{\mathbf{n}} \cdot \mathbf{E}\right) \left(\hat{\mathbf{n}} \times \mathbf{E}\right)$$

where $u = 1 - \frac{n_o^2}{n_e^2}$ is the optical anisotropy, n_o and n_e are ordinary and extraordinary refractive indices, respectively, $\mathbf{E} \equiv (E_x, 0, E_z) \equiv (E_0, 0, \eta E_0)$ is the optical electric field inside the sample, E_0 is the optical electric field outside the sample, and

$$\eta = \frac{E_z}{E_x} = -\frac{u \sin\vartheta \cos\vartheta}{1 - u \sin^2\vartheta} .$$

The boundary conditions for a hybrid aligned cell, assuming strong anchoring, are

$$\begin{aligned} \vartheta(-0.5) &= 0 \\ \vartheta(+0.5) &= \frac{\pi}{2} . \end{aligned}$$

SMALL ELASTIC ANISOTROPY APPROXIMATION

Let us observe that in absence of optical electric field ($\mathbf{\Gamma}_{opt} = \mathbf{0}$) the nematic director configuration is determined by the liquid crystal elastic anisotropy κ. However, even if the first integral of eq.(1) with $\Gamma_{opt} = 0$ is well known[8], the analytical expression of $\vartheta(\zeta)$ can be found only for special values of the elastic anisotropy κ ($\kappa = 0$ and the two extreme values $\kappa = 1$ and $\kappa \to -\infty$).

If $K_{11} = K_{33}$, i.e. in the so-called "one-constant approximation" we have $\kappa = 0$ so that the differential equation reduces to $\frac{d^2\vartheta}{d\zeta^2} = 0$ which has the trivial solution

$$\vartheta_{|\kappa=0}(\zeta) = \frac{\pi}{2}\left(\frac{1}{2} + \zeta\right). \tag{2}$$

If $K_{11} \ll K_{33}$ (or $K_{11} \gg K_{33}$), i.e. highly anisotropic material with positive (or negative) anisotropy we have $\kappa = 1$ (or $\kappa \to \infty$) and it is easy to verify that the solutions (Fig. 1) are

$$\vartheta_{|\kappa=1}(\zeta) = \arcsin\left(\frac{1}{2} + \zeta\right)$$

or

$$\vartheta_{|\kappa\to\infty}(\zeta) = \frac{\pi}{2} - \arcsin\left(\frac{1}{2} - \zeta\right).$$

Looking at Fig. 1 it is reasonable to assume that for small elastic anisotropy ($|\kappa| \ll 1$) the function $\vartheta(\zeta) - \vartheta_{|\kappa=0}(\zeta)$ is symmetric with respect to $\zeta = 0$; moreover such function must vanish for $\zeta = \pm 0.5$. Therefore let us assume that the solution of eq.(1) has the form

$$\begin{aligned} \vartheta(\zeta) &= \vartheta_{|\kappa=0}(\zeta) + \Theta_\kappa \cos(\pi\zeta) \\ &= \frac{\pi}{2}\left(\frac{1}{2} + \zeta\right) + \Theta_\kappa \cos(\pi\zeta) \end{aligned} \tag{3}$$

where the constant Θ_κ is small enough to allow us to expand all terms of eq.(1) in a power series retaining only first order terms in Θ_κ:

$$\begin{aligned} &\left(1 - \kappa\sin^2\vartheta\right)\frac{d^2\vartheta}{d\zeta^2} - \kappa\sin\vartheta\cos\vartheta\left(\frac{d\vartheta}{d\zeta}\right)^2 \\ = &\ \frac{1}{8}\pi^2\left(-\kappa - 8\Theta_\kappa + 4\kappa\Theta_\kappa + 10\kappa\Theta_\kappa\sin(\pi\zeta)\right)\cos(\pi\zeta). \end{aligned}$$

Now integrating between $\zeta = -0.5$ and $\zeta = +0.5$ we obtain

$$-\kappa - 8\Theta_\kappa + 4\kappa\Theta_\kappa = 0$$

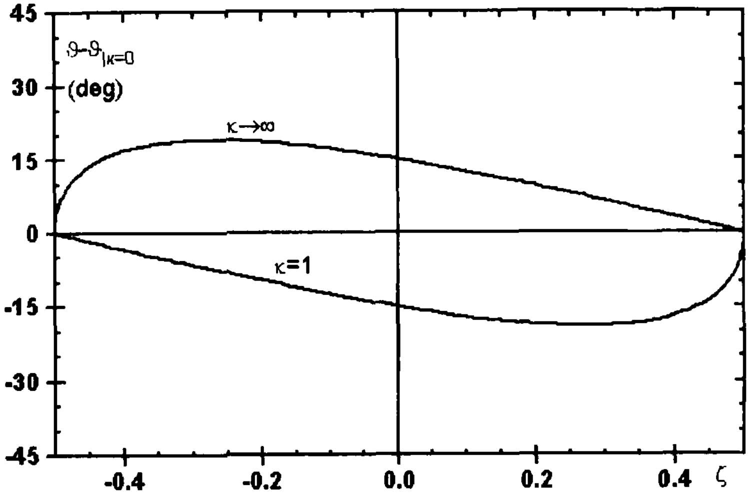

FIGURE 1 Nematic director configuration for a hybrid aligned cell: the difference with respect to the linear configuration $\vartheta_{|\kappa=0}(\zeta) = \frac{\pi}{2}\left(\frac{1}{2}+\zeta\right)$ is shown in the two extreme situations: $\kappa = 1$ (i.e. $K_{11} << K_{33}$) and $\kappa \to \infty$ (i.e. $K_{11} >> K_{33}$).

or

$$\Theta_\kappa = -\frac{1}{4}\frac{\kappa}{2-\kappa} = -\frac{1}{4}\frac{K_{33}-K_{11}}{K_{11}+K_{33}} . \tag{4}$$

We can therefore conclude that, for small elastic anisotropies

$$\begin{aligned} \vartheta(\zeta) &\simeq \vartheta_{|\kappa=0}(\zeta) + \Theta_\kappa \cos(\pi\zeta) \\ &\simeq \frac{\pi}{2}\left(\frac{1}{2}+\zeta\right) - \frac{1}{4}\frac{\kappa}{2-\kappa}\cos(\pi\zeta) . \end{aligned} \tag{5}$$

COMPARISON WITH NUMERICAL SOLUTION

In general the differential equation (1) has the first integral

$$\left(1-\kappa\sin^2\vartheta\right)\left(\frac{d\vartheta}{d\zeta}\right)^2 = C \tag{6}$$

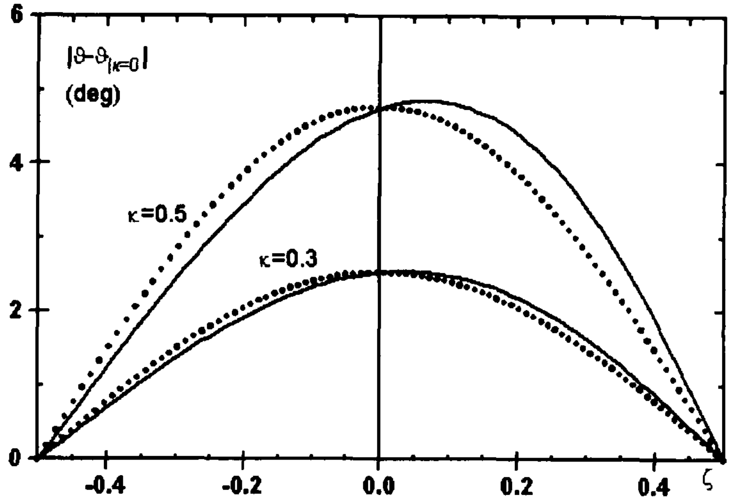

FIGURE 2 Plot of $\vartheta(\zeta) - \vartheta_{|\kappa=0}(\zeta)$. Comparison between the exact numerical solution of eq. (1) given by eq. (8) (solid lines) and the values given in the small elastic constant approximation by eq.(5) (dots).

where C is an integration constant whose value can be obtained by rewriting (6) as

$$d\vartheta\sqrt{1-\kappa\sin^2\vartheta} = \sqrt{C}d\zeta \tag{7}$$

and integrating over the whole cell thickness:

$$\int_0^{\frac{\pi}{2}} d\vartheta\sqrt{1-\kappa\sin^2\vartheta} = \sqrt{C}\int_{-0.5}^{+0.5} d\zeta = \sqrt{C}\,.$$

Now another integration of (7) gives an implicit relation which allows us to compute the inverse function

$$\zeta(\vartheta) = \frac{\int_0^{\vartheta} d\vartheta'\sqrt{1-\kappa\sin^2\vartheta'}}{\int_0^{\frac{\pi}{2}} d\vartheta'\sqrt{1-\kappa\sin^2\vartheta'}} - \frac{1}{2}\,. \tag{8}$$

The values obtained numerically integrating and inverting this expression are compared in Fig. 2 with values obtained using the small elastic anisotropy approximation (5) described in the previous section. As can be seen, since typical values of the elastic anisotropy κ are in the range

[0.1, 0.3], our approximation can be usefully applied to all practical situations.

HYBRID ALIGNED CELL DYNAMICS

In our experimental configuration, the probe beam diameter (HeNe laser beam, $2w_{HeNe} = 0.5$mm) is much smaller than the pump beam (Nd:YAG laser beam, $2w_{Nd:YAG} = 5$mm) so we can approximate the pump beam with a plane wave. Following the Ericksen and Leslie theory, the dynamics of the nematic director configuration, described by the function $\vartheta(\zeta, t)$, must satisfy the equation[9]

$$\begin{aligned} \rho_1 \frac{\partial^2 \vartheta}{\partial t^2} + \gamma_1 \frac{\partial \vartheta}{\partial t} &= \frac{K_{33}}{h^2}\left[\left(1 - \kappa \sin^2\vartheta\right)\frac{\partial^2\vartheta}{\partial\zeta^2} - \kappa \sin\vartheta\cos\vartheta\left(\frac{\partial\vartheta}{\partial\zeta}\right)^2\right] \\ &+ \frac{1}{h}\left(\frac{\gamma_1+\gamma_2}{2} - \gamma_2\cos^2\vartheta\right)\frac{\partial v_x}{\partial\zeta} + \Gamma_{opt} \end{aligned} \qquad (9)$$

coupled with the equation of the motion of the fluid

$$\begin{aligned} \rho\frac{\partial v_x}{\partial t} &= \frac{1}{h^2}\frac{\partial}{\partial\zeta}\left[\left(\frac{\alpha_3+\alpha_4+\alpha_6}{2} + \alpha_1\sin^2\vartheta\cos^2\vartheta - \gamma_2\cos^2\vartheta\right)\frac{\partial v_x}{\partial\zeta}\right] \\ &+ \frac{1}{h}\frac{\partial}{\partial\zeta}\left[\left(\alpha_3 - \gamma_2\cos^2\vartheta\right)\frac{\partial\vartheta}{\partial t}\right] \end{aligned} \qquad (10)$$

where ρ is the liquid crystal density, ρ_1 is the moment of inertia per unit volume, α_i and γ_i are the Leslie viscosity coefficients, $v_x(\zeta, t)$ is the x component of the flow velocity and the boundary conditions, assuming strong anchoring, are given by

$$\begin{array}{ll} \vartheta(-0.5, t) = 0 & v_x(-0.5, t) = 0 \\ \vartheta(+0.5, t) = \frac{\pi}{2} & v_x(+0.5, t) = 0\,. \end{array}$$

The optical torque ($\Gamma_{opt} \neq 0$) produces a molecular reorientation ($\frac{\partial\vartheta}{\partial t} \neq 0$); this induces a molecular flow ($v_x \neq 0$) which in turn influences the molecular reorientation. The simultaneous solution of eqs (9) and (10) is difficult, so we'll introduce some approximations. The rotation-flow coupling cannot be neglected when the optical field is switched on and for some time after it is switched-off. Since in the present work we are mainly interested in the relaxation time, we will neglect the molecular flow effects. Moreover, as usual, we neglect the inertial term $\rho_1 \frac{\partial^2 \vartheta}{\partial t^2}$ which is important only for subnanosecond excitation pulses[1]. Therefore, a short time after pump beam has been switched off, $\vartheta(\zeta, t)$ must satisfy the following differential equation

$$\gamma_1 \frac{\partial \vartheta}{\partial t} = \frac{K_{33}}{h^2}\left[\left(1 - \kappa \sin^2 \vartheta\right)\frac{\partial^2 \vartheta}{\partial \zeta^2} - \kappa \sin \vartheta \cos \vartheta \left(\frac{\partial \vartheta}{\partial \zeta}\right)^2\right]. \tag{11}$$

Since in a cell with hybrid alignment there is no Freedericksz threshold, we can use a low intensity pump beam and therefore we can assume that $\vartheta(\zeta, t)$ has again the form

$$\begin{aligned} \vartheta(\zeta, t) &= \vartheta_{|\kappa=0}(\zeta) + \Theta_0(t)\cos(\pi\zeta) \\ &= \frac{\pi}{2}\left(\frac{1}{2} + \zeta\right) + \Theta_0(t)\cos(\pi\zeta) \end{aligned} \tag{12}$$

similar to (3) with the constant factor Θ_κ replaced by a time dependent term $\Theta_0(t)$.

Repeating the same procedure used before to determine Θ_κ we obtain for $\Theta_0(t)$ the differential equation

$$\frac{2\gamma_1 h^2}{\pi^2 (K_{33} + K_{33})}\dot{\Theta}_0 = \frac{\kappa}{4(\kappa - 2)} - \Theta_0$$

which has the solution

$$\Theta_0(t) = \Theta_\kappa - C_0 \exp\left(-\frac{t}{\tau_{OFF}}\right) \tag{13}$$

where

$$\tau_{OFF} = \frac{\gamma_1 h^2}{\pi^2 (K_{11} + K_{33})/2} \tag{14}$$

and C_0 is an integration constant whose value is determined by the initial conditions, i.e. by the nematic director distortion when the relaxation begins. Therefore we will expect that $\Theta_0(t)$ has an exponential behavior whose starting value depends on the intensity and pulse duration, the elastic constants, the viscosity coefficients etc., but whose time constant has a very simple expression.

THE EXPERIMENT

The sample (Fig.3) is obtained by placing the liquid crystal (K15 by BDH) between two glass plates spaced h = 100 μm from each other. One glass plate has been treated with DMOAP (n,n–dimethyl–n octadecyl-3–aminopropyl–trimethoxysilyl chloride)[10] to obtain homeotropic molecular alignment, while the other has been treated with Polyvinylformal and then rubbed to induce planar alignment in the x direction.

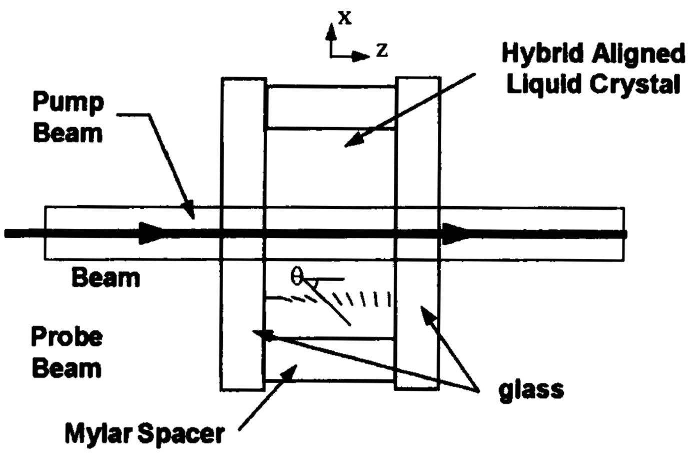

FIGURE 3 Sample configuration. Liquid crystal is K15 (4-cyano-4'-n-alkylbiphenyl): T_c = 35.5 °C, K_{11} = 5.65 pN, K_{33} = 7.40 pN, γ_1 = 0.0885 N/m^{2}s, n_e = 1.7, n_o = 1.5. Sample thickness is h = 100 μm.

The experimental setup is sketched in Fig. 4. The pump is a pulsed Nd:YAG laser beam (wavelength λ_{pump} = 1064 nm, pulse duration $\sim$ 5 ns) linearly polarized in the plane of the figure (xz plane) orthogonally impinging on the sample; the probe is a low power cw HeNe laser beam

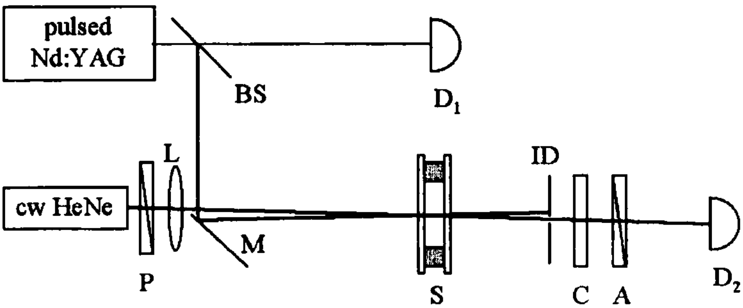

FIGURE 4 Experimental setup. BS: beam splitter, D_1: energy meter, M: mirror, S: sample, ID: iris diafragm, P: polarizer, C: compensator on a rotating mount, A: polarizer on a rotating mount, D_2: photodiode detector. Experiment was performet at $T = 26.9$ °C.

(wavelength $\lambda_{probe} = 653.8$ nm) linearly polarized at 45° with respect to the plane of the figure. Pump beam is unfocused ($2w_{Nd:YAG} = 5$ mm) while probe beam is slightly focused ($2w_{HeNe} = 0.5$ mm) at the center of the spot produced by the pump. Probe beam is detected by a photodiode in a PSCA (Polarizer-Sample-Compensator-Analizer) ellipsometric arrangement. Before each set of measures the compensator and the analizer are rotated to minimize the signal received by the detector, even if it was impossible to obtain a zero intensity condition since the sample partially depolarizes the impinging light.

Using the Jones matrix method it can be found[12] that the intensity at the detector is proportional to the square modulus of the complex number

$$
\begin{aligned}
L &= L_e \exp(i\delta_{Se}) + L_o \exp(i\delta_{So}) \\
L_e &= \cos(\alpha_C)\cos(\alpha_A - \alpha_C) - \sin(\alpha_C)\sin(\alpha_A - \alpha_C)\exp(i\delta_C) \\
L_o &= \sin(\alpha_C)\cos(\alpha_A - \alpha_C) + \cos(\alpha_C)\sin(\alpha_A - \alpha_C)\exp(i\delta_C)
\end{aligned}
$$

where δS_o and δS_e are the phase shift introduced by the sample on an ordinary or an extraordinary ray, respectively, $\delta_C = \delta C_f - \delta C_s$ is the relative phase shift introduced by the compensator between the fast and slow component, α_C and α_A are compensator and analyzer rotation angles.

If we assume $\delta_{Se}(t) = \delta^0_{Se} + \delta^*_{Se}(t)$ and preliminarly set compensator and analyzer for a null detection condition when $\delta_{Se}(t) = \delta^0_{Se}$ we get

$$L_e \exp\left(i\delta^0_{Se}\right) + L_o \exp\left(i\delta^0_{So}\right) = 0$$

and therefore

$$L = L_e\left[\exp\left(i\delta^*_{Se}\right) - 1\right]\exp\left(i\delta^0_{Se}\right) .$$

Since

$$|L|^2 = |L_e|^2 \ \left|\exp\left(i\delta^*_{Se}\right) - 1\right|^2 = 2\ |L_e|^2\ \left(1 - \cos\delta^*_{Se}\right) \tag{15}$$

the detected signal is proportional to

$$1 - \cos\left[\delta^*_{Se}(t)\right] . \tag{16}$$

The time-dependent (pump beam induced) component of the sample relative phase shift can be computed as

$$\delta^*_{Se}(t) = \frac{2\pi h}{\lambda_{probe}}\int_{-0.5}^{+0.5} n_{eff}(\zeta, t)\, d\zeta - \frac{2\pi h}{\lambda_{probe}}\int_{-0.5}^{+0.5} n_{eff}(\zeta)\, d\zeta$$

where

$$n_{eff}(\zeta, t) = \left[\frac{\cos^2\vartheta(\zeta, t)}{n_o^2} + \frac{\sin^2\vartheta(\zeta, t)}{n_e^2}\right]^{-\frac{1}{2}}$$

is the effective (extraordinary) refractive index, with $\vartheta(\zeta, t)$ given by eq.(12) and $n_{eff}(\zeta)$ is obtained by the same equation, with $\vartheta(\zeta)$ given by eq.(5).

RESULTS AND CONCLUSIONS

Fig. 5 shows the signal obtained from detector. Superimposed to the hard copy of the oscilloscope screen is the curve obtained using the theory discussed in the present paper. As can be seen, despite its semplicity, our

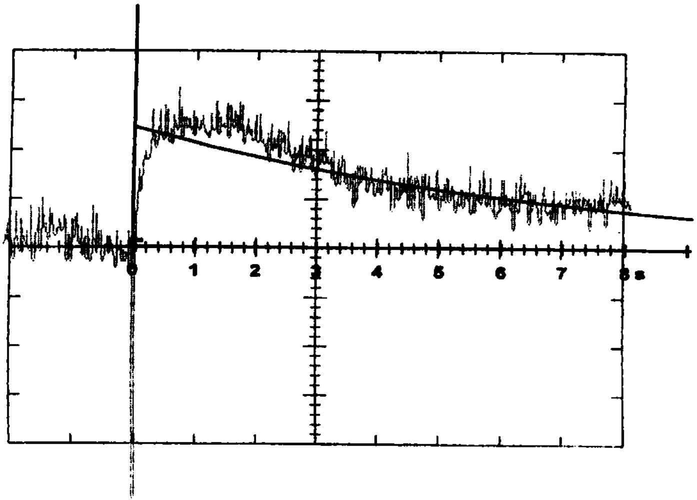

FIGURE 5 Signal obtained from the detector D_2. Superimposed to the hard copy of the oscilloscope screen is the curve given by eq.(16), using expressions (12) and (5).

model is in good agreement with experimental data. The use of a hybrid liquid crystal alignment allows to use an unfocused pump beam (energy density 0.9 J/cm^2 per pulse, pulse duration $\sim$ 5 ns) at normal incidence with no biasing static magnetic field.

An advantage of such experimental configuration is that there is no need, writing eq.(1), to take into account non-local effects due to finite size of pump beam[6]. Another advantage is that, since sample is highly transparent and the pump beam energy density is relatively low, we can neglect the thermal effect.

However our model does not describes properly the behavior of the sample for a little time (about 0.25 τ_{OFF}) after the pump beam pulse. This is probably due to the presence of flow-orientational coupling. Therefore further studies are needed to correctly describe both the switch-on and the switch-off behavior of the sample.

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