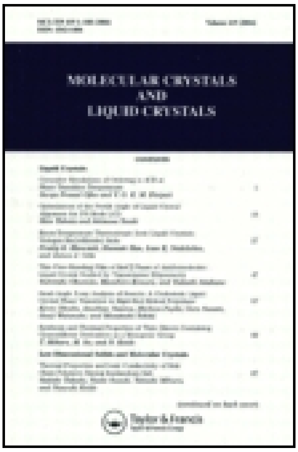

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# Effect of Temperature on the Nonlinear Optical Behavior of a Homeotropic Nematic Liquid Crystal

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# Effect of Temperature on the Nonlinear Optical Behavior of a Homeotropic Nematic Liquid Crystal

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According to the model of optical reorientation of the molecular director, we report a study of the effect of temperature variations on the nonlinear optical behavior of a homeotropic liquid crystal cell. We show that the model is in good agreement with our experimental data obtained for the K18 nematic liquid crystal. By numerical simulation we also show what is the role played by the different material parameters in determining the temperature behavior.

## INTRODUCTION

It is well known that the giant optical nonlinearities of liquid crystals which comes from molecular director reorientation are strongly affected by light induced thermal effects.[1,2] Therefore it is often necessary to take them into account to explain experimental data obtained from optical reorientation of the molecular director. For this reason it is still an open problem to experimentally distinguish the contribution of thermal phenomena from the one due to director reorientation in the nonlinear optical behavior of a nematic liquid crystal. Therefore it is important to know the effects of temperature variations on the mentioned reorientational phenomena.

In a recent paper, we have reported[3] a study of the optical Freedericksz transition performing measurements of the nonlinear phase shift at different temperatures. We have shown that the threshold and the saturation behavior are in excellent agreement with the one expected from the Meier-Saupe theory.

In this paper we present a detailed study of the influence of different material parameters on the nonlinear behavior of an homeotropic cell of nematic liquid crystal for linear polarized light at normal incidence, showing the role played by temperature variations. Moreover we show that the expected behavior is in good agreement with our experimental data obtained using the liquid crystal K18 by BDH.

In the next section we give a brief review of the theoretical framework used by us. The third section is devoted to discuss the results of the calculations and of the experiments performed on K18 at different temperatures.

## THEORETICAL FRAMEWORK

We consider a homeotropic nematic cell under strong anchoring conditions and a linear polarized laser beam normally impinging on it and traveling in the $z$ direction; moreover we assume that the cell boundaries are located at $z = -d/2$ and $z = +d/2$. Above threshold the director orientation is determined by the tilt angle $\varphi$ with respect to $z$ axis which can be calculated from the equations[4]:

$$\int_0^{\varphi(z)} G(\varphi', \varphi_{max})\, d\varphi' = z + (d/2) \quad \text{for} \quad -d/2 < z < 0 \tag{1}$$

where

$$G(\varphi, \varphi_{max}) = \sqrt{\frac{1 - K \sin^2\varphi}{f(\varphi_{max}) - f(\varphi)}} \tag{2}$$

and

$$f(\varphi) = \frac{2\, S_z \sqrt{\varepsilon_e}}{cK_3 \sqrt{1 + (\varepsilon_a/\varepsilon_o)\cos\varphi}} \tag{3}$$

$K_1$ and $K_3$ are the splay and bend elastic constants, $K = 1 - K_1/K_3$ is the elastic anisotropy, $\varepsilon_o$ and $\varepsilon_e$ are the ordinary and extraordinary dielectric constants, $\varepsilon_a = \varepsilon_e - \varepsilon_o$ is the optical anisotropy, $c$ is the speed of light, $d$ is the thickness of the sample, $S_z$ is the $z$ component of the Poynting vector which, for a Gaussian beam (power $= P$, $1/e^2$ radius $= w$), is $S_z = P/\pi w^2$.

Because of the symmetry imposed by the strong anchoring condition, $\varphi(-d/2) = \varphi(+d/2) = 0$, we have the maximum tilt angle $\varphi_{max}$ in the center of the cell. Therefore it is possible to write:

$$\int_0^{\varphi_{max}} G(\varphi', \varphi_{max})\, d\varphi' = d/2 \tag{4}$$

Thus $\varphi_{max}$ can be calculated by this equation which can be rewritten as:

$$F(\varphi_{max}; \varepsilon_e, \varepsilon_o, K, K_3) = S_z\, d^2 = \frac{P\, d^2}{\pi w^2} \tag{5}$$

where

$$F(\varphi_{\max};\, \varepsilon_e,\, \varepsilon_o,\, K,\, K_3) = \frac{2\,cK_3}{\sqrt{\varepsilon_e}}$$

$$\times \left\{ \int_0^{\varphi_{\max}} d\varphi \sqrt{\frac{1 - K\sin^2\varphi}{[1 + (\varepsilon_a \varepsilon_o)\cos\varphi_{\max}]^{-1/2} - [1 + (\varepsilon_a/\varepsilon_o)\cos\varphi]^{-1/2}}} \right\}^2 \tag{6}$$

The elliptic integral in Equation (6) may be performed using standard techniques.

When $\varphi_{\max}$ is known, we get $\varphi(z)$, as usually, from Equation (1) and finally the nonlinear phase shift through the whole sample, given by

$$\Delta\Phi = \frac{2\pi}{\varphi} \int_{-d/2}^{+d/2} [\sqrt{\varepsilon_{\text{eff}}(z)} - \sqrt{\varepsilon_o}]\, dz \tag{7}$$

where

$$\varepsilon_{\text{eff}}(z) = \frac{2\,\varepsilon_o\,\varepsilon_e}{\varepsilon_o + \varepsilon_e + \varepsilon_a \cos 2\varphi(z)} \tag{8}$$

## RESULTS AND DISCUSSION

First of all we compare the experimental data obtained using K18 by BDH, at different temperatures, and the corresponding theoretical previsions. The experimental set-up and procedure was the usual one used to observe the self phase modulation of the light through a liquid crystal film and has been reported elsewhere.[3] The number of the diffraction rings, i.e., the nonlinear phase shift divided by $2\pi$, is plotted versus the optical power $P$ impinging on the sample in Figure 1. The beam and cell parameters are: wavelength $\lambda = 0.5145\ \mu$m, $1/e^2$ beam radius $w = 107.5\ \mu$m, cell thickness $d = 115\ \mu$m, while the sample parameters at different temperatures are taken from literature.[5] We notice that the values of the threshold power can be directly evaluated from

$$P_{\text{th}} = \frac{c\pi^3 w^2}{d^2} \frac{\varepsilon_e K_3}{(\varepsilon_e - \varepsilon_o)\sqrt{\varepsilon_o}} \tag{9}$$

and coincide with the limiting values computed by program.

The agreement is good enough to induce us to use our computer program to examine the effects of various parameters on the nonlinear optical behavior of the cell. To this aim it is convenient to introduce the normalized phase shift

$$\Delta\Phi^* = \frac{\Delta\Phi}{2\pi}\frac{\lambda}{d} \tag{10}$$

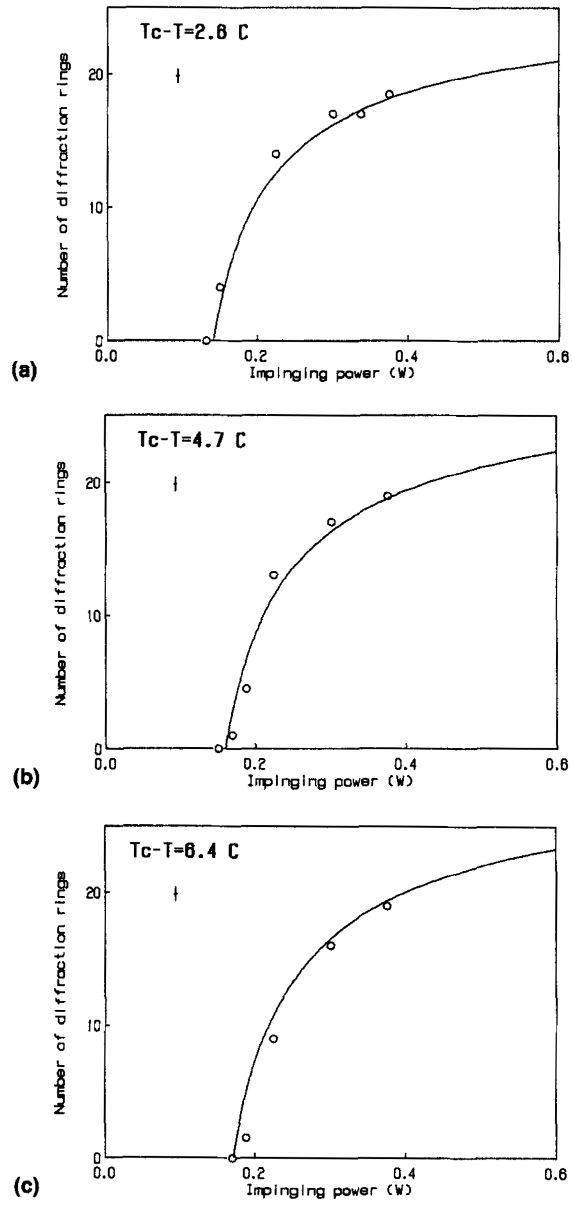


FIGURE 1 Comparison of experimental data with theoretical computed values of the number of diffraction rings vs. impinging power. The beam wavelength is 0.5145 μm and the $1/e^2$ radius is 107.5 μm. We used a homeotropic cell 115 μm thick filled with K18 by BDH in the nematic phase (clearing point temperature $T_c$ = 29°C), but at different temperatures. On the upper left corner of the figures is represented the experimental error. a) $T_c - T$ = 2.6°C: $\varepsilon_o = 2.42$, $\varepsilon_e = 2.82$, $K_1 = 3.65$ $pN$, $K_3 = 4.10$ $pN$; b) $T_c - T$ = 4.7°C: $\varepsilon_o = 2.43$, $\varepsilon_e = 2.85$, $K_1 = 4.40$ $pN$, $K_3 = 5.02$ $pN$; c) $T_c - T$ = 6.4°C: $\varepsilon_o = 2.43$, $\varepsilon_e = 2.88$, $K_1 = 4.91$ $pN$, $K_3 = 5.65$ $pN$.

and the normalized impinging power

$$P^* = \frac{P\,d^2}{\pi w^2} \tag{11}$$

to take off the dependence on the beam ($\lambda$, $w$, $P$) and cell ($d$) characteristics.

Using the known parameters of K18 we show in Figure 2 how the nonlinear behavior is affected by temperature variations, near the clearing point. As expected both the threshold and the saturation value decreases as the temperature approaches the clearing point $T_c$. It is interesting to notice that there is a value of the impinging power around which the phase shift is almost independent on temperature. This effect is clearly observable in the experimental data reported in Figure 1 where at three different temperatures we get the same nonlinear phase shift for an impinging power $P \approx 250$ mW.

To perform a more detailed analysis of the effects of the material parameters we first choose a "reasonable" set of parameters

$$\begin{aligned} K &= 1 - K_{11}/K_{33} = 0.15 \\ \varepsilon_m &= (\varepsilon_e + 2\,\varepsilon_o)/3 = 2.5 \\ K_3 &= 5.0\,pN \\ \varepsilon_a &= \varepsilon_e - \varepsilon_o = 0.5 \end{aligned} \tag{12}$$

and plot again the normalized phase shift vs. the normalized impinging power (full

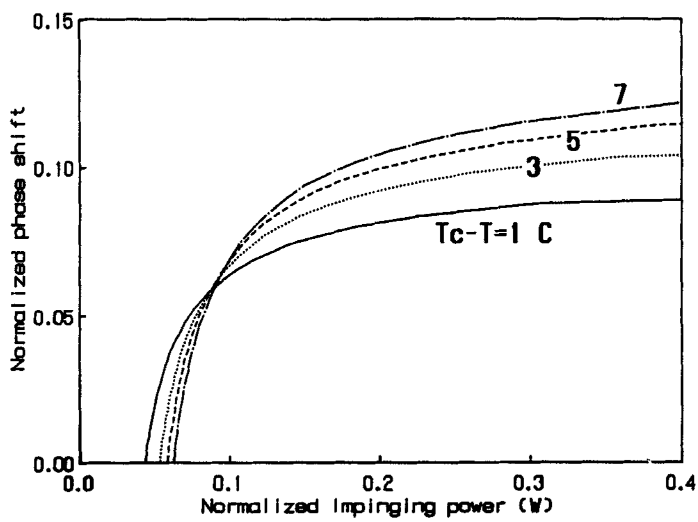


FIGURE 2 Dependence of the nonlinear behavior on the temperature for K18. Normalized phase shift is plotted vs. normalized impinging power.

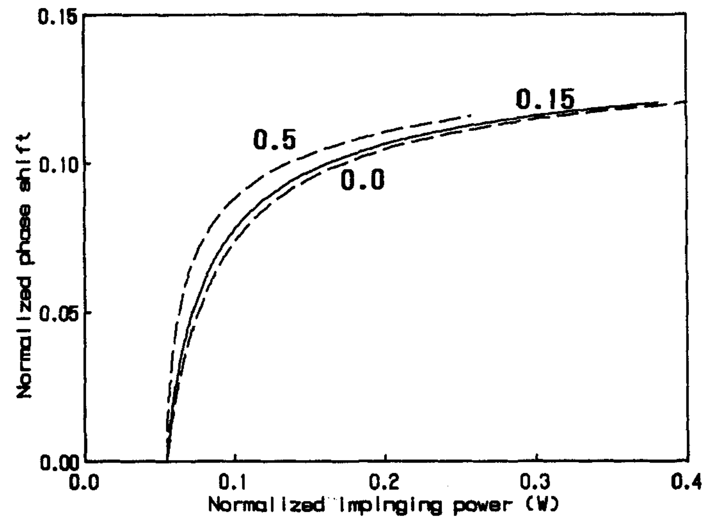


FIGURE 3 Normalized phase shift vs. normalized impinging power, with elastic anisotropy $K$ as parameter.

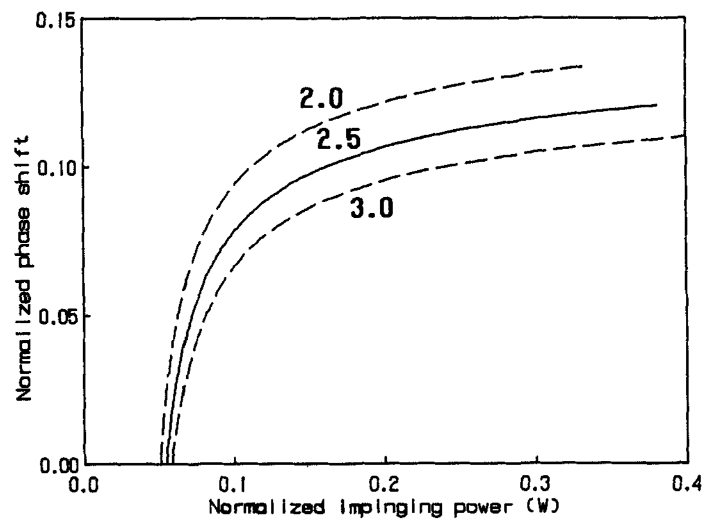


FIGURE 4 Normalized phase shift vs. normalized impinging power, with mean dielectric constant $\varepsilon_m$ as parameter.

line in Figures 3–6). Then we change by $\pm 20\%$ (except than for $K$) the value of one parameter at a time (dashed lines in Figures 3–6).

As can be seen in Figure 3 the nonlinear behavior is almost at all insensitive to the variation of the elastic anisotropy $K$. A variation of $\pm 20\%$ in $K$ does not produce any effect in this scale; a strong variation from $K = 0$ (i.e. $K_1 = K_3$) to $K = 0.5$ (i.e. $K_1 = 0.5\ K_3$) does not affect the threshold and the saturation value but slightly changes the slope of the curve. It is interesting to notice here the difference between the homeotropic and the hybrid aligned nematic (HAN) cell.

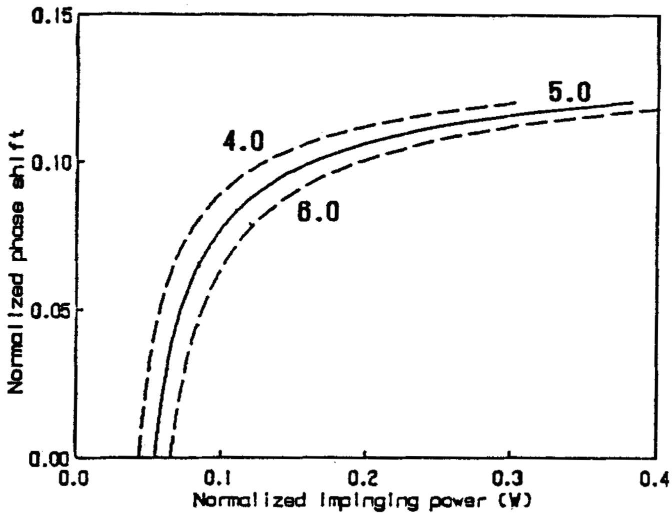


FIGURE 5 Normalized phase shift vs. normalized impinging power, with bend elastic constant $K_3$ as parameter.

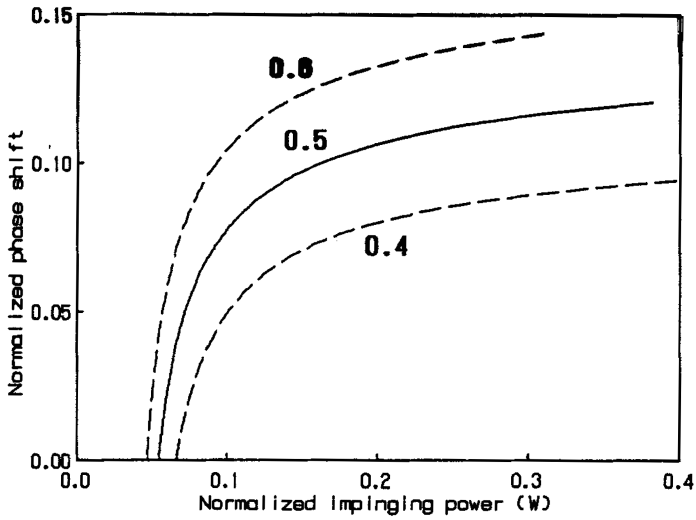


FIGURE 6 Normalized phase shift vs. normalized impinging power, with optical anisotropy $\varepsilon_a$ as parameter.

In the latter case the elastic anisotropy strongly affects the saturation value of the nonlinearity[7] because it strongly affects the initial orientation of the sample owing to the different boundary conditions on the two extremes of the cell.

The main effect of the variation of the average dielectric constant $\varepsilon_m$ (Figure 4) is a change of about 10% of the saturation value, while the threshold is only slightly affected.

A variation of the bend elastic constant $K_3$ (Figure 5) affects only the threshold value which is directly proportional to it (see Equation (9)), but does not affect the saturation value or the slope of the curve.

Finally Figure 6 shows that the most important parameter in determining the nonlinear optical properties coming from director reorientation is the optical anisotropy $\varepsilon_a$. As can be seen from the figure, by increasing $\varepsilon_a$ we get a change of the whole curve of the phase shift which decreases the threshold for the optical Freedericksz transition and produces a higher nonlinear response for each impinging power.

The former results allow us to make the following remarks above the influence of thermal effects on the optical reorientation in a homeotropic nematic sample. As far as we are dealing with the threshold behavior it is important to know the dependence of the optical anisotropy and of the bend elastic constant on the temperature; while then we are interested in the saturation behavior the main parameters to be taken into account are the dielectric constants (mainly the optical anisotropy).

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