Mol. Cryst. Liq. Cryst., 1990, Vol. 179, pp. 1–12
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

ANOMALOUS OPTICAL FREEDERICKSZ TRANSITION IN AN ABSORBING LIQUID CRYSTAL

I. JÁNOSSY
Central Research Institute for Physics; H-1525 Budapest, P.O.Box 49, HUNGARY

A.D. LLOYD and B.S. WHERRETT
Heriot-Watt University, Edinburgh, EH14 4AS, U.K.

Abstract Laser-induced reorientation was studied in absorbing nematic films. The optical Freedericksz threshold occured at an intensity level of 50 W/cm^2 in contrast to the normally observed few kW/cm^2 value for transparent layers. Thermomechanical coupling is considered as a possible explanation of the observed anomaly.

1. INTRODUCTION

In the last decade the optical field induced Freedericksz transition in nematics has been widely studied both theoretically and experimentally [1]. The basic mechanism of this transition in non-absorbing homeotropically aligned films is essentially the same for optical fields as for quasistatic electric ones. The interaction between the electric field and the induced electric polarization yields a torque with non-zero time average; this d.c. torque drives the reorientation of the nematic director. The threshold light power for reorientation in the case of normal incidence can be readily calculated taking into account this mechanism. The result of such calculations are in satisfactory agreement with experimental data[2].

In this paper we report on light-induced reorientation observed in a homeotropic absorbing nematic layer. Preliminary investigations were published earlier[3], here we present a more comprehensive study on the subject. In § 2 and 3 we present the experimental details. As described there a reorientation effect was found with a threshold power almost two orders of magnitude smaller than expected for the usual optical Freedericksz transition. The decisive role of absorption in the observed reorientation process became evident also from the fact that no effect could be detected in corresponding non-absorbing samples. Therefore the interpretation of the experimental findings requires a mechanism which is effective only in the presence of absorption.

In §4 we discuss one possibility: thermally induced reorientation originating from the coupling between the temperature field and the director deformation. The relevant coupling constant is estimated from the experimental data and is compared with theoretical predictions.

2. EXPERIMENTAL

The liquid crystal investigated was the commercial nematic mixture D82E63 developed at BDH for use in guest-host cells. This mixture is a solution of a blue dichroic antraquinone dye in the cyanobiphenyl host E63. This dye provides a strong absorption at 633 nm; for the ordinary ray at room temperature we measured $\alpha = 170\ cm^{-1}$. The clearing point of the mixture was found to be $83^{\circ}C$. This value is $4.5^{\circ}C$ lower than the date given by BDH for pure E63.

The liquid crystal was sandwiched between two glass substrates coated with transparent electrodes. Homeotropic alignment was ensured by treating the surfaces with octadecyldimethyl-(3-trimethoxysilyl-propil)ammonium cloride. The thickness of the nematic films varied between 20 and 40 μm for different samples.

In a typical experiment the beam from a He-Ne laser capable of 30 mW was focussed onto the sample at normal incidence. The illuminated region of the layer was projected onto a screen using microscope objectives. Care was taken to obtain the real image of the liquid crystal layer on the screen. This was confirmed by observing the edge of the spacer, dust particles or orientational defects. In some experiments a weak probe beam was used to test the changes within the illuminated area. The probe beam was observed by blocking the main beam for a short interval.

3. RESULTS

On increasing the input laser power reorientation of the nematic film was observed above a specific threshold. This reorientation was best seen with the help of the probe beam polarized at 45° with respect to the main beam and placing an analyzer between the sample and the screen. Below the threshold no light from the probe beam passed through the analyser. At the threshold (typically 1.5 mW) a bright spot appeared at the centre of the image which developed into a system of concentric bright rings as the input power increased. A significantly smaller contrast of the ring system was observed when the polarization of the probe beam was parallel or perpendicular to that of the main beam.

The above pattern can be interpreted as a Freedericksz type of deformation in the layer. The molecules are rotated within (or closely to) the polarization plane of the light beam. The director tilt is maximum at the centre of the beam and decreases gradually with increasing radial distance. The intensity maxima correspond to radial distances at which the phase difference between the ordinary and extraordinary ray is a half-integer multiple of π.

Further observations connected to this reorientation process were the following.

a.) The application of an external electric field normal to the boundaries increased the threshold input power (Fig. 1.). As it can be seen from the figure the threshold increased proportionally to the square of the applied field. This observation excludes the possibility that the ring pattern would arise from pure thermal lensing effects as this latter effect cannot be sensitive to a stabilizing electric field.

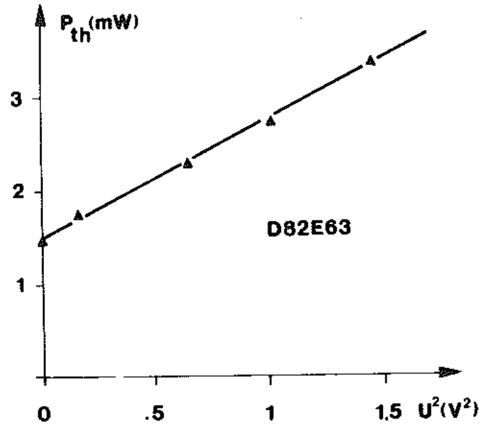

FIGURE 1. Threshold power as a function of applied voltage. Cell thickness and laser spot radius 40 μm.

b.) When the direction of the input polarization was rotated abruptly by 90^0 (taking care not to displace meanwhile the laser beam) the ring system first shrank then expanded again in a few seconds. No significant difference was found in the threshold for vertically and horizontally polarized beams. In addition we checked that the reorientation processes were very similar -at least qualitatively- when the sample was set both vertical and horizontal. (Precise quantitative comparison of the thresholds in the two cases has not yet been carried out). These latter circumstances indicate that onset of thermal convection was not the primary cause of the reorientation (see §4).

c.) Illumination by circularly polarized light generated self-oscillation in the polarization state of the transmitted beam (Fig.2.) The frequency of this self-oscillation increased linearly with the input power (Fig.3.). Assuming that the reorientation threshold corresponded to zero frequency we found that the threshold for circularly polarized light exceeded the threshold for linear polarization by a factor of 2.2 .

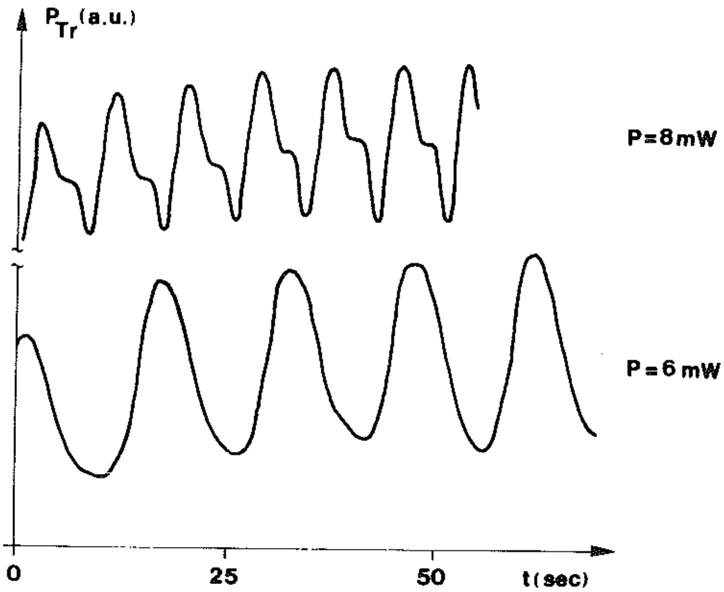

FIGURE 2. Transmission oscillations generated by circularly polarized inputs. The signal was detected behind a $\lambda/4$ plate and a polarizer.

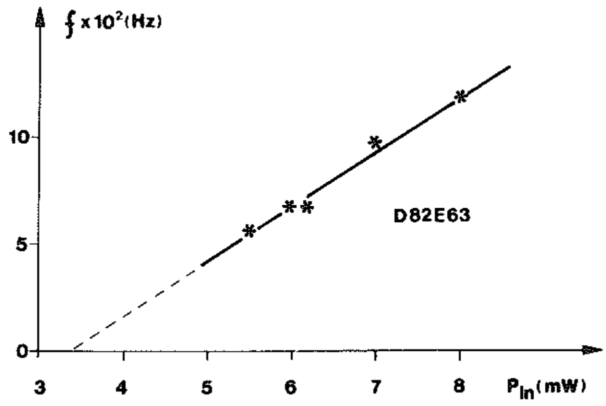

FIGURE 3. Frequency of oscillation as a function of input power.

On increasing the input power above the reorientation threshold the appearance of a sharp loop was observed at a second threshold. This loop was best seen in the direct image coming from the main beam, without using analyser. The loop-threshold increased also with increasing applied field. At high fields saturation of the threshold was found (Fig.4.). We assume that this phenomenon was connected to the formation of an isotropic droplet within the nematic film; the loop was the image of the nematic-isotropic interface.

Finally we note that the homeotropic orientation in our samples degraded rather easily under the influence of laser radiation, especially when the reorientation threshold was exceeded significantly. During the measurements care was taken to always choose well-oriented parts in the film.

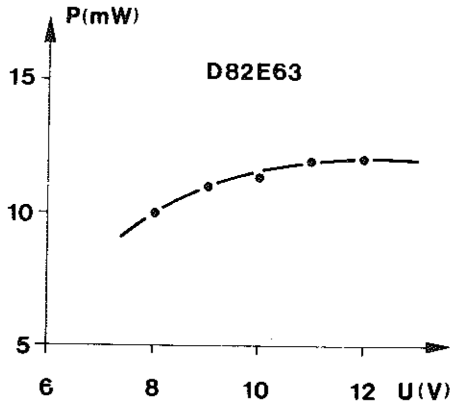

FIGURE 4. Threshold power for melting as a function of applied voltage.

4. DISCUSSION

The reorientation effect described in the previous paragraph shows some characteristic features of the optical Freedericksz transition such as dependence on external fields[4] or self-oscillation at circularly polarized input[5]. However the measured threshold power was anomalously low compared to the value expected for corresponding non-absorbing samples. As shown in[2] for a laser spot radius comparable to the film thickness the normal Freedericksz threshold is around 100 mW while in the present case we observed values below 2mW. This anomaly was directly proved by the fact that no reorientation was produced in pure E63 samples even when the highest aivable power was applied.

In view of the above fact one must search for a mechanism in which absorption plays an essential role. One possibility could be the generation of a flow due to the temperature gradient present in the sample. However in the case of thermal convection the vertical direction should play a distinguished role. In particular, the flow generated by buoyancy forces is essentially vertical which would produce a director tilt within a vertical plane. In contrast to this expectation we observed that the director tilted within the plane of polarization of the laser beam. Hence we believe that the reorientation was not caused by the onset of thermal convection.

We consider here another possibility, namely the direct coupling between the nematic director and the temperature field generated by the laser beam. The existence of this coupling (thermomechanical effect) is well-known in cholesterics[6,7]. It was shown theoretically[8,9] that coupling between a temperature gradient and a non-uniform director field might exist as well in the nematic phase. There can be both a static effect and also kinetic contributions arising from the heat flow. In the following we make a rough estimation of the coupling constant supposing that the reorientation described above is due to the thermomechanical effect.

The torque arising from the thermomechanical coupling includes appropriate linear combination of terms like[9]

$$\xi \, \partial n_i / \partial x_j \cdot \partial T / \partial x_k$$

where $\underline{n}$ is the director, ξ is a coupling coefficient. Assuming that at threshold this torque is balanced by the elastic torque

$$K\,\partial^2 n_i/\partial x_j \partial x_k$$

(K is a Frank elastic constant) we can make the following estimation for ξ. $\partial n_i/\partial x_j$ and $\partial^2 n_i/\partial x_j \partial x_k$ are in the order of θ/L and θ/L^2 resp. where θ is the tilt angle, L is the sample thickness or laser spot size. $\partial T/\partial x_k$ is in the order of $\Delta T/L$ where ΔT is the maximum temperature rise in the film. The balance of torques yields

$$\xi \sim K/\Delta T_{th}$$

ΔT_{th}, i.e. the maximum temperature rise at the reorientation threshold can be estimated for our experimental circumstances in the following way. As shown in the experimental part 12 mW was needed to melt the film when the reorientation was supressed by an external field. This corresponded to a temperature rise of $63^{o}C$ for D82E63. Supposing that the temperature rise in the homeotropic state is essentially proportional to the input power one obtains that at 1.6 mW (the threshold for reorientation in the absence of field)

$$\Delta T_{th} = 8.5^{o}C$$

Hence with $K=10^{-11}N$

$$\xi \sim 10^{-12} N/^{o}C$$

According to the theoretical estimation by Akopyan and Zeldovich $\xi \sim 10^{-11} N/^{o}C$. From an alternative point of view it can be predicted that $\xi \sim \partial K/\partial T$, which gives 10^{-13}-$10^{-12} N/^{o}C$. Hence our estimated experimental value of the coupling coefficient is more or less in agreement with the theoretical considerations. On the other hand it is not clear why the presumed thermomechanical coupling should lead to a Freedericksz type of deformation.

In conclusion, we observed in an absorbing nematic layer a new type of optical Freedericksz transition. The effect was systematically found in all samples investigated. Thermomechanical coupling was suggested as a possible source of the reorientation. As further possibilities one may consider changes in the surface anchoring properties due to the decomposition of the dye under the influence of illumination[3] or effects arising from radiation pressure. Further work is planned on the subject in order to clarify the proper mechanism leading to the phenomena described in the paper.

REFERENCES

1. N.V. Tabyrian and B.Y. Zeldovich Mol. Cryst. Liq. Cryst., 136,1 (1986) and references therein.
2. L. Csillag, I.Jánossy, V.F. Kitaeva, N. Kroó and N.N. Sobolev Mol. Cryst. Liq. Cryst., 84, 125 (1982)
3. I.Jánossy, M.R.Taghizadeh and E.Abraham in Optical Bistability III. (Springer-Verlag, 1986), p.160.
4. L. Csillag, N. Éber, I.Jánossy, V.F. Kitaeva, N. Kroó and N.N. Sobolev Mol. Cryst. Liq.

Cryst., 89, 287 (1982)
5. E. Santamato, D. Daino, M. Romagnoli, M. Settembre and Y.R. Shen Mol. Cryst. Liq. Cryst., 143, 89 (1987)
6. F. Leslie Proc. Roy. Soc., A307, 359 (1968)
7. N.Éber and I.Jánossy Mol. Cryst. Liq. Cryst. Lett., 72, 233 (1982) and 102, 311 (1984)
8. H. Pleiner and H. Brand J. Physique Lett., 41, L-491 (1980)
9. R.S. Akopyan and B.Y. Zeldovich Soviet Physics JETP, 60, 953 (1984)