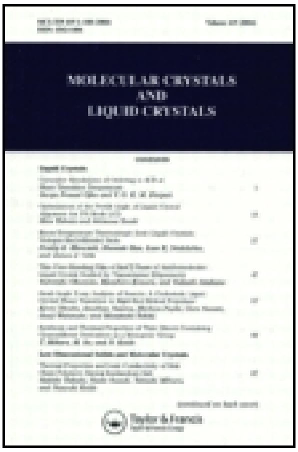

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Low-Power Optical Reorientation in Dyed Nematics

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Low-Power Optical Reorientation in Dyed Nematics

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The optical reorientation process in nematic guest-host mixtures is investigated. It is found that for certain dyes the optical Freedericksz threshold is two orders of magnitude lower as in the pure host. The observations can be interpreted in terms of an enhanced optical torque which increases with the dye concentration.

INTRODUCTION

In a recent publication[1] we called attention to the anomalous behaviour of a dyed nematic liquid crystal in optical fields. It was found that in homeotropic layers of a guest-host mixture the optical Freedericksz threshold is two orders of magnitude smaller at the He-Ne wavelength than expected for a corresponding transparent nematic. In Reference 1 thermomechanical coupling, i.e. the coupling between the laser-heating induced temperature gradients and the director field, was suggested tentatively as a possible source of the anomaly. However, the need of further investigations was emphasized.

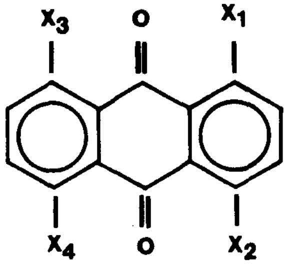
In the present paper we report on new experimental results in connection with the above phenomenon. The previous studies were extended in two respects. Firstly, we investigated the dependence of optical reorientation on the dye concentration and compared the effect of different dyes. We found that the reorientation process is not directly connected to the strength of absorption in the liquid crystal. In particular, different dyes had different influences on the reorientation, even when the absorption coefficient of the mixtures was the same. This fact reveals that, in contrast to our previous speculation, laser-heating cannot be responsible for the observed low-power reorientation.

Secondly, we studied the optical-field induced distortion of the layer at oblique incidence of the light beam. The observations at oblique incidence—together with the ones at normal incidence, reported in Reference 1—showed that there is a complete qualitative correspondence between the reorientation processes in transparent and dyed nematic films. Quantitatively, however, the input power level necessary to produce a given distortion decreases significantly in the presence of the dyes. This circumstance can be described formally with the help of a dye-induced torque which has the same form as the usual optical torque, except that the dielectric anisotropy (i.e. the difference between the squares of the refractive indices) is replaced by a new material factor. The new parameter characterizes the action of the dye; for a specific dye and concentration its value can be deduced from the Freedericksz threshold.

For the time being, there is no detailed microscopic model to explain the dye-induced enhancement of the optical torque. As a possibility, we mention the influence of the light-induced dipoles of the dye molecules. The electric field of these dipoles may be much stronger than the optical field, inducing therefore a reorientation at anomalously low input light levels.

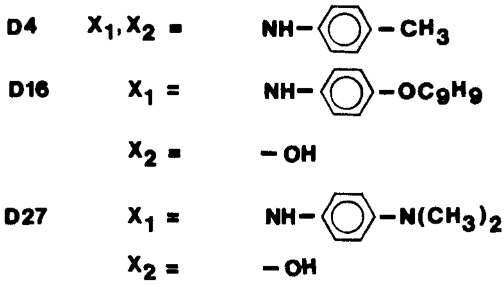
MATERIALS

We investigated the anthraquinone dyes 1,8-dihydroxy-4,5-diamino-2,7-di-isobutyl-AQ and 1,8-dihydroxy-4,5-diamino-2,7-di-iso-pentyl-AQ. Here AQ stands for anthraquinone. In the following we refer to these dyes as AQ1 and AQ2, respectively. In addition we carried out experiments with the dyes D4, D16 and D27 produced by BDH, which are also based on the general anthraquinone structure:



Scheme 1

For these dyes the X_3 and X_4 groups are missing; the groups X_1 and X_2 are as follows:



Scheme 2

As a host we used E63, which is mainly a mixture of biphenyls; its clearing point is at 87.5°C. In Table I we provide some data on the absorption coefficients for 633 nm.

In order to investigate the influence of the dyes on some material parameters of the nematic, we studied the d.c. electric field-induced Freedericksz transition in planar cells of pure E63 and E63 doped with 2% of AQ1. The capacitance of the cells and the optical phase difference between the ordinary and extraordinary rays were measured as a function of the applied voltage. These measurements allow for the determination of the static dielectric constants, the splay elastic constant and the birefringence, $n_e - n_o$. For E63 we found $\varepsilon_a^{static} = 10$, $K_1 = .7 \times 10^{-11}$ N and $n_e - n_o = .22$ (at $\lambda = .633$ μm). Regarding the dyed material, we found no change in the d.c. Freedericksz threshold; a decrease of 3% in ε_a^{static} and—within the precision of our measurements (≈10%)—no change in the birefringence.

MEASUREMENTS OF THE OPTICAL FREEDERICKSZ THRESHOLD

As well known, in homeotropic layers and at normal incidence of the laser beam, deformation of the director field takes place only above a threshold input power (Freedericksz threshold). The simplest way to detect the distortion is to observe the self-focusing of the illuminating beam. At the threshold the beam divergence behind the sample starts to increase and on raising the input power the familiar abberational ring pattern develops. From the number of rings the optical phase change at the centre of the laser spot can be estimated in a simple way and thus the light-induced birefringence can be calculated.[2,3]

In order to determine the threshold, it is sufficient to detect only the initial increase of the beam divergence. For this purpose, we passed the transmitted beam through an aperture. The ratio of the power behind the aperture and the input power was monitored as a function of the input power. At the threshold a sharp decrease of the ratio was observed as a consequence of the increase in the beam divergence.

The measurements were carried out on cells with thickness of 60 μm ± 2 μm. Two lenses with focal lengths of 135 mm and 70 mm were used, producing laser spot sizes of 31 μm and 17 μm resp. (1/e radii).

In Figure 1 the Freedericksz threshold is shown for the system AQ1-E63 as a function of the dye concentration. We choose to characterize the materials by the absorption coefficient $\alpha_\perp$. The highest $\alpha_\perp$ value on the curves corresponds to ap-

TABLE I

MATERIAL	$\alpha_\parallel$ (cm^{-1})	$\alpha_\perp$ (cm^{-1})
2% AQ1	2800	390
1% D4	1110	380
1% D16	720	140

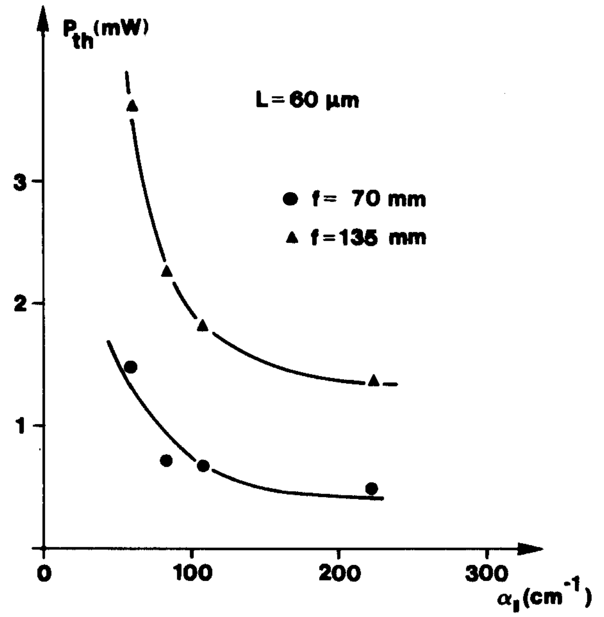


FIGURE 1 Threshold power in the AQ1-E63 system as a function of the absorption coefficient for two spot sizes.

proximately 1% of AQ1 in E63. For higher concentrations of AQ1, fast degradation of the homeotropic alignment under the influence of laser irradiation was observed and thus the threshold measurements were unreliable. In the diluted mixtures the degradation rate was very much reduced, but the bleaching of the dye was still evident and caused some uncertainty in the determination of the thresholds. Very slow scans led to increased threshold powers due to the bleaching during the scan. At faster scans the long response time of the nematic (few seconds) became a source of error. We estimate the error in the measured thresholds to be around 15%.

The dye AQ2 in E63 behaved in a similar way as the AQ1-E63 system. For example, with a concentration of 1% of AQ2, the Freedericksz threshold was 0.6 mW at a spot size of 17 μm.

On the other hand, with the other dyes optical Freedericksz transition could not be induced. On increasing the power no reorientation effect was observed until melting of the layer occurred. We note that one can easily distinguish between melting and reorientation in the nematic phase: melting gives rise to a characteristic far-field diffraction pattern and has a much shorter relaxation time than reorientation.

The above facts indicate that it is not simply the presence of absorption which causes the low-power reorientation in the dyed systems. E.g. D4 has almost the same $\alpha_\perp$ value as 2% AQ1 (see Table I) yet the reorientation effects are markedly different. The same statement holds when D16 is compared with the appropriate AQ1-E63 mixture that has the same $\alpha_\perp$ as D16.

In order to take further this point, we prepared a mixture of AQ1, D4 and E63 in which both the parallel and the perpendicular components of the absorption coefficients were approximately the same as in the 1% D16-E63 system. In Figure 2 the far-field diffraction patterns are shown for 4 mW input power. For the AQ1-D4-E63 mixture 4 mW is well above the Freedericksz threshold, while in the case of D16-E63 there is no sign of reorientation. From this fact it is evident that reorientation cannot originate from laser heating. Laser heating is determined by the value of the absorption coefficients which are close to each other in the two systems, yet the reorientation processes deviate significantly.

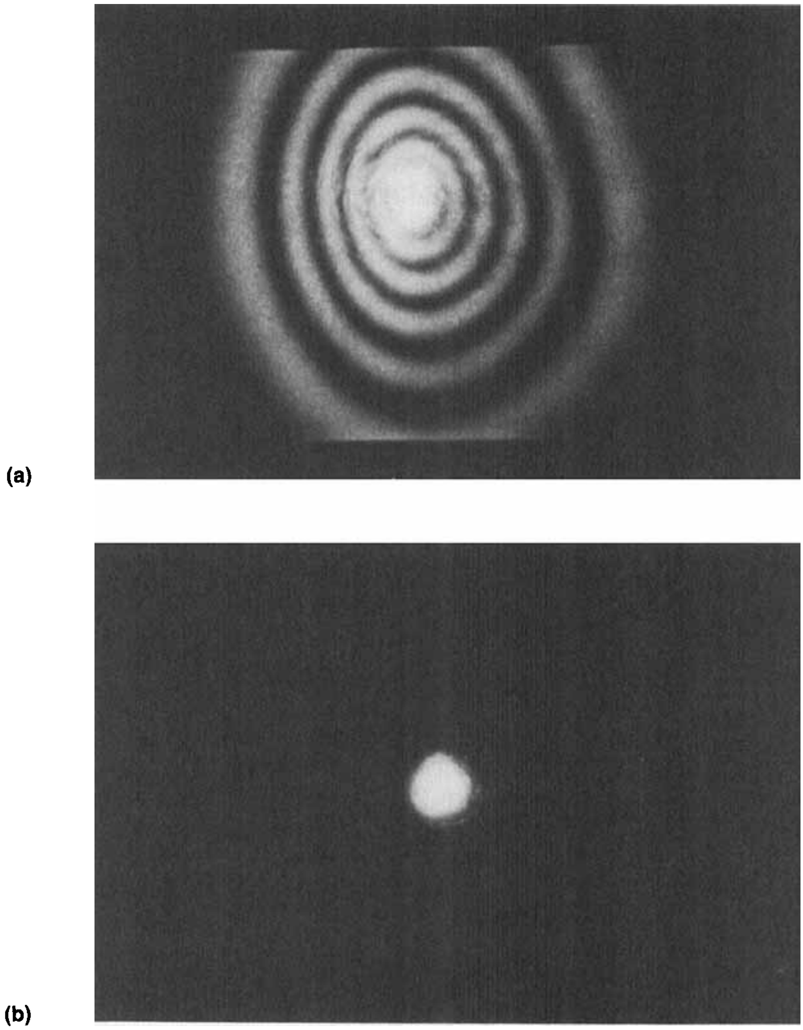


FIGURE 2 Far-field diffraction patterns for a; .375% AQ1 and .25% D4 in E63 b; 1% D16 in E63. Normal incidence, input power 4 mW, spot size 17 μm.

REORIENTATION AT OBLIQUE INCIDENCE

At normal incidence of the laser beam the reorientation in the AQ1-E63 and AQ2-E63 layers takes place in a similar way as in transparent nematics. The reorientation of the director starts at a threshold input power (optical Freedericksz transition). For linear polarization, above the threshold the nematic molecules tilt within or near to the polarization plane of the light beam. For circularly polarized input, the threshold is about two times larger than for linear input polarization; furthermore above the threshold continuous precession of the director around the normal direction takes place. All these effects are well-known for non-absorbing nematics.[2,4,5]

In further experiments we proved that this similarity holds for oblique incidence as well. We found that, in accordance with the corresponding observations in transparent layers, the reorientation process is very different for the extraordinary and the ordinary rays. For *e*-polarization (i.e. polarization in the plane of incidence of the light beam) reorientation occurred without threshold. For ordinary input (polarization normal to the plane of incidence) the threshold power increased drastically as the angle of incidence was increased. At angles larger than 10° the threshold became so large that melting occurred before reorientation could have started. The observations are illustrated in Figure 3, showing some characteristic far-field patterns.

DISCUSSION

Optical reorientation in transparent nematics can be interpreted by considering the balance between the torque exerted by the light field and the elastic torque, arising from the distorsion of the director pattern. The "optical" torque is given as

$$\Gamma_{opt} = \mathbf{P} \times \mathbf{E} = \varepsilon_0 \varepsilon_a (\mathbf{nE})(\mathbf{n} \times \mathbf{E}) \tag{1}$$

with

$$\varepsilon_a = n_e^2 - n_o^2.$$

Here **P** is the polarization, **E** is the electric field of the light beam, **n** is the director. The elastic torque, in the one-constant approximation is

$$\Gamma_{elastic} = K\,\mathbf{n} \times \nabla^2 \mathbf{n} \tag{2}$$

where K is a Frank elastic constant. As shown by previous authors, combining these expressions for the torques with a careful consideration of light propagation allows all aspects of optical reorientation to be accounted for.

The optical Freedericksz threshold in transparent layers, calculated according to the above consideration, is in good agreement with the measured values.[6] For the experimental circumstances used in the present experiments, this threshold is well

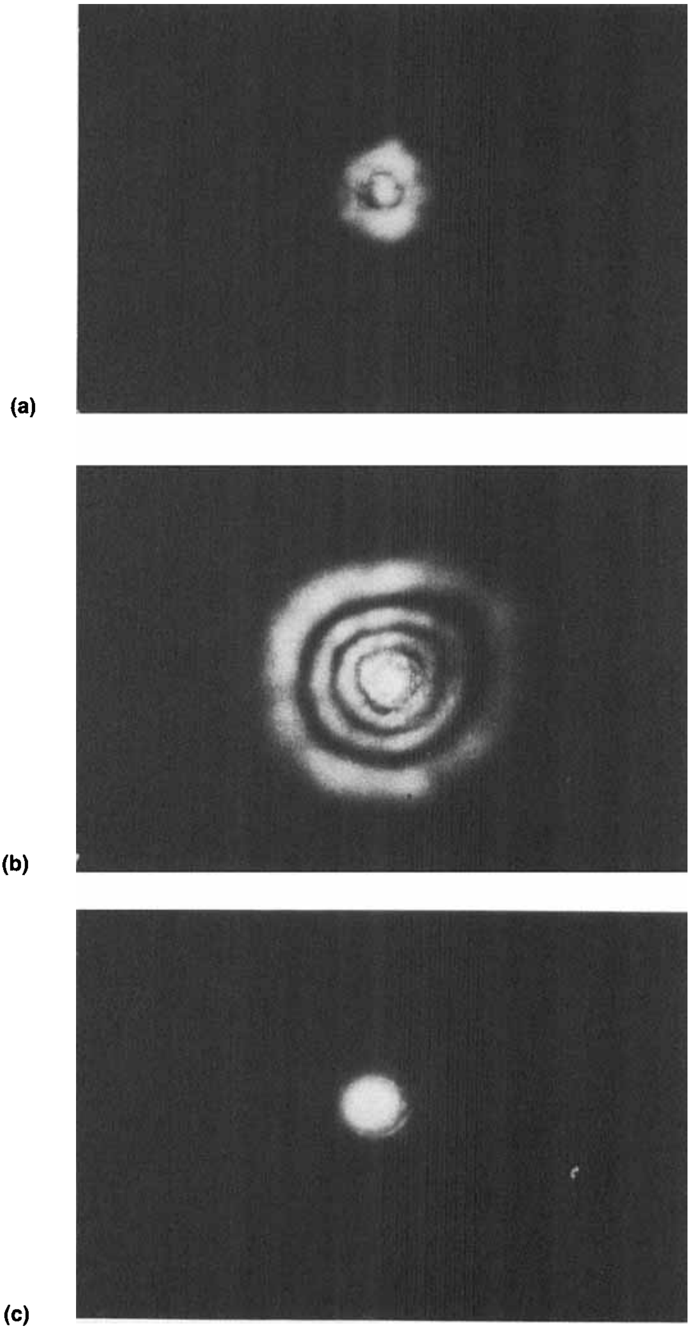


FIGURE 3 Far-field diffraction patterns for .375% AQ1 and .25% D4 in E63. Angle of incidence 45°. a; *e*-polarization, input power 0.5 mW. b; *e*-polarization, input power 2 mW. c; *o*-polarization, input power 4 mW.

above 100 mW. As we observed, however, in the presence of certain dyes (AQ1, AQ2) the Freedericksz threshold drops to power levels around 1 mW.

This dramatic reduction of the optical Freedericksz threshold power, caused by the dyes, cannot be explained by the change of the relevant material parameters (ε_a, K). These parameters would have to change by orders of magnitude to produce the observed effects. In reality, we found that these constants deviate only by a few percent in the dyed materials from their values in the pure host.

In our opinion, the very low optical Freedericksz thresholds indicate that in the presence of these dyes an additional light-induced torque is superposed on the normal optical torque. On the basis of the similarity between the qualitative features in the reorientation processes in dyed and transparent nematics, we propose that this extra torque, Γ_{dye}, (or rather its time average) is proportional to the time average of the optical torque:

$$\langle \Gamma_{dye} \rangle = \eta \langle \Gamma_{opt} \rangle \tag{3}$$

where the coefficient η is a characteristic of the dye.

With the above phenomenological form of Γ_{dye}, one can account for a number of experimental observations (threshold, oscillation at circularly polarized input, behaviour at oblique incidence of the light beam) in the same way as in the case of non-absorbing nematics. The intensity, necessary to produce a given distortion in the layer is reduced, however, by the factor $1 + \eta/\varepsilon_a$. The decrease of the Freedericksz threshold by two orders of magnitude in the cases of AQ1 and AQ2 indicates that η is at least 100 times larger than the birefringence for these materials.

On a microscopic level, the enhancement of the optical torque can be a result of the build-up of an internal electric field within the illuminated volume. This kind of internal field might originate from the dipole moments of the excited dye molecules; its strength would depend on the properties of the excited state of the particular dye, like lifetime or magnitude and direction of the dipole moment. The details of such a microscopic model has to be worked out in the future.

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