Notes on the paper I. Jánossy and T. Kósa "Laser-induced effects in dyed nematics"

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The paper contains a number of misprints in the equations from Eq. 3 to 7. The correct text is as follows:

"The temperature rise can be calculated by using Hankel-transformation. According to this method, the temperature is expressed as a function of zero-order Bessel functions, J_0:

$$T(r,z) = \int_0^\infty \lambda \tilde{T}(\lambda, z) J_0(\lambda r) d\lambda. \tag{3}$$

Inserting Eq. 3 into Eq. 2, one obtains for $\tilde{T}(\lambda, z)$

$$-\lambda^2 \kappa_\perp \tilde{T}(\lambda, z) + \kappa_\| \frac{\partial^2 \tilde{T}}{\partial z^2} = -\frac{P\alpha_\perp}{2\pi} e^{-\alpha_\perp z} e^{-\lambda^2 w^2/4} \tag{4}$$

In deriving Eq. 4, we made use of the relation

$$e^{-r^2/w^2} = \frac{w^2}{2} \int_0^\infty \lambda e^{-\lambda^2 w^2/4} J_0(\lambda r) d\lambda.$$

The solution of Eq. 4 is

$$\tilde{T}(\lambda, z) = \frac{P\alpha_\perp}{2\pi\kappa_\perp} \frac{e^{-\lambda^2 w^2/4}}{\lambda^2 - (\alpha_\perp \xi)^2} (e^{-\alpha_\perp z} + F_1 e^{\lambda z/\xi} + F_2 e^{-\lambda z/\xi}) \tag{5}$$

with $\xi = (\kappa_\| / \kappa_\perp)^{1/2}$. The F_1 and F_2 functions can be found from the boundary conditions, which require for arbitrary λ

$$\left. \frac{\tilde{T}}{\partial \tilde{T}/\partial z} \right|_{z=0} = - \left. \frac{\tilde{T}}{\partial \tilde{T}/\partial z} \right|_{z=L} = \frac{1}{\lambda} \frac{\kappa_\|}{\kappa_s}$$

(κ_s is the heat conductivity of the substrates.) Explicitly, F_1 and F_2 are the solutions of the linear equations

$$(1+k)e^{\lambda L/\xi} F_1 + (1-k)e^{-\lambda L/\xi} F_2 = -e^{-\alpha_\perp L}(1 - \alpha_\perp \xi k/\lambda) \tag{6}$$

$$(1-k)F_1 + (1+k)F_2 = -(1 + \alpha_\perp \xi k/\lambda) \tag{7}$$

where the notation $k = \sqrt{\kappa_\| \kappa_\perp} / \kappa_s$ was introduced."

Note on the evaluation of the integral in Eq. 3:

In Eq. 3 the integrand contains a singularity at $\lambda = \lambda_0 = \alpha_\perp \xi$. The integral, nevertheless, is not singular. It should be evaluated as

$$T(r,z) = \lim \varepsilon \to 0 \quad \left(\int_0^{\lambda_0 - \varepsilon} \lambda \tilde{T} J_0(\lambda r) d\lambda + \int_{\lambda_0 + \varepsilon}^\infty \lambda \tilde{T} J_0(\lambda r) d\lambda \right).$$

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LASER-INDUCED EFFECTS IN DYED NEMATICS

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Abstract. The interaction of a light beam with dye-doped nematic liquid crystals is discussed. We investigate both theoretically and experimentally the laser-induced melting of homeotropically aligned nematic films.

INTRODUCTION

In the present paper we discuss various laser-induced effects in dye-doped nematic liquid crystals. In the first part we consider pure thermal effects arising from the absorption of the light beam. We derive a simple estimation for the threshold input light power, at which the melting of a homeotropically aligned nematic starts. The corresponding experimental data are in reasonable agreement with the calculated values. We compare the melting threshold to the optical Freedericksz threshold and point out some observed anomalies.

In the second part of the paper, we investigate the behaviour of the liquid-crystalline film above the melting threshold. We describe experimental studies carried out with two dichroic dyes. With one of the dyes, we observed the expected phase-transition optical bistability.[1,2,3] With the other dye, oscillations between the isotropic and nematic phases were detected at the melting threshold. The latter phenomenon is similar to that observed recently by Simoni *et al.*[4] We suggest that the oscillations are connected to mass-diffusion within the illuminated area.

LASER-INDUCED MELTING IN A HOMEOTROPIC NEMATIC FILM

We consider a homeotropic nematic layer, sandwiched between two glass plates. A Gaussian laser beam penetrates trough the film in normal direction, i.e. along the z axis. The intensity at a point with r, z coordinates is

$$I(r,z) = \frac{P}{\pi w^2} e^{-r^2/w^2} e^{-\alpha_\perp z}. \tag{1}$$

Here P is the input power, w is the laser spot radius and $\alpha_\perp$ is the perpendicular component of the absorption coefficient.

The steady-state heat conductivity equation in the film is

$$\kappa_\perp\left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r}\right) + \kappa_\parallel \frac{\partial^2 T}{\partial z^2} = -\alpha_\perp I, \tag{2}$$

where $\kappa_\perp$ and $\kappa_\parallel$ are the perpendicular and parallel components of the heat conductivity tensor. We neglect the temperature dependence both of the heat conductivities and of the absorption coefficient. The usual boundary conditions are assumed; the temperature and the normal component of the heat flow are supposed to be continuous at the substrate-nematic interfaces ($z = 0$ and $z = L$).

The temperature rise can be calculated by using Hankel-transformation.[5,6] According to this method, the temperature is expressed as a function of first-order Bessel functions, J_0 :

$$T(r,z) = \int_0^\infty \lambda \tilde{T}(\lambda, z) J_0(\lambda r) d\lambda \tag{3}$$

Inserting Eq.3 into Eq.2 one obtains for $\tilde{T}(\lambda, z)$

$$-\lambda^2 \kappa_\perp \tilde{T}(\lambda, z) + \alpha_\perp^2 \kappa_\parallel \frac{\partial^2 \tilde{T}}{\partial z^2} = -\frac{P\alpha_\perp}{2\pi} e^{-\alpha_\perp z} e^{-\lambda^2 w^2/4} \tag{4}$$

In deriving Eq.4, we made use of the relation

$$e^{-r^2/w^2} = \frac{w^2}{2} \int_0^\infty \lambda e^{-\lambda^2 w^2/4} J_0(\lambda r) d\lambda.$$

The solution of Eq.4 is

$$\tilde{T}(\lambda, z) = \frac{P\alpha_\perp}{2\pi\kappa_\perp} \frac{e^{-\lambda^2 w^2/4}}{\lambda^2 - (\alpha_\perp \xi)^2} (e^{-\alpha_\perp z} + F_1 e^{\lambda z/\xi} + F_2 e^{-\lambda z/\xi}) \tag{5}$$

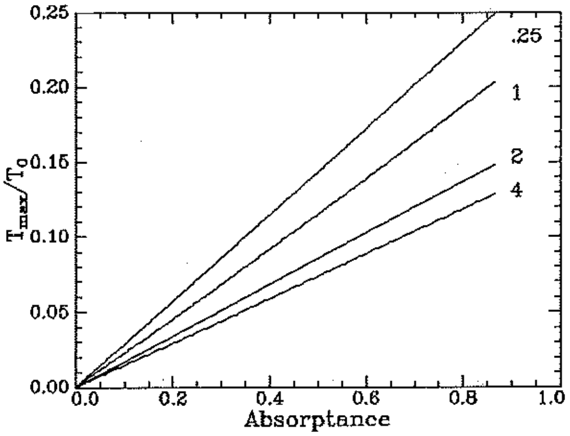

Figure 1: The maximum temperature rise as a function of the absorptance, for different w/L values. $\kappa_{\|}/\kappa_{\perp} = 2$; $\kappa_{\|}/\kappa_{s} = .27$.

with $\xi = (\kappa_{\|}/\kappa_{\perp})^{1/2}$. The F_1 and F_2 functions can be found from the boundary conditions, which require for arbitrary λ

$$\left.\frac{\tilde{T}}{\partial\tilde{T}/\partial z}\right|_{z=0} = -\left.\frac{\tilde{T}}{\partial\tilde{T}/\partial z}\right|_{z=L} = \kappa_{\|}/\kappa_{s}$$

(κ_s is the heat conductivity of the substrates.) Explicitly, F_1 and F_2 are the solutions of the linear equations

$$(1+k)e^{\lambda L/\xi}F_1 + (1-k)e^{-\lambda L/\xi}F_2 = e^{-\alpha_{\perp}L}(1 - \alpha_{\perp}\xi k/\lambda) \tag{6}$$

$$(1-k)F_1 + (1+k)F_2 = -1 + \alpha_{\perp}\xi k/\lambda) \tag{7}$$

where the notation $k = \sqrt{\kappa_{\|}\kappa_{\perp}}/\kappa_s$ was introduced.

With the help of relations 3,5,6 and 7 the temperature rise can be readily calculated in any point of the film. In Fig.1., we show the maximum temperature rise in the layer, in units of the quantity $T_0 = P/2\pi\kappa_{\perp}w$. In the calculations we set $\kappa_{\|}/\kappa_{\perp} = 2$ and $\kappa_{\|}/\kappa_{s} = .27$, which are typical numbers for nematics and float glass substrates. On the abscissa the absorptance of the film is plotted, which is related to the absorption coefficient as $A = 1 - e^{-\alpha_{\perp}L}$.

As it can be seen from the figure, T_m is almost proportional to the absorptance. Hence, in a good approximation

$$T_m \approx T_0 A f(w/L) = \frac{PA}{2\pi\kappa_{\perp}w} f(w/L). \tag{8}$$

f is plotted as a function of its argument in Fig.2. We note that in the limit of

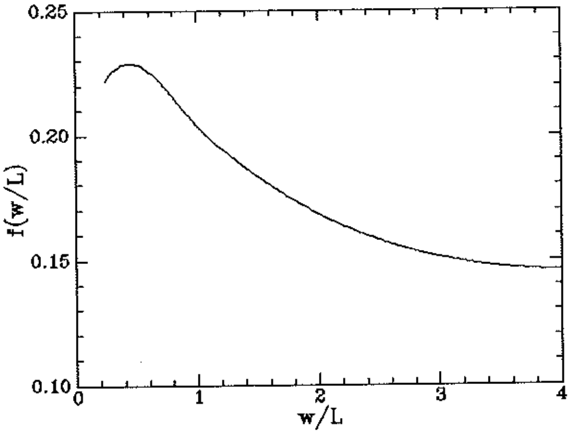

Figure 2: The $f(w/L)$ function. The heat conductivity parameters are the same as in Fig.1.

very large spot sizes, $w/L \gg 1$, the relation

$$T_m = \frac{PA}{4\sqrt{\pi}\kappa_s w}$$

holds.[7] Hence in this limit $f \rightarrow \frac{\sqrt{\pi}}{2}\kappa_\perp/\kappa_s$.

With the help of Eq.8, a simple estimation can be given for the melting threshold power, i.e. the input power at which the melting of the nematic layer starts. At this power

$$T_m = T_{NI} - T_{ambient}$$

where T_{NI} denotes the nematic-isotropic phase transition temperature. Thus, from Eq.8 we obtain

$$P_{th} \approx \frac{2\pi\kappa_\perp w(T_{NI} - T_{ambient})}{Af(w/L)} \tag{9}$$

In order to check the validity of the above relation, we carried out experiments with two dyes. We utilezed the anthraquinone dyes D16 and D102, supplied by BDH. 2% D16 was dissolved in 5CB. The nematic-isotropic transition was not modified by this dye; it took place at the same temperature as for the pure 5CB (35.5^{0}C). The dye D102 is available only in the cyanobiphenyl host E63. We mixed the commercial D102E63 system with 5CB in a ratio 1:3. The melting of this mixture started at 47^{0}C, but nematic droplets were observable up to 50^{0}C.

The measurements were performed with a beam from a He-Ne laser. The input power on the sample was regulated with the help of an electro-optic

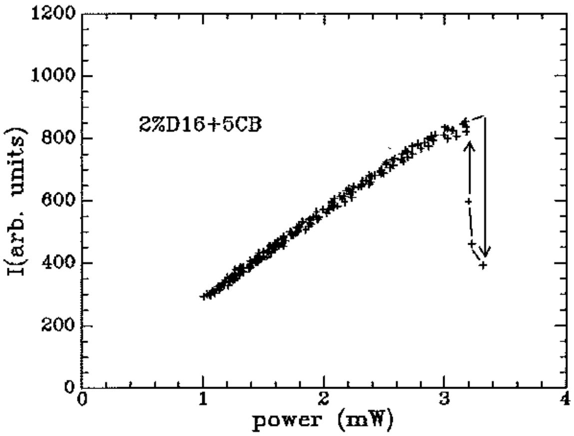

Figure 3: The transmitted signal as a function of the input power. $A = .332, w = 12\mu\text{m}$, $L = 10\mu\text{m}$.

modulator and a polarizer; the voltage on the modulator was controlled by a personal computer. The absorption coefficient for the D16 and D102 doped material was 332 cm^{-1} and 155 cm^{-1} respectively. $10\mu\text{m}$ thick cells were used. A strong electric field (10 V) was applied in order to prevent any reorientation effect. The spot radius of the laser beam in the layer was $12\mu\text{m}$. The ambient temperature during the experiments was 21^0C.

The melting of the layer was detected by observing the far-field diffraction pattern of the laser beam. The formation of an isotropic droplet in the layer was accompanied by the development of a characteristic ring system behind the sample. We measured the light intensity at the center of the diffraction pattern as a function of the input power; at melting an abrupt decrease of the signal was detected (Fig.3.).

The melting thresholds were 3.2 mW for the D16-5CB mixture and 13.5 mW for the D102E63-5CB system. Assuming $\kappa_\perp = 1.5 \times 10^{-3}\text{W}/^0\text{C cm}$, taking $w = 12\mu\text{m}$ and $w/L = 1.2$, the corresponding theoretical values from Eq.9 are 2.9 mW and 10.4 mW. These data are in reasonable agreement with the measured values; the deviations may arise from the uncertainty of the precise values of the heat conductivities and because of neglecting the temperature dependence of the material parameters.

It is interesting to compare the melting threshold with the threshold of the optical Freedericksz transition. For transparent layers, the Freedericksz threshold can be written as[8]

$$P_{F_r} = \frac{c\pi^3 K}{n_e^2 - n_o^2}\frac{n_e^2}{n_o}\left(\frac{w}{L}\right)^2 g(w/L) \qquad (10)$$

where n_e and n_o are the extraordinary and ordinary refractive indices resp., K is an elastic constant and c is the light velocity. The geometrical factor $g(w/L)$ gives the correction for the finite beam size; it approaches unity for very large spot sizes.

By comparing Eqs.9 and 10, one can calculate a critical absorptance, at which the two thresholds become equal. E.g. with $w/L = .5$, $T_{NI} - T_{ambient} = 50^0\mathrm{C}$ and $w = 10\mu\mathrm{m}$, furthermore supposing $K = 10^{-11}\mathrm{N}$, $n_e = 1.7$ and $n_o = 1.5$, one obtains $A_{cr} = 2 \times 10^{-2}$.

From the above example, it appears that it could be difficult to observe optical Freedericksz transition in films with significant absorptance. However, we found that in the case of certain dyes (D82 and D102), the optical Freedericksz threshold decreases drastically as the dye is added to the material.[9] With these dyes it is possible to induce optical reorientation well below the melting threshold even for large absorption. A detailed account of this effect is given in another publication.[10]

PHASE-TRANSITION BISTABILITY AND RELATED OSCILLATIONS

The idea of phase-transition optical bistability is illustrated in Fig.4. In a homeotropic sample, below the melting threshold the absorption coefficient for the laser beam is $\alpha_\perp$. Above the threshold, an isotropic droplet is formed in the film in which the absorption coefficient is α_i. For most dichroic dyes $\alpha_i > \alpha_\perp$, thus the heat production in the isotropic phase is higher than in the nematic state. The extra heating in the isotropic part leads to a further expansion of the melted droplet which, in turn, produces more heat. This positive feedback may lead to a runaway effect and to related optical bistability. A detailed theoretical discussion of this effect will be presented elsewhere. Here we describe observations with the dyes D16 and D102.

The experimental conditions were the same as given in the preceding section. The ratio $\alpha_i/\alpha_\perp$ was 1.90 and 2.74 for the D16 an D102 doped materials respectively. These numbers were deduced from transmission measurments using the same samples and the same He-Ne laser as in the melting experiments. In the case of the D16 sample, the runaway effect was observed, as expected, although the corresponding bistable loop was very narrow (Fig.3.). The bistable region was between 3.2 and 3.35 mW, as indicated by the arrows in the figure. We note that in spite of the narrow loop, it was possible to stabilize both branches of the transmission curve for several seconds (while the swithing time from one branch to the other was around a milisecond).

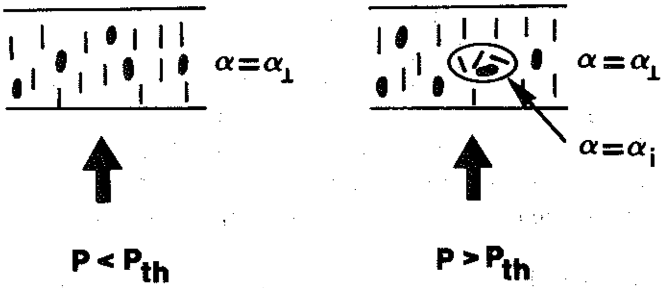

Figure 4: Schematic representation of the phase-transition bistability

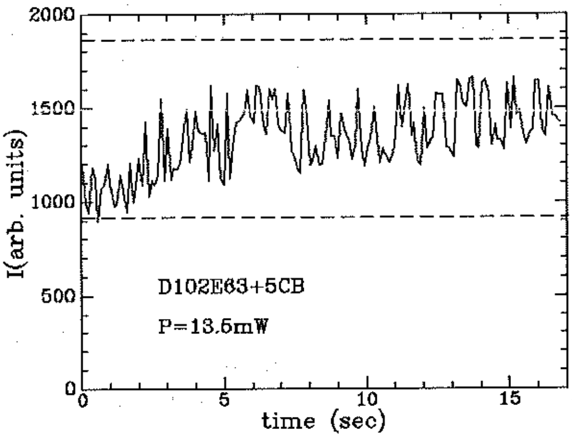

Figure 5: Time dependence of the transmitted signal. The upper and lower dashed lines represent the signal in the nematic state and after the first switch-off respectively.

For the D102-doped material, a wider bistable loop had been expected because of the higher $\alpha_i/\alpha_\perp$ ratio in this case. In contrast to this supposition, no bistable loop could be observed at all. Instead, oscillations in the far-field diffraction pattern was seen which corresponded to strong fluctuations in the size of the isotropic droplet (Fig.5.). The oscillation phenomenon was seen only in a narrow window of the input power, ranging from the melting threshold to a power approximately 5% above this threshold. (The curve shown in the figure was taken just above the melting threshold.) In a preliminary investigation it was found that at larger laser spot sizes the oscillations are less pronounced.

A similar phenomenon was reported recently by Simoni *et al.* in a more complicated geometry.[4] A comparison of our observations with theirs indicates that the geometry of the cell is not relevant in the development of the oscillations, while the specific guest-host mixture chosen for the experiment plays an important role in it. Simoni *et al.* suggested that either the occurrence of light diffraction by the droplet, or heat conduction in longitudinal direction is responsible for the oscillations. As an alternative possibility, we propose that the oscillations are caused by the interplay of a fast thermal process and a much slower mass-diffusion process. Our proposition is based on the following circumstances.

The system in which the phenomenon was found is a multi-component mixture. The host material itself is composed of a number of substances and, as observed experimentally, when heated uniformly it exhibits an unusually wide temperature range (few degrees) in which the isotropic and the nematic phases coexist. This fact implies a large difference between the chemical potentials of the components in the isotropic and the nematic states. Hence, when an isotropic droplet is created by laser heating, a strong diffusion process through the interface is expected to take place, which alters the composition of the material in the vicinity of the irradiated spot. This change in the composition, in turn, modifies the local clearing point and may cause the disappearence of the isotropic region.

Although the mathematical details of the above model has not yet been worked out, the oscillation period can be estimated in a simple way. As thermal diffusion is much faster than mass diffusion, it is the latter process which determines the rate of oscillations. The "lifetime" of the isotropic droplet is comparable to the time under which a molecule diffuses from the center to the boundary of the melted region. It can be estimated as

$$\tau \approx r_i^2/D \tag{11}$$

where r_i is the radius of the melted region and D is the diffusion constant. Taking $r_i = 10\mu\text{m}$ and $D = 5 \times 10^{-7}\text{cm}^2/\text{sec}$, one obtains $\tau = .5$ sec, which is of the same order of magnitude as the experimental value (Fig.5.).

In conclusion, in this paper we demonstrated the complex nature of the interaction between laser radiation and dye-doped nematics. Besides of pure thermal effects, other processes must be considered also in order to interpret the observed phenomena in these systems.

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