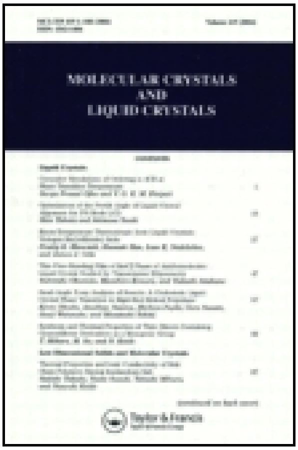

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Dynamic Light Scattering at Optical-Field-Induced Freedericksz Transition in Nematic Liquid Crystal

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Dynamic Light Scattering at Optical-Field-Induced Freedericksz Transition in Nematic Liquid Crystal

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The collective orientational fluctuations near optical-field-induced Freedericksz transition in a homeotropically aligned cell of the nematic liquid crystal 5CB were studied by dynamic light scattering. The beam size of the transition inducing Gaussian shape laser beam was considerably smaller than the thickness of the cell. A softening of the orientational fluctuation mode corresponding to the transverse inhomogeneity of the optical field was detected in a region which extended from about 70 % of the threshold intensity up to the threshold intensity. Measurements above the threshold intensity showed that at large scattering angles the time response of the fluctuations is very close to the usual exponential behaviour with a quadratic dispersion of the inverse relaxation time. At small scattering angles in addition a very gradual "tail" appears at long correlation times, signifying the instability of the perturbed structure.

Keywords: optical-field-induced Freedericksz transition; orientational fluctuations; dynamic light scattering

INTRODUCTION

The optical-field-induced Freedericksz transition (OFT) in nematic liquid crystals[1] is due to molecular director reorientation induced by the light field above the threshold. A variety of interesting nonlinear phenomena occurs due to a pronounced exchange of angular momentum and energy between the optical field and the nematic director field[3, 4, 6]. Characteristic of the OFT is also the inhomogeneous response of a nematic liquid crystal due to the transverse spatial intensity distribution of the focused laser light. This leads to beam size dependent critical intensity, and the well known prominent optical self focusing and self phase modulation[8, 9, 10].

The occurrence of the Freedericksz transition is generally interpreted by the slowing down of some critical thermal fluctuation mode of the director field which "freezes" at a specific threshold magnitude E_{tr} of the externally applied field. In homogeneous external fields the spatial profile of the critical mode is sinusoidal and the threshold field E_{tr} is determined, besides the material properties, predominantly by the thickness L of the liquid crystal cell[2] . The analysis of light scattering noise from variously aligned samples have shown that in homogeneous fields the relaxation time and the square mean value of the amplitude of the critical fluctuation are increasing with the increasing field as $(E_{tr}^2 - E^2)^{-1}$[11, 12].

In OFT the profile of the critical mode is more complex than in static fields, as it depends also on the spatial properties of the optical beam. The threshold optical intensity I_{tr} is significantly affected by the size of the beam waist w_0[7]. The evidence of strong influence of the transversal beam profile on the form of the critical mode and the threshold intensity I_{tr} stimulated us to investigate the dynamics of the thermal director fluctuations in the vicinity of the OFT by dynamic light scattering technique (DLS). The main goal of our experiment was to determine the role played by finite beam size in the behaviour of the critical fluctuation mode, which becomes soft near the OFT. In addition we investigated also the characteristics of the fluctuations in strongly perturbed orientational structure which forms when the input intensity I is above the threshold value I_{tr}.

EXPERIMENT

A homeotropically aligned sample of nematic liquid crystal 4-pentyl-4'-

cyanobiphenyl (5CB, Merck Ltd.) was prepared with two silane (DMOAP) covered glass plates. The thickness of the sample was 120 μm. The sample was mounted on a rotation stage and put in the center of a conventional DLS setup. In the setup the two CW laser beams were used simultaneously: the molecular reorientation inducing Ar ion laser beam (pump beam) with λ = 514.5 nm and input power up to 200 mW, and a weaker He-Ne laser beam (probe beam) with λ = 632.8 nm and input power of 5 mW (Fig. 1). The beams were focused onto the sample by a lens of f = 100 mm which resulted in a beam waist of $w_0 \approx 20\ \mu m$ for the pump beam and about $w_0' \approx 15\ \mu m$ for the probe beam. The sample was positioned into the waist by slowly translating it along the beam and observing the far field diffraction rings produced by self phase modulation of the beam at the incident power above the Freedericksz threshold.

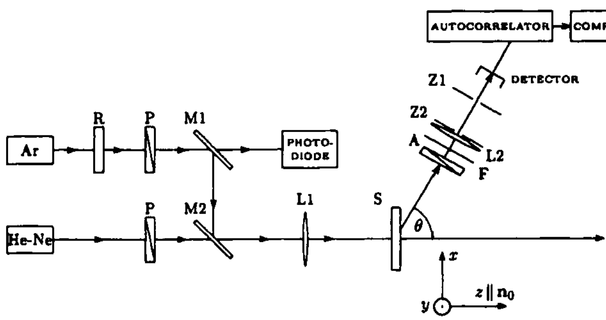

FIGURE 1: Experimental setup with polarization rotator R, polarizers P, analyser A, lenses L1, L2; pinholes Z1, Z2; dicroic mirrors M1, M2; color filter F, Ar laser beam (pump beam), He-Ne laser beam (probe beam), and sample S.

All the measurements were performed at room temperature. The intensity of the scattered light I_s of definite polarization and propagation direction was detected by a photomultiplier and its time dependence was analyzed on a digital autocorrelator providing the intensity autocorrelation function $g^{(2)}(t) = < I_s(t')I_s(t'+t) >$[13]. The system of two pinholes and a collimating lens was applied to arrange the observation of a single coherence area. The detection of either the pump or the probe beam light scattering was selected by use of an appropriate optical interference filter mounted in front of the detector.

Before starting the DLS measurements we checked the OFT of the sample by observing the far field diffraction patterns on the observation screen. The dependence of a divergence of the output beam and the number of the self diffraction rings on the incident power of the pump beam was measured. The OFT only appeared with the proper choice of the incident optical field polarization. This indicates that the transition was not due to laser heating, but to the light field torque, that affects the nematic molecules. The threshold power P_{tr} for the corresponding bend-splay Freedericksz transition was determined as a power at which the divergence of the outgoing beam started to increase and was found to be of about 40 mW. This value remained the same after repeating the measurements for several times, which shows that there was no sample degradation under irradiation. If the transverse effects of the Gaussian beam profile $I = I_0\, e^{-2r_\perp^2/w_0^2}$ were negligible, the induced director reorientation would be equivalent as in the plane wave situation and the critical field E_c would be[10]

$$\frac{1}{2}\frac{\varepsilon_\perp}{\varepsilon_\parallel}\varepsilon_0\varepsilon_a E_c^2 = K\left(\frac{\pi}{L}\right)^2 \tag{1}$$

where L is the sample thickness, $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the components of the optical dielectric tensor along and perpendicularly to the nematic director $\mathbf{n}$, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is optical anisotropy and K is an average Frank elastic constant. Taking into account the values of these material parameters for 5CB as reported in the literature[5, 14], expression (1) gives the critical laser power $P_c \approx 3$ mW. The about 13 times larger value of P_{tr} observed in our experiment shows the importance of the finite beam size and is in agreement with other results obtained at similar ratio of L/w_0[7].

Measurements at low scattering angles

The first set of the DLS measurements was performed at scattering angle θ of around 0.7°. The pump beam as well as the probe beam propagated in direction normal to the sample walls i.e. along the z axis and were linearly polarized along the x axis (Fig. 1). The polarization of the scattered light was selected to be within the scattering plane, that is in the xz plane. In homeotropic alignment such a geometry probes solely the bend-splay fluctuations, i.e. the fluctuations in the xz plane[2]. The components of the corresponding scattering wave vector at $\theta = 0.7°$ are $q_x = 14.9 \cdot 10^4\,\mathrm{m}^{-1}$ and $q_z = 4.7 \cdot 10^2\,\mathrm{m}^{-1}$ for the pump beam, and $q_x = 12.4 \cdot 10^4\,\mathrm{m}^{-1}$ and $q_z = 3.8 \cdot 10^2\,\mathrm{m}^{-1}$ for the probe beam, while for comparison: $\pi/w_0 = 15.7 \cdot 10^4\,\mathrm{m}^{-1}$. Due to the small scattering angle and nearly parallel in and out going polarizations a lot of elastic scattering was present in the experiment so that the regime of the detection was entirely heterodyne[13]; the inelastic light scattering contributed only about one percent of the total scattered power. For the incident optical power below the threshold value P_{tr} the time dependence of the measured autocorrelation function $g^{(2)}(t)$ was reasonably well fitted to a single exponential decay.

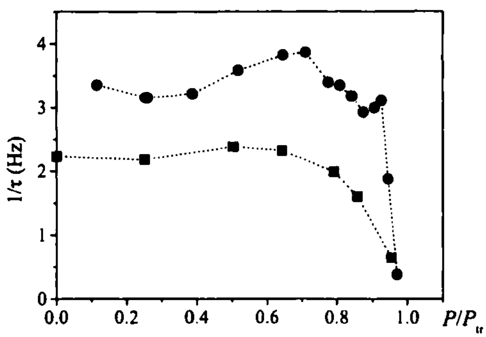

FIGURE 2: The dependencies of the inverse relaxation time of the bend-splay fluctuations on the incident optical power of the pump wave. The circles correspond to the pump beam scattering, the squares correspond to the probe beam scattering. The dotted lines connecting the data are just a guide for the eye.

Fig. 2 shows the dependence of the inverse relaxation time $1/\tau$ of the fluctuations as a function of the incident optical power P as obtained from the DLS of the pump beam and the DLS from the probe beam. The values differ due to different scattering wave vectors associated with the pump and the probe beam scattering. From $P = 0$ to $P \sim 0.7\,P_{tr}$ the inverse relaxation time is more or less constant. At about $0.7\,P_{tr}$ the behaviour changes significantly, the value of $1/\tau$ starts to decrease with increasing P and reduces by about 2 Hz till the threshold power P_{tr} is reached. For $P > P_{tr}$ the form of autocorrelation function $g^{(2)}(t)$ becomes nonexponetial (Fig. 4).

Fig. 3 shows the dependence of the ratio of inelastic to elastic light scattering intensity (I_{inel}/I_{el}) on the incident optical power P for the case of pump beam scattering. The data were obtained from the corresponding heterodyne autocorrelation function $g^{(2)}(t)$ as $I_{inel}/I_{el}=\frac{1}{2}\left[\left(g^{(2)}(t=0)/g^{(2)}(t=\infty)\right)-1\right]$[13]. Similarly to the dependence of $1/\tau$, again almost nothing

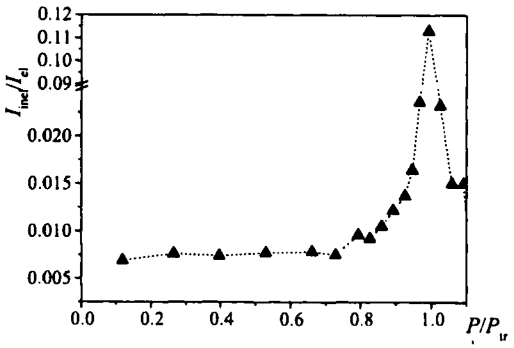

FIGURE 3: The dependence of the inelastic light scattering intensity of the pump wave on the incident optical power. The dotted line connecting the data is just a guide for the eye.

changes up to about $0.7\,P_{tr}$. At $P = 0.7\,P_{tr}$ the value of I_{inel}/I_{el} starts to increase and then grows in magnitude until the threshold is reached. Both the prominent decrease of the observed relaxation rate and the increase of the dynamic part of the scattering intensity are the evidence of the soft mode in OFT.

To elucidate the origin of the non-exponential form of the $g^{(2)}(t)$ above the threshold we performed a set of measurements at some slightly larger values of the scattering angle, that is at $\theta = 2°$, $4°$ and $6°$. The scattering related to the bend-splay fluctuations was analysed. The corresponding results for $g^{(2)}(t)$ obtained at $P = 1.2\,P_{tr}$ are shown in Fig. 4. The amplitude

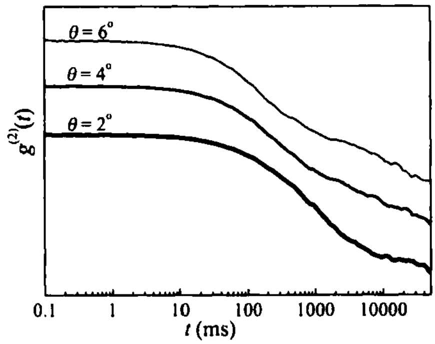

FIGURE 4: Intensity autocorrelation function associated to the scattering from bend splay fluctuations measured at three different scattering angles.

of $g^{(2)}(t)$ at different scattering angles varies due to different contributions of inelastic light scattering. In order to present only the form of these functions, which indicates the time dependence, the obtained autocorrelation functions were renormalized and then shifted with respect to the ordinate axis. The angle of the divergence of the outgoing beam at $P = 1.2\,P_{tr}$ was around $\theta_d = 10°$, therefore all the measurements correspond to the DLS detector positioned within the "ring" structure of the outgoing beam related to the self phase modulation. It can be noticed that at short correlation times in the ms region the $g^{(2)}(t)$ starts as the usual exponential relaxation, which then at longer correlation times transforms to a gradually decreasing "tail" extending over several decades and exhibiting nearly a logarithmic time dependence. The relaxation rate associated with the faster process increases with increasing scattering angle - suggesting that it corresponds to the usual director fluctuation modes. The tail, on the contrary, retains the same shape at all scattering angles. We observed that the tail vanishes when the scattering angle θ becomes larger than the angle of the divergence of the outgoing beam θ_d.

Measurements at large scattering angles

The next set of measurements regarded the properties of the DLS at scattering angles larger than the value of the θ_d at large incident powers. The dependence of the $g^{(2)}(t)$ on the incident optical power was studied for $\theta = 60°$. The components of the corresponding scattering wave vector for pump beam scattering are $q_x = 10.6 \cdot 10^6\,\mathrm{m}^{-1}$ and $q_z = 2.5 \cdot 10^6\,\mathrm{m}^{-1}$, and are both much larger than the ratio of $\pi/w_0 = 15.7 \cdot 10^4\,\mathrm{m}^{-1}$, which is characteristic for the spatial profile of the reorientational disturbance. The regime of scattering was practically homodyne at all the incident powers. In contrast to the low angle measurements, the form of the $g^{(2)}(t)$ revealed to be single exponential also for $P > P_{tr}$.

Fig. 5 shows the dependencies of the inverse relaxation time $1/\tau$ of the bend-splay mode and the inelastic light scattering intensity I_{inel} on the incident optical power P. For $P < P_{tr}$ the inverse relaxation time $1/\tau$

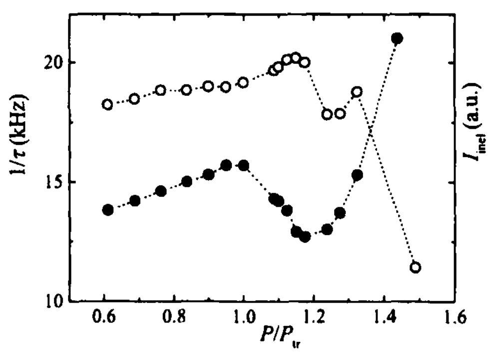

FIGURE 5: The inverse relaxation time of the bend-splay fluctuations (full circles) and the corresponding inelastic light scattering intensity (open circles) as functions of the incident optical power measured at scattering angle $\theta = 60°$ and at incident angle $\alpha = 0°$. The dotted lines are just a guide for the eye.

slightly increases with the increasing power. This increase very probably originates from the refractive index changes due to the laser heating of the sample, which affect the magnitude of the q_z component of the scattering wave vector[14]. For $P > P_{tr}$ the variations of $1/\tau$ are much more radical, exhibiting a characteristic minimum at $P \approx 1.2\, P_{tr}$. The dependence of I_{inel} is very similar - except that the position of its minimum is slightly shifted to some lower value of P.

By repeating the measurements the characteristic minima shown on Fig. 5 sometimes disappeared. This may be explained by the degeneracy of the direction of the molecular reorientation at OFT. In optical field polarized along the x axis the nematic director can rotate from its initial direction $\mathbf{n_0} = (0, 0, 1)$ either toward $\mathbf{n} = (1, 0, 0)$ or toward $\mathbf{n} = (-1, 0, 0)$. The occurence of one or the other specific direction of the reorientation, which is in principle random, affect the dependencies of relaxation rate and the scattering cross section on the input optical power. This is illustrated in Fig. 6, which shows the dependencies of $1/\tau$ and I_{inel} as functions of P for

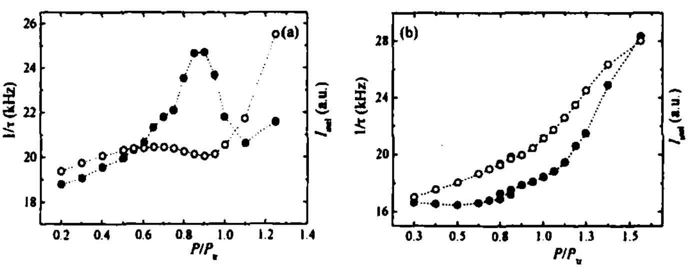

FIGURE 6: The inverse relaxation time of the bend-splay fluctuations (full circles) and the corresponding inelastic light scattering intensity (open circles) as functions of the incident optical power measured at $\theta = 60°$ and at incident angle $\alpha = +5°$ (a) and $\alpha = -5°$ (b). The threshold power is related to the measurements at normal incidence. The dotted lines are just a guide for the eye.

the two situations when the pump beam entered the sample at an incident angles α of 5° and −5° respectively (see also Fig. 1). At these incident angles no threshold behaviour is present, and the direction of the molecular reorientation is well defined. For $\alpha = 5°$ the director rotates toward the direction of the DLS detection (60°), and for $\alpha = -5°$ it rotates away from the direction of the DLS detection. One can see that in the first case the dependencies of $1/\tau(P)$ and $I_{\rm inel}(P)$ exhibit significant minima and are very similar to the ones shown in Fig. 5, while for $\alpha = -5°$ the monotonous behaviour is present. From qualitative comparison of the behaviours given by Fig. 5 and Fig. 6 it can be concluded that in the OFT experiment corresponding to the Fig. 5 the director reorientation also took place in direction toward the direction of the DLS detection.

The angular properties of the DLS signal at large scattering angles are given in Fig. 7. which shows the dependence of the inverse relaxation time

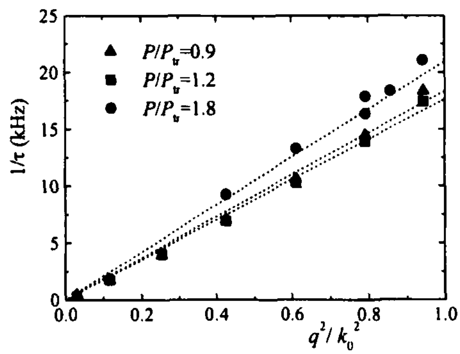

FIGURE 7: The dependence of the inverse relaxation time of the fluctuations on the scattering wave vector q at three different incident powers (at normal incidence), where k_0 denotes the wave vector of light in vacuum. Dotted lines are fits to the quadratic dispersion relation.

$1/\tau$ as a function of the square of the scattering wave vector $\mathbf{q}$ at three different incident optical powers. The scattering wave vector $\mathbf{q}$ corresponding to the various scattering angles was calculated taking into account the refractive indices of the initial $\mathbf{n}_0 = (0, 0, 1)$ director field. Solely the $1/\tau$ data obtained for scattering angles $\theta > \theta_d$ are given at a specific incident power. The observed dependencies fit relatively well to the usual quadratic dispersion relation, which is characteristic for homogeneous liquid crystalline phase[2].

CONCLUSIONS

We have experimentally observed the critical slowing down of the director fluctuations in the vicinity of the optical field induced Freedericksz transition in a homeotropic nematic liquid crystal cell. The observed dependence of the inverse relaxation time of the critical mode on the input optical power is very different from the linearly decreasing behaviour, which is characteristic for the Freedericksz transition in homogeneous external fields.

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