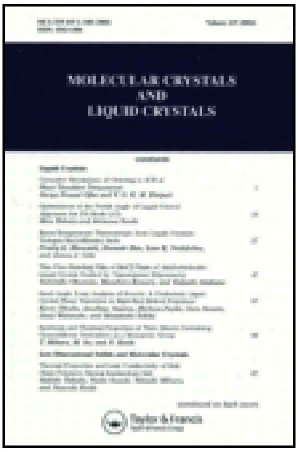

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Stationary Optical Noise in Planar Nematic Liquid Crystals Near the Fréedericksz Transition

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We measure, under stationary conditions, the power spectrum of the transmitted and scattered light intensity in a planar nematic liquid crystal cell as a function of the applied voltage in the neighbourhood of the electrically-induced splay Fréedericksz transition. The Fréedericksz threshold is independently determined using a standard interferometric technique. The low-frequency noise spectral density of the transmitted light displays a well defined peak at the critical voltage; this feature is instead hidden in the scattering noise. We critically examine the performances of these different experimental techniques and show that noise measurements are an adequate tool to study orientation transitions in nematic liquid crystals.

Keywords: Fréedericksz transition; Scattering; Noise.

1. INTRODUCTION

As it is well known, the analysis of the thermal fluctuations of the director gives useful informations on the viscous and elastic constants of a nematic liquid crystal.[1] The fluctuations are usually detected by measuring the mean value and the spectral density of the intensity of the light scattered by a homogeneously oriented liquid crystal sample. Recently, noise analysis techniques have also been proposed to study surface anchoring energies[2] and field-induced orientation transitions in planar cells.[3] The effects of the finite sample thickness have also been investigated.[2,4]

In Reference 3 the electrically induced splay Fréedericksz transition—a second order transition in which the maximum distortion angle, in the middle of the cell, plays the role of an order parameter[5]—was studied by looking at the low frequency behaviour of the noise-spectrum of the intensity of the light scattered at large angles. The idea was to exploit the indirect modulation, through the variation of the geometric factors, by the soft critical and quasi-critical fluctuation modes on the fast relaxing noncritical modes responsible for the large-angle scattering, a modulation that should give rise to a peak in the low frequency spectrum in correspondence to the Fréedericksz transition: this second order effect, similar to the Raman effect, was enhanced by collecting a large number N of coherence zones, in such a way that the high frequency spectral components turn out to be equal to N-times the contribution of each coherence zone, since they come from uncorrelated contributions of different fluctuation modes of the director, while the low frequency modulation is equal to N^2-times the contribution of each coherence zone. In fact this modulation is due to the director rotation within the illuminated volume coming from the critical and quasi-critical modes, namely from the modes whose wavevector $\mathbf{q}$ has components $q_\perp = \pi/d$ and $q_\parallel << \pi/d$, where d is the sample thickness and $\parallel$ and $\perp$ refer to the sample plane.

Actually this approach has severe experimental drawbacks. In fact to collect a sufficiently large number of coherence zones one is forced to enlarge the acceptance angle of the detector and/or the incident beam waist: this in turn increases the contribution of the defects to the scattered light and thus conceals the low frequency noise spectrum, an effect which is particularly strong when one deals with thin samples (typically less than 50 μm). Moreover, above the Fréedericksz transition, the continuous distortion of the director profile modifies the power spectrum of the scattered intensity in a way which might override the critical behaviour. Last but not least it must be noted that a sharp splay Fréedericksz transition is to be expected, in principle, only in strictly planar samples[6]: a non-negligible surface pretilt angle (typically on the order of a tenth of a degree) will destroy the critical noise.

In the present work, in order to overcome most of these difficulties, we measure, in the neighbourhood of the electrically-induced splay Fréedericksz transition, the noise power spectrum of the light transmitted by a planar nematic liquid crystal cell, placed between crossed polarizers: this has the advantage of being a first-order effect, since the critical and quasi-critical fluctuation modes directly contribute to the phase of the transmitted beam.[4]

In Sect. 2 we present our experimental set-up and we determine, using a standard interferometric technique,[7] the splay Fréedericksz transition voltage of the cell that we will employ for the subsequent noise measurements. In Sect. 3 we measure the noise spectrum near the Fréedericksz threshold both in the scattered and in the transmitted light, showing that in the latter case there is a definite anomaly in the low frequency behaviour, a feature which is instead hidden in the scattering noise. Finally in Sect. 4 we briefly discuss our experimental results.

2. EXPERIMENTAL SET-UP

A highly homogeneous nematic liquid crystal sample with strong planar anchoring was prepared in a class-1000 clean room by Tecdis S.p.A. by means of a standard assembly technique. The cell consists of two glass plates coated with ITO conducting electrodes and surfactant polyimide layers, having a thickness of 0.6–0.9 μm, mechanically rubbed in one direction in order to obtain a strong planar anchoring. The empty cell thickness was interferometrically measured and found to be (8.3 ± 0.05) μm. The cell was then filled by capillary action in a low pressure glass-bell with the BDH K15 nematic liquid crystal, which has a positive dielectric anisotropy.

The splay Fréedericksz transition is induced by a zero mean value sinusoidal voltage, having a frequency of 1 KHz, applied to the cell electrodes. The RMS amplitude of the electric signal is controlled by a PC. All the measurements were performed at room temperature ((23 ± 0.5)°C).

The light source used to investigate the transition is provided by a 10 mW He-Ne laser ($\lambda = 632.8$ nm), spatially filtered and focused by a set of diverging and converging lenses in order to reduce the beam waist on the sample to ≈ 200 μm. In all the measurements the director is always in the incidence plane and the incident beam is linearly polarized. The light emerging from the sample is passed through a polarizer, collected by an objective lens (diameter 1 cm, focal length 5 cm, acceptance angle 10°) and focused onto a low-noise photodiode connected to a high-gain amplifier. The output of the amplifier is sent to a digital FFT-spectrum analyzer to measure the power spectral density of the detected light-intensity and to a digital oscilloscope to monitor the mean value: both instruments are interfaced to the PC. The detecting stage is mounted on a platform that can rotate in the incident plane around an axis coincident with the sample, which in turns can independently rotate about the same axis in order to change the incidence angle. All the various measurements that we performed were realized by suitably adjusting the incidence angle, the detection angle and the polarization of the incident and detected light.

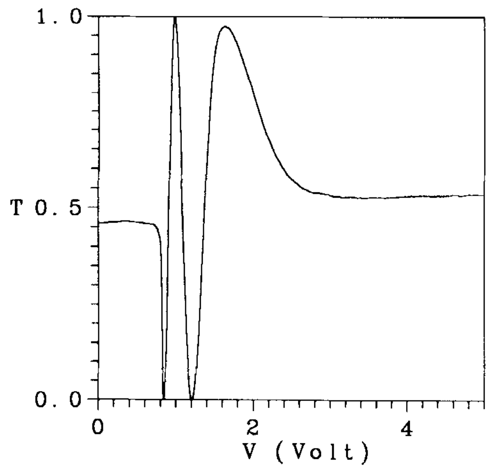

FIGURE 1 Transmittance T as a function of the RMS applied voltage V for an incidence angle $\vartheta_i = 30°$. The sample is placed between cross polarizers making an angle of 45° with respect to the incidence plane, in which the nematic director lies.

First of all, in order to locate the Fréedericksz threshold and to control the quality of the cell, we performed standard fringes measurements as a function of the applied voltage: this technique is based on the interference effect between the ordinary and the extraordinary ray propagating in the liquid crystal sample.[7] The measurement is done by detecting the mean intensity of the transmitted beam, I, with the polarizer and the analyzer crossed with respect to each other and at 45° with respect to the incidence plane. The incidence angle can be selected at will, but the highest sensitivity will be reached when the initial transmittance is around 0.5. In Figure 1 we show the resulting fringes measurements for an incidence angle of 30°: since the incident intensity I_0 provided by the laser is sufficiently stable during the measurement cycle, it was not necessary to monitor the incident intensity and the transmittance $T = I/I_0$ was computed by assuming equal to one the maximum transmitted intensity.

In principle, by fitting the experimental curve with the theoretical prediction, it is possible to extract many physical parameters of the system: in practice, if no further informations are available (such as the exact values of the indices of refraction and the thickness of the cell), the numerous degrees of freedom make it difficult to separately and accurately determine each parameter. For the case at hand we limited ourselves to determining the Fréedericksz transition voltage V_F—which marks the onset of the fringe pattern—and to get an estimate of the mean surface pretilt angle—which is responsible for the smoothing of the transition—: precisely we find $V_F = (0.774 \pm 0.008)V$, while the mean pretilt angle is in the range $3\text{–}5 \times 10^{-2}$ degrees. Different choices of the incidence angle, and of the illuminated spot of the sample, gave equivalent results, thus assuring the homogeneity of the cell.

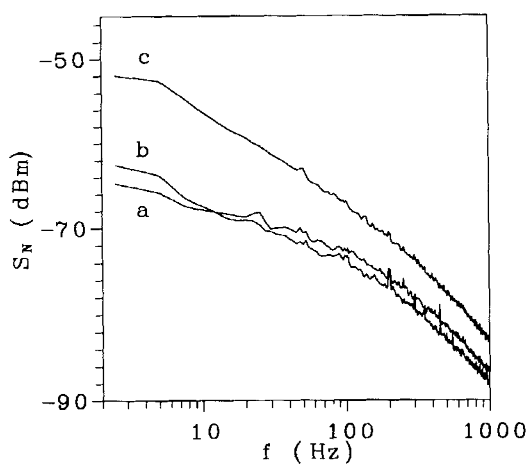

FIGURE 2 Normalized spectral density S_N of the scattered-light noise as a function of the frequency f for three different RMS applied voltages V (curve a, V = 0V; curve b, V = 0.53V; curve c, V = 0.82V). The incidence angle is zero while the mean scattering angle is 10° with an acceptance angle of 10°; the polarization of both the incident and scattered fields is extraordinary (E-E configuration).

3. NOISE MEASUREMENTS

Two types of noise measurements in the neighbourhood of the splay Fréedericksz transition were performed, using the previously described experimental set-up, in order to uncover a low frequency peak related to the critical soft mode.

In the first instance, following the guide-lines of Reference 3, we measured the noise power spectrum in the scattered intensity away from the transmitted beam: different combinations of polarizations and angles for the incident and scattered beams were tested. The geometry in which we observed the strongest effects as a function of the applied voltage corresponds to normal incidence and small scattering angles with E-E polarization (incident and scattered field both linearly polarized in the incidence plane, which in our case coincides with the scattering plane and

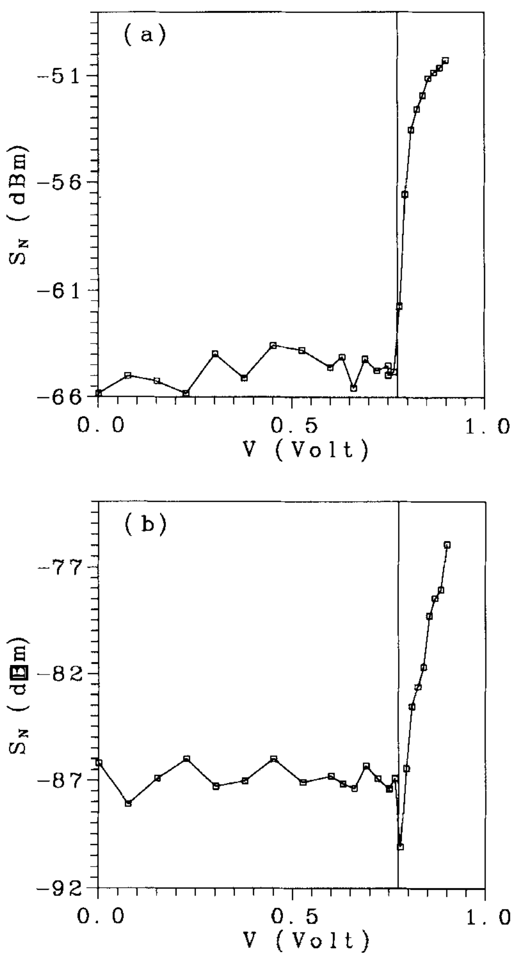

FIGURE 3 Same as Figure 2 but for a fixed frequency f as a function of the RMS applied voltage V. a) f = 2.5 Hz; b) f = 960 Hz. Here and in the following figures the vertical lines correspond to the Fréedericksz threshold V_F = 0.774V determined by the fringes measurements.

with the plane in which the director field distortion takes place). The observed power spectra (see Figure 2) do not show a Lorentzian shape due to the fact that, owing to the objective lens, the detected light is composed of contributions scattered at various angles, and thus corresponds to a wide range of exchanged momenta: in our case the external mean scattering angle was 10°, and thus, given the acceptance angle of the lens, the external scattering angles ϑ_s extended in the interval $5° \leq \vartheta_s \leq 15°$. The presence of the lens, together with the reduced cell thickness, also implies that we are always working in the heterodyne regime.

In Figure 3 we show the normalized power spectral densities S_N of the scattered light noise as a function of the RMS applied voltage V at fixed frequencies f: precisely in Figure 3a we have $f = 2.5$ Hz, while in Figure 3b $f = 960$ Hz. The normalization is performed by dividing each spectrum by the corresponding mean value of the scattered light intensity: in this manner the changes in the noise level merely related to variations of the signal intensity are largely compensated. The experimental data do not show any evidence of a low frequency peak related to the Fréedericksz transition, and in particular no evidence of a pretransitional effect: the curves are essentially flat until the Fréedericksz threshold is reached; from this point on an abrupt change of the slope is observed, a feature which is present at all the frequencies and which is due to the continuous distortion of the director profile above the Fréedericksz threshold; in fact, as a first rough approximation, the inhomogeneous distortion of the director can be assimilated to a rigid rotation by a suitably chosen mean angle: in our conditions (normal incidence and low scattering angles with E-E polarization) this induces a marked increase in the scattering cross-section. Unfortunately the experimental points display a rather broad uncertainty—largely due to the reduced thickness of the sample (8.3 μm) as compared to the surface defect density—particularly below the Fréedericksz threshold, where the scattering cross-section is lower. The variation of the noise spectrum at low and high frequencies is different, as shown by Figure 4, where the

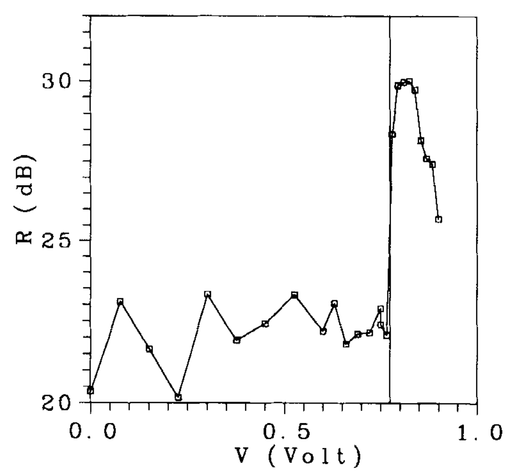

FIGURE 4 Ratio R between the low-frequency ($f = 2.5$ Hz) and the high-frequency ($f = 960$ Hz) spectral densities shown in Figures 3a and 3b, respectively, as a function of the RMS applied voltage V.

ratio between the power spectra plotted in Figures 3a and 3b, respectively, is displayed (since all the spectra are plotted in log scale, this ratio actually corresponds to a difference between the two plotted values). The peak which is observed is significantly shifted above the Fréedericksz threshold and is not directly related to the transition.

These results can be coherently interpreted according to the analysis already outlined in the Introduction: in fact under the present conditions the effect of the critical and quasi-critical fluctuation modes on the detected scattered fields is only indirect, through a second order mechanism, and in these circumstances it is largely hidden by various spurious effects, as e.g. the enhancement of the scattering cross-section consequent to any small rotation of the director, since in the undistorted

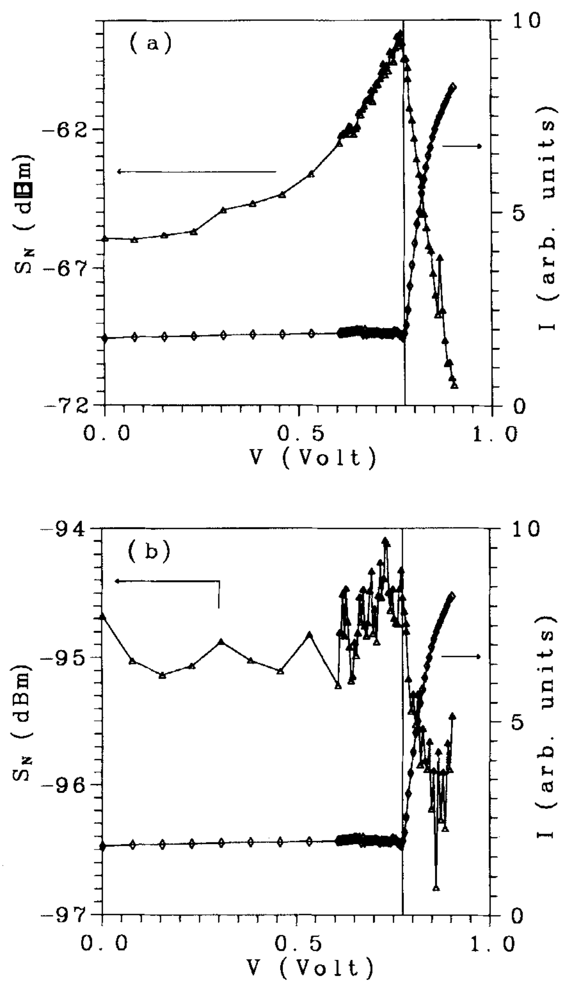

FIGURE 5 Normalized spectral densities of the transmitted-light noise S_N at a fixed frequency f (Δ, left-hand scale) and transmitted-light intensity I ($\Diamond$, right-hand scale) as a function of the RMS applied voltage V. The experimental conditions are the same as in Figure 1, but with an incidence angle equal to 50°. a) f = 2.5 Hz; b) f = 960 Hz.

state the E-E scattering cross-section, for normal incidence and low scattering angles, has a deep minimum.[1] On the other hand the fluctuation modes directly responsible for the scattered light do not have any critical behaviour due to the large transverse component of their wavevectors.

In order to directly detect the critical fluctuation mode we performed noise measurements in the transmitted light. The experimental geometry is the same as in the case of the fringes measurements and thus has the advantage of allowing a contemporaneous and independent monitoring of the behaviour of the director profile using a well established and reliable technique. The real benefit of the transmitted-light measurements consists in picking up virtually all the director thermal fluctuation modes, thus including the critical one.

The experimental spectra of a typical transmittance-noise measurement as a function of the RMS applied voltage V are shown in Figures 5, for low frequencies (a, f = 2.5 Hz) and high frequencies (b, f = 960 Hz). The incidence angle is equal to 50°. The spectra are again normalized with respect to the average value of the transmitted intensity, which corresponds to the first part of an interference fringes pattern and is reported, in arbitrary units, on the same graphs. Now, in correspondence to the beginning of the first interference fringe, a maximum in the noise level is indeed observed at low frequencies, while it practically disappears in the high-frequency limit. These results remain practically unchanged if one does not normalize the spectra with respect to the mean values.

It is therefore possible to conclude that the present measurements provide unequivocal evidence for a critical behaviour of the thermal noise at low frequencies in the neighbourhood of a second order orientation transition.

4. CONCLUDING REMARKS

In the present work we have shown the existence of an anomaly in the noise power spectrum of the intensity of the light emerging from a planar nematic liquid crystal in correspondence to a second order orientation transition of the director field. Large-angle light scattering measurements have proven to be rather unpractical in evidencing this noise anomaly, mainly because the results are exceedingly influenced by the sample conditions, such as surface pretilt angles and defects. Although the Fréedericksz transition can be located, even if with a rather large uncertainty, in noise measurements in the scattered-light, it appears not directly related to a maximum in the noise intensity, but to a continuous modification of the spectra. In the light of these results, the noise-intensity peak reported in a preliminary paper[3] might be related to an enhancement of the scattering cross section and to a modification of the spectral shape in an already distorted sample.

Transmitted-light noise measurements evidence, beyond any doubt, the anomalous behaviour of the noise at a second-order orientation transition: this new method has the advantage of directly detecting the critical thermal fluctuation mode. The noise anomaly at the threshold $V = V_F$ is observed to increase in magnitude with decreasing analysis frequency. This result is in agreement with the general views about the dynamical behaviour of a physical system undergoing a second order phase or orientation transition, characterized by a critical slowing-down of certain fluctuation modes. Such behaviour explains the occurrence at $V = V_F$ of a maximum in the low-frequency spectral density of the transmitted-light noise. We note that a true divergence is expected only at zero frequency and only for a linearized model (e.g. truncating a Landau expansion of the free-energy around the undistorted configuration at the quadratic terms).

Our results show the adequacy of noise-analysis techniques in the study of electrically or magnetically-induced transitions in nematic liquid crystals. Improved noise-analysis methods, such as the simultaneous study of beams scattered at different angles by means of cross-correlation techniques, might be a useful tool in more advanced studies of orientation or phase transitions in liquid crystals.

References

1. See e.g. E. Miraldi, L. Trossi, P. T. Valabrega and C. Oldano, *Il Nuovo Cimento B*, **60**, 165 (1980) and references quoted therein.
2. J. Papanek, *Mol. Cryst. Liq. Cryst.*, **179**, 139 (1990).
3. P. Allia, C. Oldano, P. T. Valabrega and L. Trossi, *Phys. Scripta*, **44**, 388 (1991).
4. B. Ya. Zel'dovich and N. V. Tabiryan, *Zh. Eksp. Teor. Fiz.*, **81**, 1738 (1981) [*Sov. Phys. JETP*, **54**, 922 (1981)].
5. See e.g. P. G. de Gennes, "The Physics of Liquid Crystals," Clarendon Press, Oxford (1974).
6. P. Galatola and C. Oldano, "Fréedericksz Transitions in Zero-Field Distorted Nematic Liquid Crystals," *Phys. Rev. A*, in press.
7. A. Saupe, *Z. Naturforsch.*, **15A**, 815 (1960); C. Oldano, E. Miraldi, P. T. Valabrega, *J. Physique*, **45**, 755 (1984).