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The Electrohydrodynamic Instability in a Homeotropically Aligned Nematic: Experimental Results

Visual observations and measurements of the onset of electroconvection in a homeotropically aligned nematic liquid crystal MBBA are presented. Because of its negative dielectric anisotropy, the first instability to occur when increasing the voltage across the layer is the electrically driven bend Fréedericksz transition. Further increase of the voltage leads to electroconvection. This paper characterizes the convection pattern close to the onset of convection in order to allow a comparison with a linear stability analysis of the Fréedericksz state.

1. INTRODUCTION

In a recent interesting paper, Hertrich et al.[2] pointed out the possibility of complicated spatio-temporal behavior close to the onset of convection, when a nematic liquid crystal with negative dielectric anisotropy in a homeotropically aligned configuration is used. The reason for this expectation is the fact that, because of the rotational symmetry of the boundary conditions, no orientation of the expected roll pattern is preferred. Because of the negative dielectric anisotropy and the

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homeotropic alignment, the first instability to occur is the Fréedericksz transition. This is not a pattern-forming instability, but a spontaneous break in the rotational symmetry. Under idealized conditions there are no forces fixing the orientation of the director in the plane of the layer; thus, this state is expected to be metastable. If convection sets in and if the rolls are normal to the director projection in the plane of the layer, there is no reason to believe that this metastability would be distorted. If, on the other hand, the rolls are oblique with respect to this director projection, this additionally broken symmetry gives rise to the expectation that the orientation of the rolls would not be metastable any more: the rolls might constantly reorient their position. As such a reorientation cannot be expected to occur simultaneously over the entire cell, one might have a complicated, time-dependent pattern directly at threshold. The necessary condition for this interesting scenario is the existence of oblique rolls. The Hertrich paper[2] did indeed predict oblique rolls for small frequencies of the driving electric field, and normal rolls for larger frequencies. The goal of the work presented here is a first attempt to check this prediction experimentally. The most interesting result is that we do not find oblique rolls, but rather normal rolls within the whole experimentally accessible frequency regime. At high frequencies, however, the normal rolls break the left-right symmetry in a peculiar way which is not yet understood.

2. EXPERIMENTAL SETUP

The nematic liquid crystal MBBA is sandwiched between two transparent electrodes with a spacing of 24 μm. In order to achieve homeotropic alignment, these electrodes are sputtered with chrome[1]. An attractive feature of this novel procedure seems to be a good stability of the sample—within half a year there were no obvious signs of deterioration. A disadvantage of this coating is a tremendous reduction of the light intensity by the chrome layer, which has a thickness of about 200 Å. Using an exposure time of 0.1 s, we get an image measured by means of a 512×512 square pixel, 14-bit slow-scan CCD-camera. The Fréedericksz transition and the electroconvection are driven by applying an ac-coupled sinusoidal current to the cell. In order to allow an easy access to the cell, we did not use any temperature control of the sample. Thus, most of the measurements shown here have been performed at (24 ± 2) °C, unless otherwise stated.

3. RESULTS

Figure 1 is the phase diagram of our sample. We measured the voltage across the sample for the Fréedericksz transition and the onset of electroconvection for different frequencies of the driving field. The Fréedericksz transition occurs at an rms voltage of $V_F = 3.2$ V. Within our resolution and in the range of frequencies shown here, we found no sign for a frequency dependence of this threshold (such an effect is not expected, either); thus, the threshold $V_F(f)$ is just a straight line. The curved line shows the onset of electroconvection. It has been determined by visual inspection of the sample, a method which gives reasonable results within a resolution of 1%. In order to give an idea of the reproducibility of this measurement, open squares are shown representing the measured onset of convection when increasing the frequency of the ac-field, and solid circles correspond to the measurement when decreasing the frequency. The visual deviation of these two points at 3000 Hz might reflect the temperature influence in this sensitive part of the threshold curve. In order to avoid deterioration of the sample, we did not perform measurements above 25 V. From a comparison of this threshold curve with the one presented by Hertrich et al.[2] (see Figure 5 therein), we would expect the predicted Lifshitz-point, where the crossover from normal to oblique rolls takes place, to be located around a driving frequency of 1600 Hz.

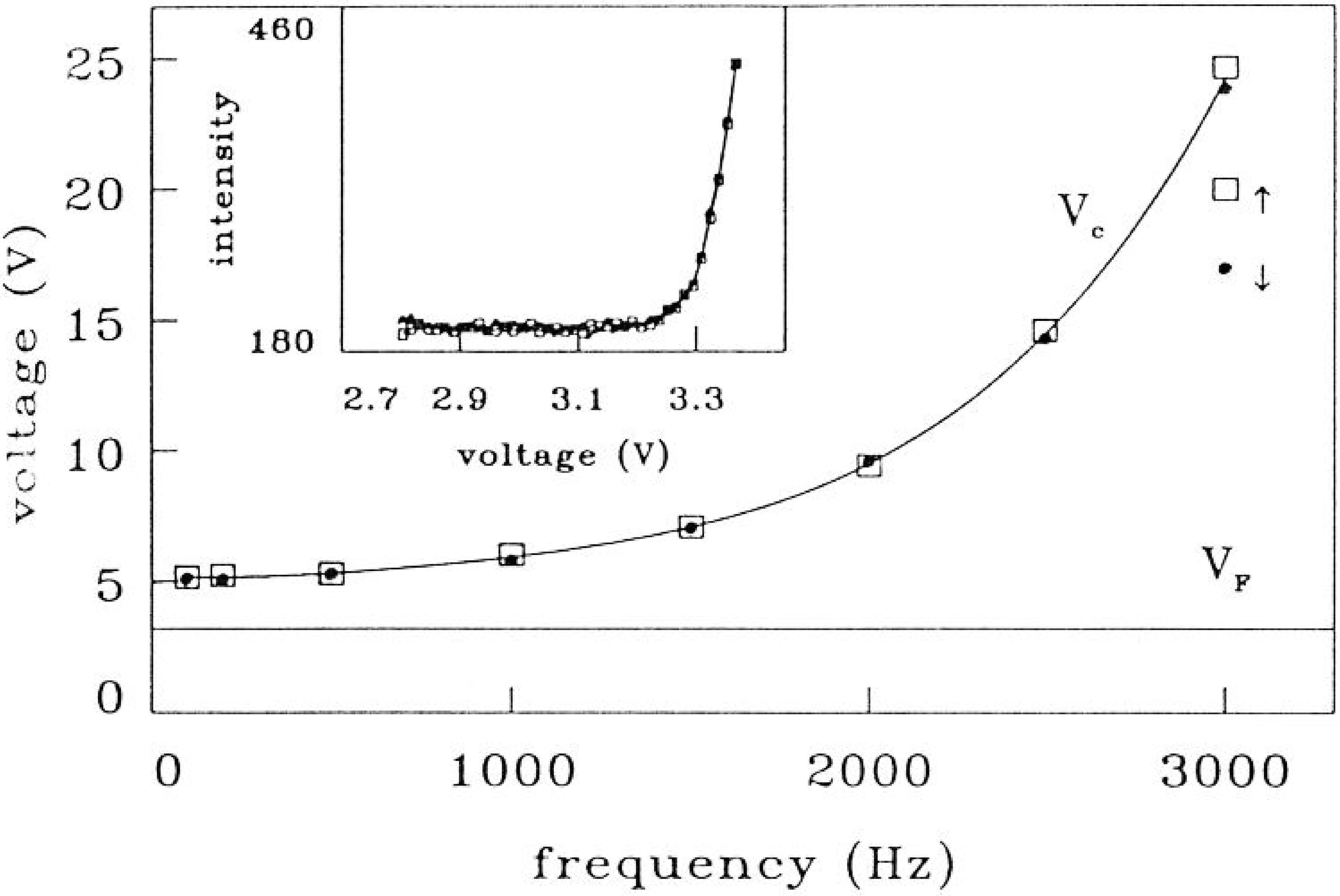

FIGURE 1 Phase diagram showing the Fréedericksz transition and the onset of convection. The inset is a detailed measurement of the Fréedericksz transition.

It is important to notice that the Fréedericksz transition is not perfect; it is rather slightly rounded as indicated by the onset measurement shown in the inset of Figure 1. Here, we crossed the polarizer and the analyzer and measured the intensity of one single pixel of the CCD-array. Due to the absorption of the polarizer and the analyzer, an exposure time of 1 s has to be used in this case. The underlying idea of this procedure is the fact that the CCD-array, being dc-coupled to the 14-bit AD-converter, is capable of measuring absolute light intensities. We measured these light intensities for different voltages of the driving electric field. The open squares correspond to increasing voltage, and the solid circles were obtained by decreasing the voltage. We would like to mention that there is a specific difficulty with this method for the homeotropic alignment. If the orientation of either the polarizer or the analyzer is parallel to the director, the image remains black above threshold. For any other orientation, an increase in the intensity occurs once the director starts to reorient. Fortunately, it turns out that the director orients always in the same direction, if the transition is performed sufficiently slow. Thus it is possible to orient the polarizer and analyzer in the appropriate direction in order to be sensitive to the bend Fréedericksz transition.

We believe that the rounding of the measurement curve shown in the inset of Figure 1 indicates that the homeotropic alignment is not geometrically perfect, although there is no obvious distortion of this alignment when observing the cell under crossed polarizers below threshold. This imperfection presumably also manifests itself in the fact that the orientation of the director is reproducible, which is considered as a great help performing the measurements of the convection onset.

The measurements were made in a frequency range from 15 Hz up to 3 kHz. We observed two different kinds of convection patterns with a crossover taking place at about 100 Hz. Below this frequency we believe to get normal rolls as demonstrated below, and for higher frequencies a peculiar pattern. We choose two frequencies, 30 Hz and 1000 Hz, to demonstrate the two different regions.

The orientation of the director is measured by the procedure indicated in Figure 2. Here we rotated the crossed polarizer and analyzer, and measured the resulting intensity within a small fraction of the observation field (150 × 150 pixels), where the orientation of the director was fairly homogeneous. The rotation angle $\alpha = 0$ of the polarizer is chosen arbitrarily. In Figure 2 (a) we represent the result of this procedure above the Fréedericksz transition and below the onset of convection. The data are taken at a frequency of 30 Hz and a voltage of 4.02 V. From this curve we conclude that the mean orientation of the director φ_d is either 0° or 90°. The lower part of the figure shows the result obtained above the onset of convection (30 Hz, 5.76 V). The solid line, which is supposed to be a guide for the eye, is a fit to harmonical functions. Our interpretation of these two figures is that the convection did not change the mean orientation of the director, at least within the resolution given by this procedure (about 10°).

In Figure 3 we show the same comparison for a higher frequency of 1000 Hz. In Figure 3, view(a) is taken at 4.31 V, and (b) at 6.64 V. The conclusion is

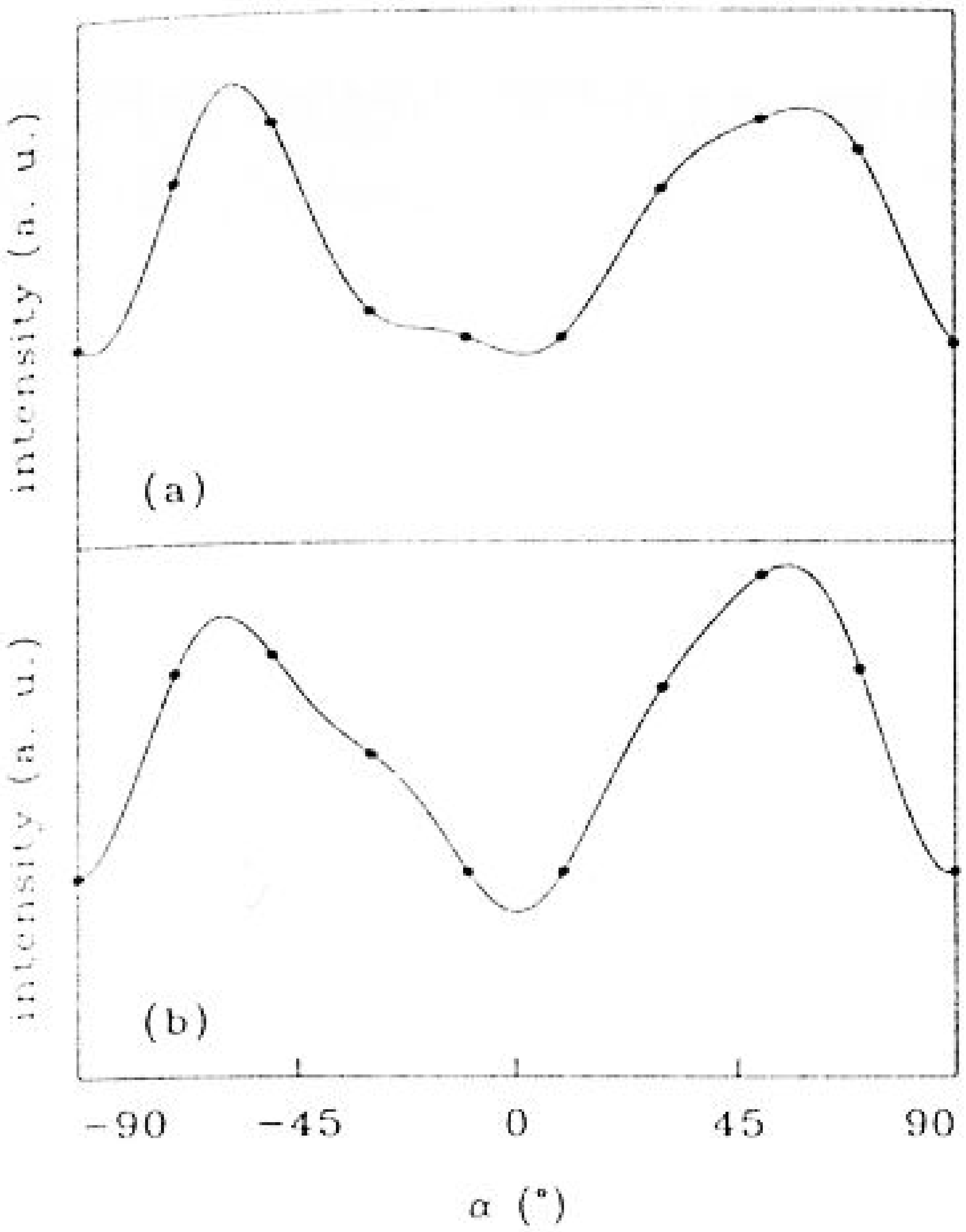

FIGURE 2 Determination of the director orientation at 30 Hz. (a) Below the onset of convection, 4.02 V, (b) with convection, 5.76 V. The solid line is a guide for the eye.

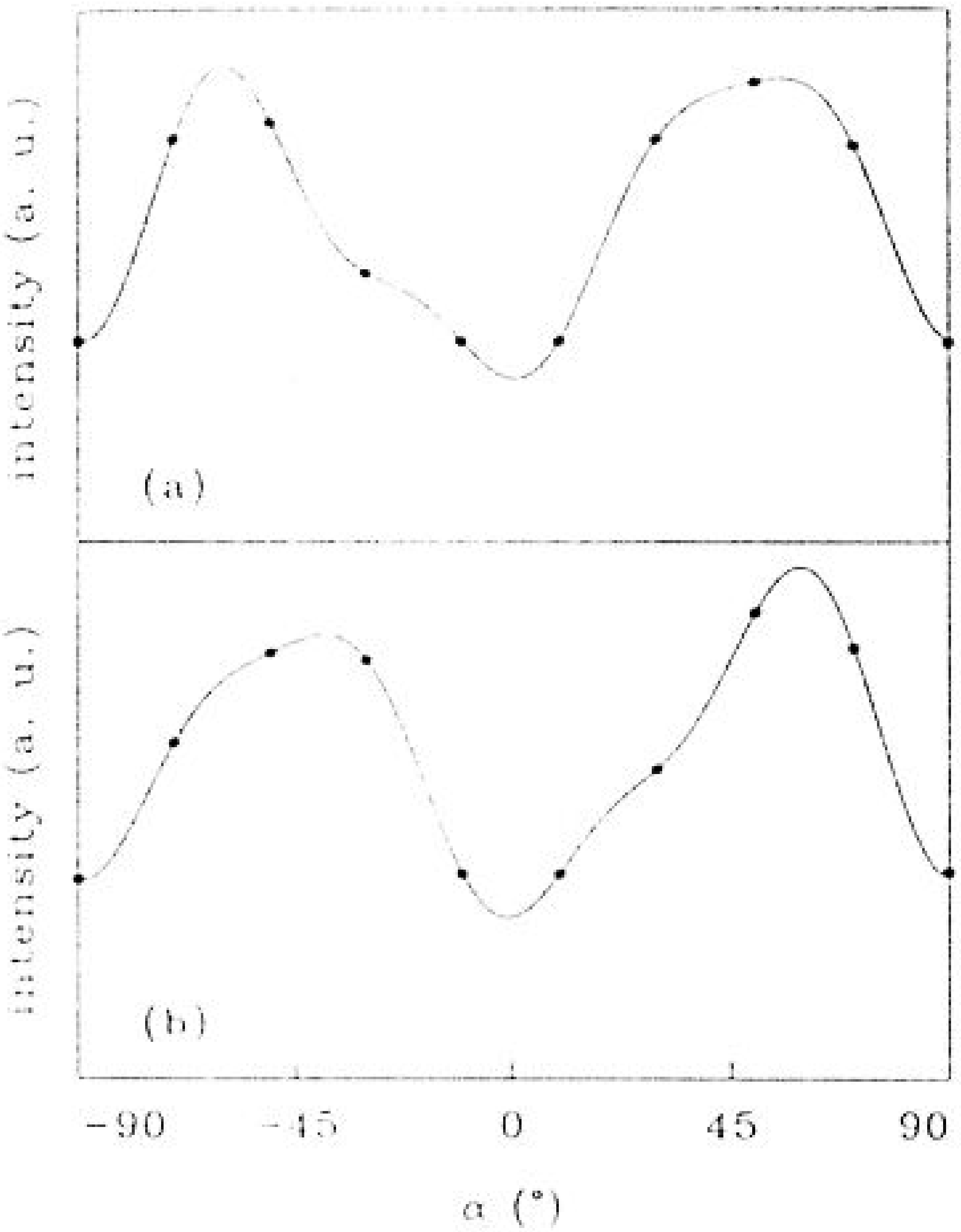

FIGURE 3 Determination of the director orientation at 1000 Hz. (a) Below the onset of convection, 4.31 V, (b) with convection, 6.64 V.

the same as in Figure 2, namely that the mean orientation of the director field is not much changed by the onset of convection. This statement seems to apply for all frequencies. The angle for the orientation of the director is measured as $\varphi_d = 0° \pm 10°$.

FIGURE 4 Image of the convection pattern of an area of 1000 μm × 1000 μm at 30 Hz, $\epsilon = (V/V_c)^2 - 1 \approx 0.1$ (28 °C).

FIGURE 5 Image of the convection pattern of an area of 1000 μm × 1000 μm at 1000 Hz, $\epsilon = (V/V_c)^2 - 1 \approx 0.1$ (28 °C).

The convection patterns above threshold for these two frequencies of 30 Hz and 1000 Hz are shown in Figures 4 and 5. They indicate that there is clearly a preferred direction for the rolls which varies slightly with the position in the sample, presumably caused by the change of the preferred orientation of the director. There is no obvious change in the orientation of the rolls as a function of the frequency. Thus, by means of visual observations like these, together with the determination of the orientation of the director as indicated above, and the contrast measurements described below, we draw the conclusion that the rolls are normal to the director field for any frequency within the range shown in Figure 1. The theory, on the other hand, would predict oblique rolls for small frequencies below 1600 Hz or so. It is worth mentioning that from the measurements of Figures 2 and 3 it cannot be excluded that the roll orientation is parallel to the director. This would, however, be very improbable from a theoretical point of view. Moreover, it would make the interpretation of the following contrast measurements very difficult.

There is something peculiar about the convection rolls measured above about 100 Hz. At first sight this effect is best described as a somewhat unsatisfactory optical contrast of the images. In order to quantify this statement, we measured the contrast of an image as a function of the orientation of the polarizer—with no analyzer used in this case. The contrast is defined as A_0/I_0, where A_0 is the amplitude of the first harmonic of the spatially periodic light intensity modulation, and I_0 is the mean value of the light intensity. This method is well understood for planarly aligned nematics in the normal roll case. There, the contrast is high when the director is aligned parallel to the polarizer, and almost zero when the director is aligned perpendicular to the polarizer, as shown in Figure 6. For normal rolls in the homeotropic case, one would expect similar behavior.[4] As indicated by Figure 7(a), which was measured at 30 Hz and at 5.76 V, this seems to be the case: The contrast is indeed low when the polarizer is parallel to the roll axis. Thus these rolls can consistently be interpreted as normal rolls, although this fact seems to be in disagreement with the theoretical calculations. If the rolls were oriented parallel to the director, they would not be expected to change its direction and, thus, they would be invisible.

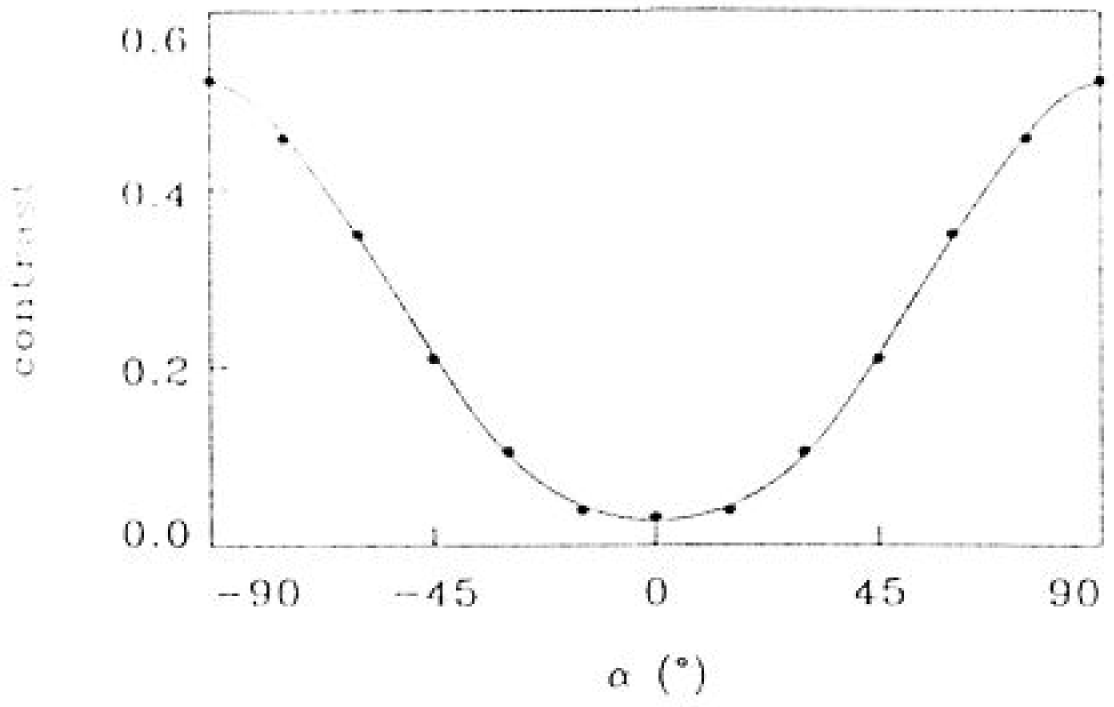

FIGURE 6 Optical contrast of normal rolls in planar alignment.

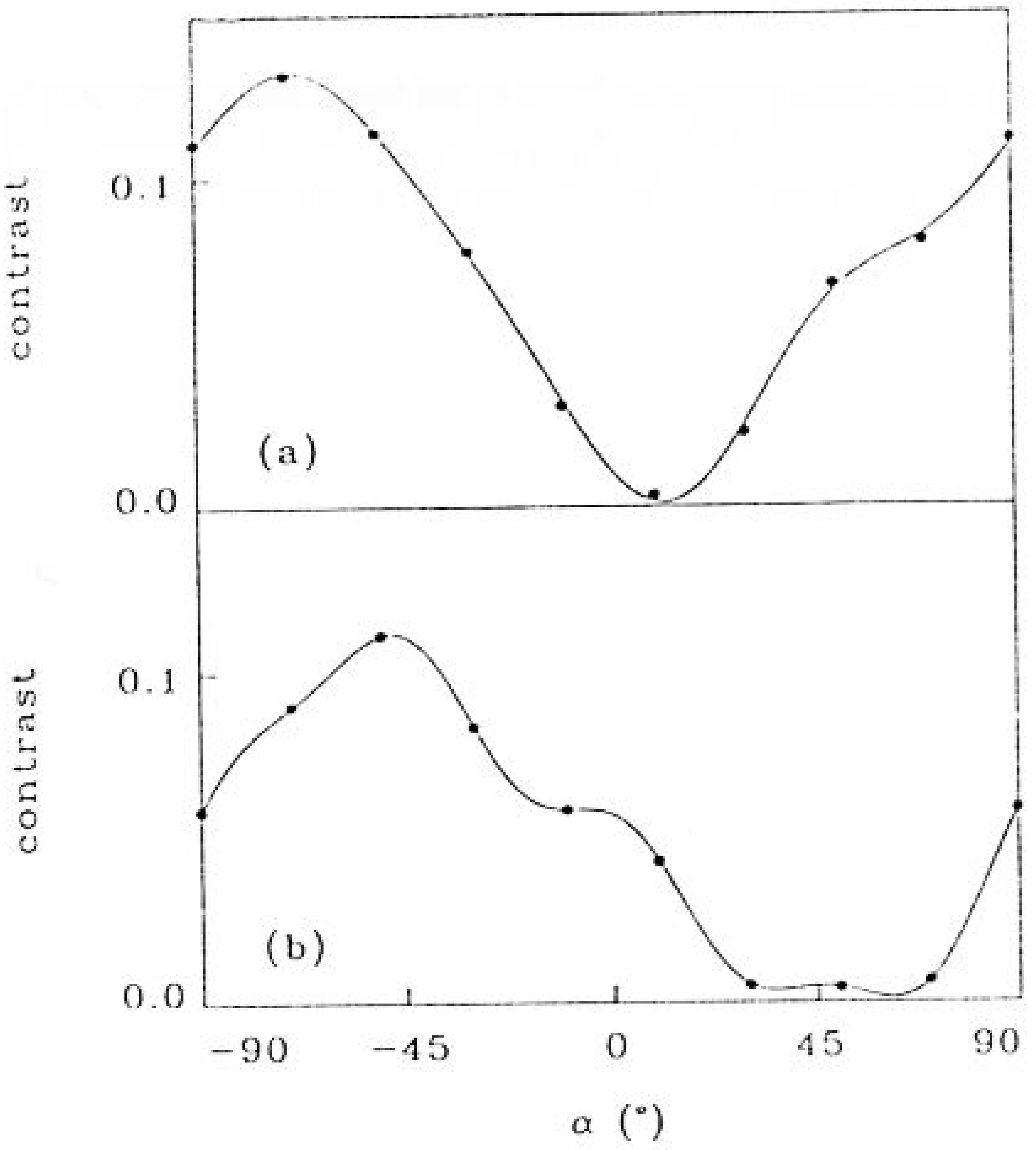

FIGURE 7 Optical contrast of convection rolls in homeotropic alignment, (a) 30 Hz, 5.76 V, (b) 1000 Hz, 6.64 V.

The strangest behavior observed so far is shown in Figure 7(b): These rolls do not behave like normal rolls. They have a minimal contrast when the polarizer has an angle of approximately 45° with the roll axis, and a maximum when this angle is about −45°. Thus, these rolls are not oblique rolls in a usual sense, because they appear to be aligned normal to the mean orientation of the director. They are not regular normal rolls, either, because they have obviously broken the symmetry of the normal rolls. We thus call them abnormal rolls.

There is no ideas about the geometrical shape of the abnormal rolls. They do not seem to agree with the prediction from the linear stability theory. As mentioned by Hertrich et al.,[2] however, the linear stability theory in the case of oblique rolls might be of limited use, because the ensuing mode is unstable, roughly speaking with respect to rotations of the roll. Thus, the abnormal rolls might already be some manifestation of this instability of the linear solution. In order to make closer contact to the theory, we tried to measure the most unstable mode by analyzing subcritical thermal fluctuations below the onset of electroconvection, a method which has been successfully applied in the planar case.[6] The result is shown in Figure 8 and must be considered as negative: No obvious pattern can be observed. The reason for this difficulty in observing the unstable mode is unclear: The optical sensitivity is expected to be as twice as high compared to the planarly aligned sample.[4]

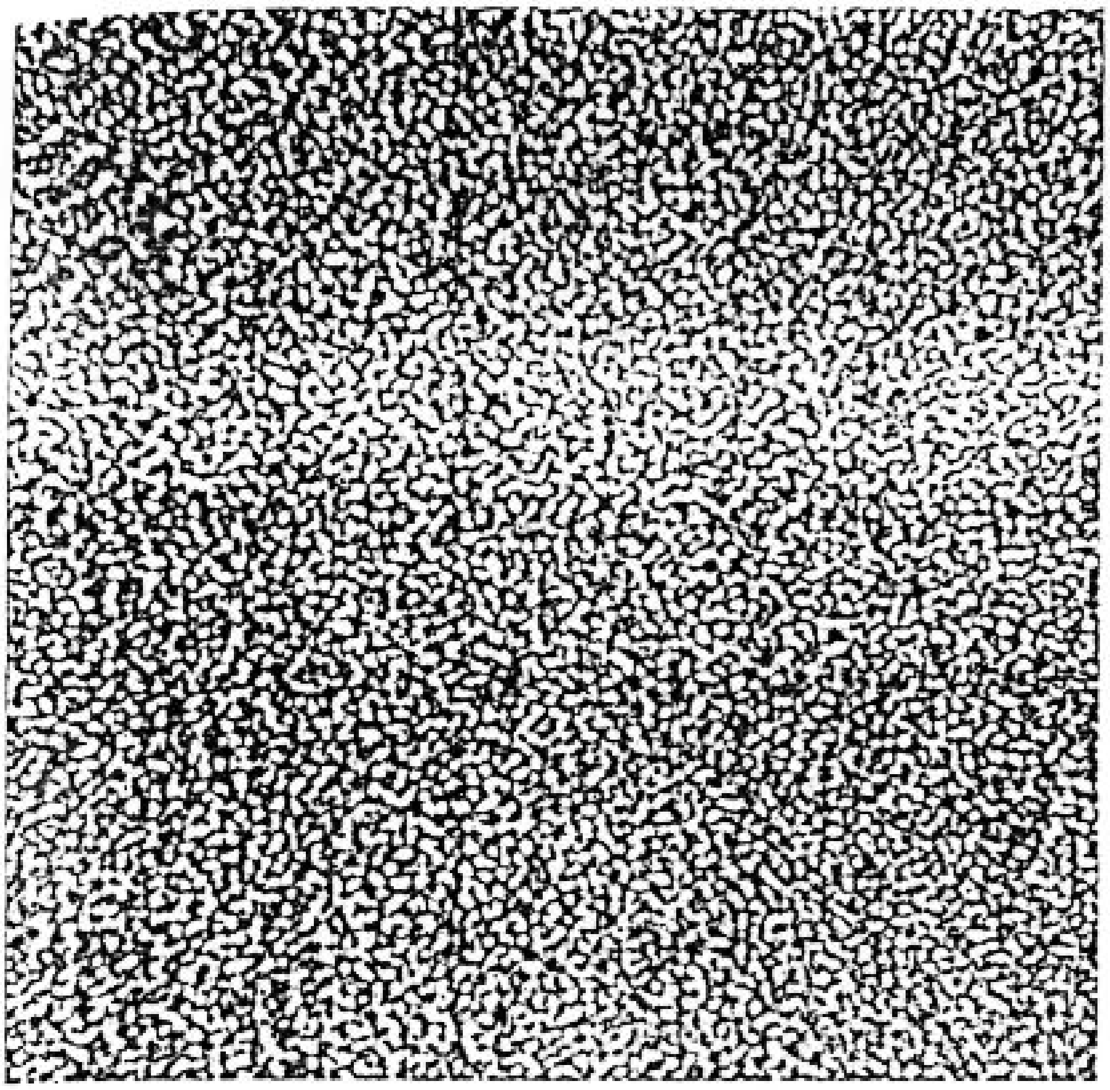

FIGURE 8 Attempt to observe subcritical convection patterns, $\epsilon \approx -0.1$, 30 Hz (28°C).

In summary, there seems to be a qualitative disagreement between the theoretical predictions and experimental observation in this interesting system. One speculation we would like to investigate in the near future is the question of the possible time dependence of these patterns: Since they appear on an already tilted director orientation, the electric ac-field might induce oscillations of this field phase-locked with the driving frequency. This effect is also present in planarly aligned normal rolls, where it is enhanced when approaching the cut-off frequency,[5] but it might be stronger in the present case where the convection is an instability originating from a less trivial ground state. The natural way to explore the time dependence would be stroboscopic illumination, a method that is harder to implement here because of the large light absorption due to the chrome coating.[1]

Finally, we would like to mention that spiral convection patterns are observed in our cell as well (two images have been presented by Pesch et al.[3]). Before exploring their behavior in detail, a clarification of the nature of the most unstable mode at the onset of convection seems to be the most pressing task.

ACKNOWLEDGMENTS

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