

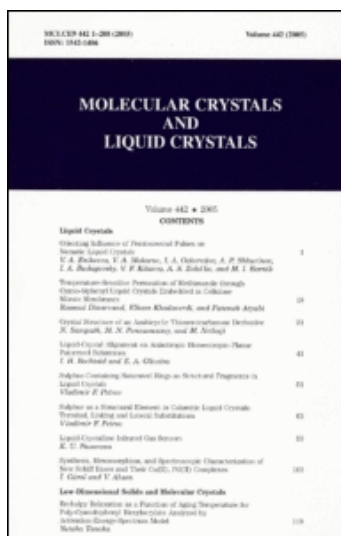
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## Molecular Crystals and Liquid Crystals

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### The Electrooptic Characteristics of a Nematic Liquid Crystals Cell with Asymmetrically Treated Electrodes

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# The Electrooptic Characteristics of a Nematic Liquid Crystal Cell with Asymmetrically Treated Electrodes

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We present electrooptic measurements on sandwich nematic liquid crystal cells with asymmetrically coated electrodes. The cells have one electrode coated with silicon monoxide at an oblique angle and the other is either rubbed or coated with aluminium oxide. Most of the observations have been made on materials with positive dielectric anisotropy, and a few observations on materials with negative dielectric anisotropy. With salt dopants, there is vigorous electrohydrodynamic motion under an applied field beyond a threshold value if the SiO-coated electrode is positive. The motion is subdued when the voltage is reversed. If the medium is viewed between crossed polarizers, there is a considerable change in the transmission coefficient when the polarity of the exciting voltage is reversed. The phenomenon is attributed to the fact that it is mainly the positively charged ions that carry the current in such cells. If the medium is doped with an electron acceptor, the situation is reversed since in this case negatively charged ions carry the current. Both DC and AC responses are reported.

## 1 INTRODUCTION

Electrohydrodynamic effects on nematic liquid crystals with negative dielectric anisotropy have been studied extensively. The phenomena have also been well understood theoretically. On the other hand, observations on electrohydrodynamic effects in liquid crystals of *strong* positive dielectric anisotropy ( $>2$ ) are relatively few. Recently flow patterns have been observed under applied electric fields in such substances.<sup>1-3</sup> Depending upon the voltage and frequency, either a finger-print or a spherulite pattern is observed in such cases. Barnik *et al.*<sup>2,3</sup> have observed only a field threshold in materials with dielectric anisotropy up to  $+3$ , and they describe the instability regime as

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† Presented at the Seventh International Conference on Liquid Crystals, Bordeaux, July 1-5, 1978.

corresponding to the well known "dielectric" regime of negative dielectric anisotropy materials.<sup>4</sup> Further, they have noticed that the threshold field for instabilities agree with the value observed even for the isotropic phase, and hence they have concluded that the instability does not essentially depend upon the dielectric anisotropy. Thus the anisotropy of the medium is useful only in the visualization of the phenomenon, rather than in causing it. We noticed recently that cyanobiphenyls which have a dielectric anisotropy of about +10 also give rise to similar electrohydrodynamic phenomena. Further, we observed an interesting polarity-dependent electrooptic effect when the two electrodes were treated asymmetrically. The rectification action in similar liquid crystal cells with asymmetrically treated electrodes has been reported recently by Sussman.<sup>5</sup>

## 2 EXPERIMENTAL

We used the standard sandwich geometry for the observations. The liquid crystal is contained between two tin-oxide coated glass plates, separated by mylar spacers. The thickness of the cell was measured using channel spectra before the liquid was filled in, and was usually in the range 10–25  $\mu$ . We tried a few different methods of treating the electrodes asymmetrically. Usually, one of the electrodes was coated with SiO at an oblique angle. We used only the "high" angles of coating, viz., 30–35° so that the electrode would orient the liquid crystal with its director practically parallel to the glass plate.<sup>6</sup> The thickness of the coating was about 300–500 Å. In many studies, the other electrode was rubbed unidirectionally with a filter paper. The resulting cell is similar to the one used by Sussman<sup>5</sup> in his experiments. We also tried to coat the second electrode with different materials. We found that aluminium oxide (Al<sub>2</sub>O<sub>3</sub>) was a useful material to get a good alignment and reproducible results.

We have made observations on 4'-*n*-pentyl-4-cyanobiphenyl (5CB), 4'-*n*-heptyl-4-cyanobiphenyl (7CB) and E7, a mixture of cyanobiphenyls and cyanoterphenyls marketed by BDH. The results are similar in all the cases.

The observations were made through a Spencer polarizing microscope using a sodium lamp as the source of light. The electrooptic response was also recorded using an MRD 810 phototransistor of Motorola Company, and the signal was amplified in the DC studies using a Keithley microvoltmeter, Model 155. The DC current through the sample was measured using a Keithley electrometer, Model 610C. Both the current and the electrooptic response were recorded on a Rikadenki 3-Pen recorder, Model No. B-381H. In the case of AC studies, we used a helium-neon laser (Spectra Physics,

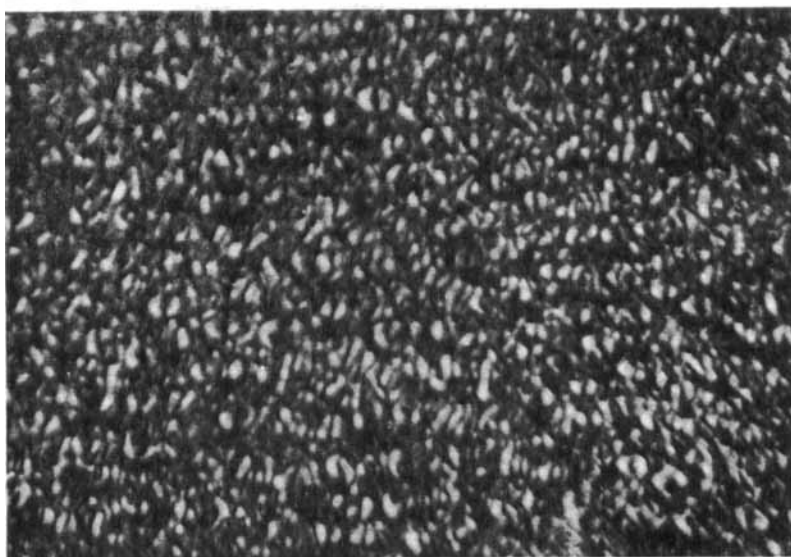
Model No. 133) as the source of light. The output of the phototransistor was amplified using a Burr-Brown amplifier (No. 3640) and displayed on an ECIL storage oscilloscope, Model No. OS768-S.

The sample area was about 1 square centimeter and the typical sample resistance was a few megohms.

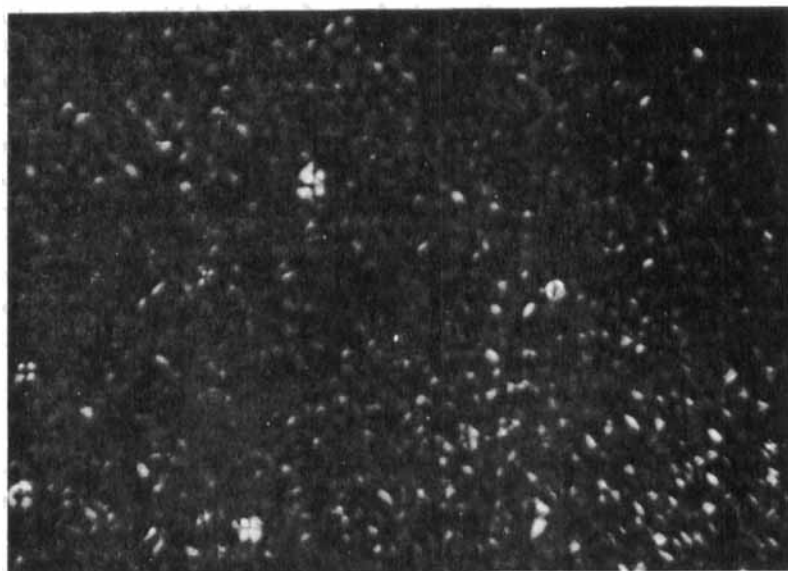
### 3 RESULTS AND DISCUSSION

We will consider the cell with one of the electrodes coated with SiO and the other one rubbed. The liquids we used already contained some (unknown) conducting impurities so that the resistivities were in the range of  $10^9$ – $10^{10}$  ohm-cm. In these cases, we did not add any further impurities to enhance the conductivity. Starting with the homogeneously aligned sample, we found that as the DC voltage was increased, initially the director pattern was distorted (Freedericksz transition) at about 1.2 V. Then, depending on the thickness and conductivity of the sample, the electrohydrodynamic instability started beyond a voltage of about 5 V. However, since the threshold is a field threshold, the phenomenon could be seen even at 2 V for thin samples ( $\sim 6 \mu$ ). The pattern was usually in the form of small bubbles, but sometimes the finger-print texture noted by earlier workers<sup>1–3</sup> was also seen. The bubbles which look like maltese crosses between crossed polars were caused by a circular fluid motion in a plane perpendicular to the field of view.<sup>3</sup> The hydrodynamic motion was distinctly more vigorous when the SiO-coated electrode was positive than when it was negative. As was found by Sussman,<sup>5</sup> there was an asymmetry in the DC current also. The current was somewhat higher when the SiO-coated surface was positive than when it was negative. However, we must mention that the asymmetry in current (or rectification) was not as high as reported by Sussman in his system which used a Schiff-base formulation and indium oxide coated electrodes. Also, we have not tried to normalise the current measurements with reference to the in-phase value, as was done by Sussman. The difference in the electrooptic response was made very obvious by placing the cell between crossed polaroids. When the SiO-coated side was positive, the vigorous electrohydrodynamic motion allowed a considerable amount of light to pass through the system so that the field of view appeared bright (Figure 1(a)). On the other hand, when the SiO-coated side was negative, the field of view more or less remained crossed out (Figure 1(b)). Hence, there was a polarity-dependent electrooptic response, which is obviously of interest for possible applications.

We also studied the polarity dependent response by observing the scattering of a laser beam by the sample. Under proper conditions, with SiO-coated electrode positive, there was a strong scattering of light due to the motion



(a)



(b)

**FIGURE 1** Photomicrograph of the electrohydrodynamic pattern in a cell containing E7 under DC excitation. Voltage applied 30 V. The cell is viewed between crossed polarizers. Magnification  $\times 200$ . (a) Silicon monoxide coated electrode positive; (b) Silicon monoxide coated electrode negative.

in the medium. Further, the spherulite-like flow pattern gave rise to a scattering in the form of a ring. The ring diameter corresponds to the spherulite size being equal to the thickness of the sample. When the voltage was reversed, the scattered light disappeared since there was no noticeable flow in the medium.

We studied the contrast and the current asymmetry as a function of the applied DC voltage. The method was to reverse the polarity using a DPDT switch. The current usually had a transient response which decayed in a few seconds to a steady value. The electrooptic response had a more complicated behaviour. In some samples, immediately after the field was reversed (to make the SiO-coated electrode positive), the optical transmission coefficient increased to a high value. Over tens of seconds, it reduced to a steady value (Figure 2). In some other cases, the increase was not so high but in course of time the intensity gradually attained a steady value (Figure 3). The optical contrast between the positive and negative voltages increased as the voltage

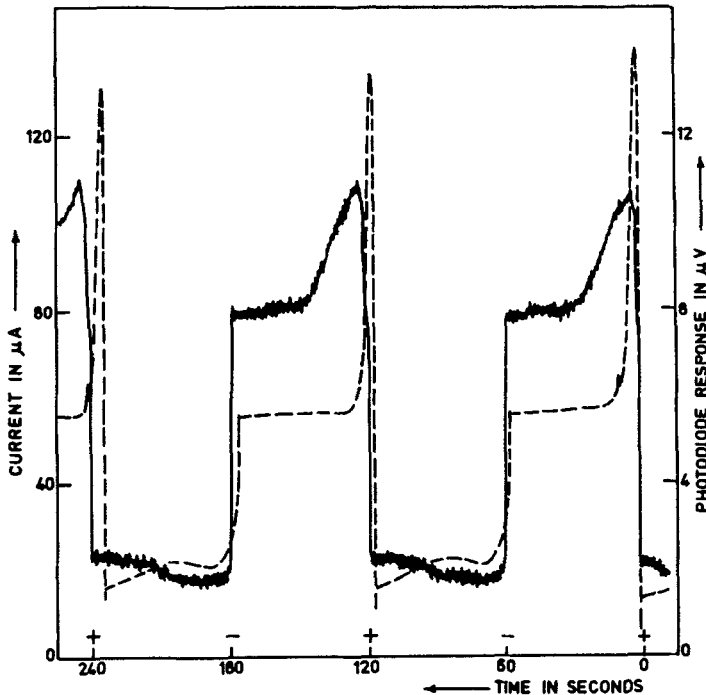


FIGURE 2 Electrooptic response (continuous line) measured by a photo-diode in a cell with one electrode coated with SiO and the other rubbed. The dashed line indicates current. The + and - signs at the bottom of the figure stand for the DC polarity to which the SiO-coated electrode has been switched. Thickness of the cell  $\sim 15 \mu$ . Voltage applied 40 V.

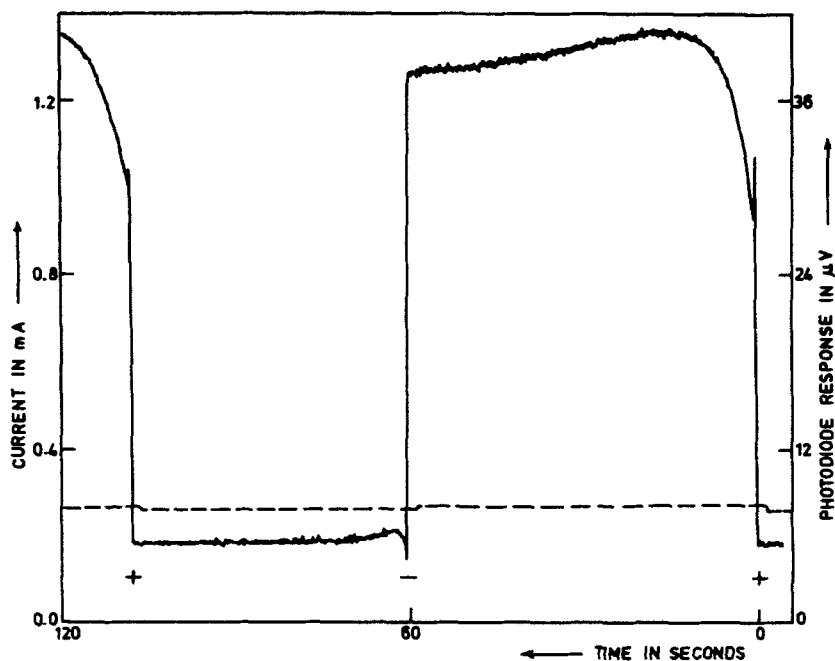


FIGURE 3 The electrooptic response in another cell with SiO-coated and  $\text{Al}_2\text{O}_3$ -coated electrodes containing E7. Applied voltage 15 V. Other details as in Figure 2.

is increased initially and an optimum contrast was obtained at about 15–20 V (for sample thickness in the range 15–20  $\mu$ ). If the sample is very thin (< 10  $\mu$ ) the maximum contrast is obtained at very low voltages—even as low as about 4 V.

A further increase in voltage usually reduced the transmitted intensity, for both the signs of the applied voltage. The current in both the situations increased with the applied voltage. However, the current asymmetry usually decreased with increasing voltage. If the sample was held at a high voltage (40 V or above) for a long time, one could usually observe a damage to the SiO coated surface and hence a deterioration in the alignment of the sample. (In one or two samples where the conductivity was high, good contrast was obtained only at high voltages, i.e., beyond about 40 V).

### AC Studies

We have also examined the electrooptic response under AC excitation of the cell. Most of the studies were made at 20 Hz since the response was good at that frequency. As the frequency was increased, the threshold field for electrohydrodynamic instability increased<sup>2,3</sup> and further, the maximum

contrast that could be obtained decreased. However, we could observe a small asymmetry in the electrooptic response even at a frequency of 1.5 KHz.

At 20 Hz, usually the best response was obtained when the cell was viewed between crossed polarizers, set at  $45^\circ$  to the field-free orientation direction of the director. Figure 4 illustrates the response observed in one such experiment. Square wave excitation normally gave a somewhat poorer response (Figure 5). Figures 4 and 5 illustrate the response at different voltages. Usually there are variations with time in both the DC and AC responses. However, the AC response in particular stabilises after sometime and then does not vary even after continuously operating the cell for several hours. We have also tested the repeatability of the response after resting the cell for a day or two.

The maximum contrast ratios that we could obtain are:

1 : 8 in DC excitation, and 1 : 4 in AC excitation (20 Hz),

where we have defined the contrast ratio as the ratio of the peak responses in the two halves of the exciting voltage. Perhaps a more appropriate measure would be the ratio of the integrated intensities in the two halves, i.e., the areas under the response curves (see Figures 4 and 5). In that case also, the maximum contrast ratio is about the same.

We tested the twisted nematic configuration for observing the asymmetry. There were no particular improvements in this case.

We tried other coatings for observing this effect. Aluminium oxide was found to be quite useful; when it is coated at an oblique angle ( $30-35^\circ$ ), it gives a good alignment of the sample. If the coating is thin, and the other electrode is rubbed, the electrooptic asymmetry of the cell is very small. Hence we studied cells with one electrode coated with SiO and the other with  $Al_2O_3$ . Such cells give better reproducibility of the results compared to the SiO-coated and rubbed system, but the transmitted intensity was somewhat less.

We also tried the experiment with two electrodes of different resistances. If one of the electrodes had a relatively high resistance ( $\sim 10\text{ K}\Omega$ ), we could again notice a slight asymmetry in the electrooptic response.

Sussman has concluded, based on detailed observations on the rectification action of the cells with SiO-coated and rubbed electrodes, that negative ions are electrosorbed more efficiently at the positive electrode than the positive ions at the negative electrode. Presumably, this arises because the positive and negative ions have different rates of electrode-reaction. Hence the current is mainly carried by the positive ions. When the SiO-coated electrode is positive, the full area of the uncoated negative electrode is available for the current flow. On the other hand, when the SiO-coated electrode is negative, the positive ions see effectively a reduced area of the electrode and hence the measured current density is lowered. This gives an



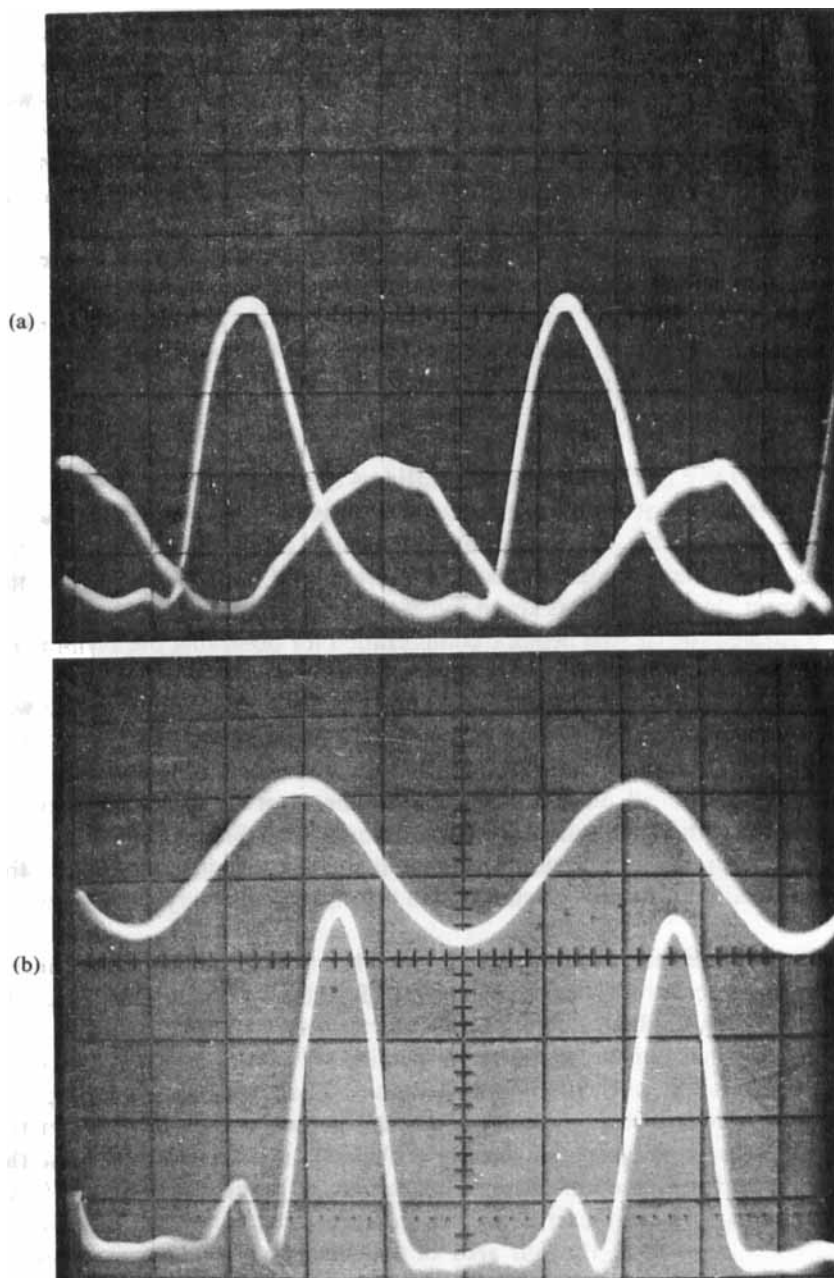


FIGURE 4 Electrooptic response as seen on an oscilloscope in the case of 20 Hz sine-wave excitation of a cell containing E7. One of the electrodes was coated with SiO and other was rubbed. Thickness  $\sim 12 \mu$ . The zero level for the electrooptic response is the bottom most line. The applied sine-wave voltage is also shown. (a) RMS voltage, 6 V; (b) RMS voltage, 10 V.

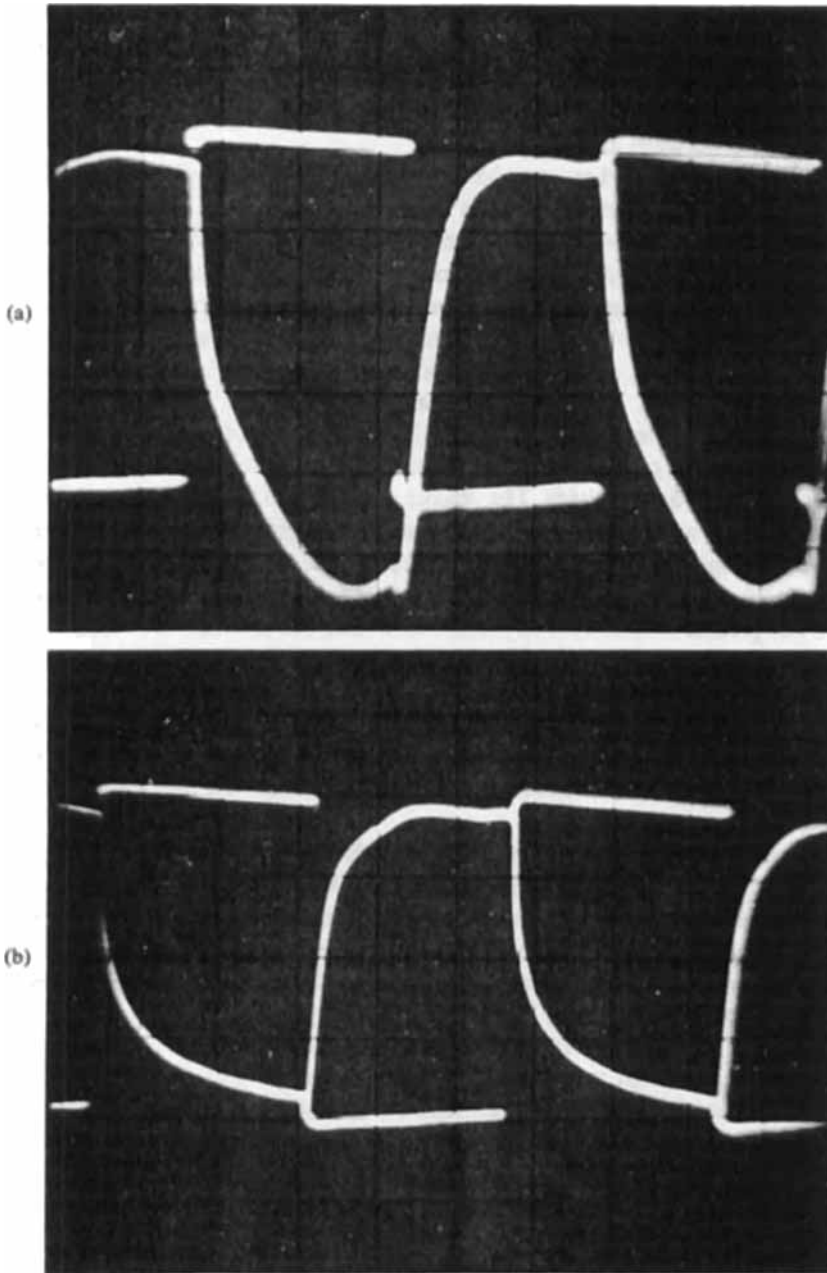


FIGURE 5 The electrooptic response of a cell similar to the one used in Figure 4 under square-wave excitation (20 Hz). Thickness of the cell  $\sim 7 \mu$ . The exciting voltage pattern is also shown. (a) Applied voltage, 2V; (b) Applied voltage, 4V.

explanation of the rectification action in such cells. It also explains our observations on the electrooptic effect. The greater current flow when the SiO-coated side is positive obviously gives rise to a greater electrohydrodynamic flow and hence the greater intensity of light transmitted through the cell placed between crossed polaroids.

As mentioned earlier, we do not know the nature of the conducting impurities in our sample. If, as is likely, the impurity is of a salt type, the liquid crystal molecules are also expected to take part in the conduction mechanism.<sup>7-9</sup> In order to test the explanation given above, wherein positive ions contribute mainly to the current flow, we wanted to have a system in which negative ions mainly contribute to the current flow. For this purpose, we dissolved 1-2% of an electron acceptor, viz., 7,7',8,8'-tetracyanoquinodimethane (TCNQ). It is now well established that with such redox dopants, which have (in the case of acceptors) a reduction potential which is much lower than that of the liquid crystal, the current flow will mainly take place through the redox dopants. The charge acceptors pick up negative charges at the cathode, and the negative ions thus formed get injected into the sample. Hence we might expect that with the doping, the electrooptic response would be reversed. And it is indeed what is observed. With SiO-coated side *negative*, we get greater electrohydrodynamic flow and the contrast is reversed. Thus for instance, the scattering of the laser light is much stronger when SiO-coated side is negative. Further, when we dope the sample with a salt like cetyl pyridinium bromide, the usual polarity dependence of the electrooptic response is observed, i.e., the field of view is brighter when SiO-coated electrode is positive. These observations establish the mechanism of the effect discussed earlier.

As mentioned in the introduction, the electrohydrodynamic instability of the type we have studied is not peculiar to liquid crystals. The instabilities are seen even in isotropic liquids and arise from electroconvective processes.<sup>2</sup> The instability becomes easily discernible in the nematic liquid crystal because of its optical anisotropy. The dielectric anisotropy of the material does not have a major role to play.

Indeed the asymmetry of electrohydrodynamic response of the liquid crystal in sandwich cells similar to the ones described above can be seen even in materials with negative dielectric anisotropy. However, it should be noted that in this case, at low frequencies, we have the 'conduction' regime, unlike the positive dielectric anisotropy materials, which are in a situation analogous to the 'dielectric' regime which occurs in negative dielectric anisotropy materials only above a critical frequency.

Further experiments are in progress with different dopants and liquid crystal materials with a view to improving the electrooptic response of the asymmetrically treated cells.

**Acknowledgment**

We are deeply indebted to Professor S. Chandrasekhar for several useful suggestions.

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