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# Non-Uniform Distortions During Electric Field Induced Unwinding of a Cholesteric Liquid Crystal

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# Non-Uniform Distortions During Electric Field Induced Unwinding of a Cholesteric Liquid Crystal

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Direct experimental evidence is reported for non-uniform distortions of a cholesteric array undergoing an electric field induced nematic transition. The effect is most pronounced when sample thickness is comparable to the pitch, when the helix axis is parallel to the walls, and when the electric field is applied perpendicular to the helix axis. Thicker samples display helix expansion as a uniform process as field is applied, in accord with theory, and similar to the experimentally observed thermal pitch distortions.

## INTRODUCTION

An important field effect in cholesteric liquid crystals of positive diamagnetic or positive dielectric anisotropy is the unwinding of the cholesteric helix, often called the cholesteric-nematic phase transition[1]. For the case in which an electric or a magnetic field is applied perpendicular to the helix axis,[2,3] it has been shown in several experiments[4-7] that helix unwinding occurs as a continuous process—the helix expands in a given field range followed by a conversion to a nematic phase. A theory for this continuous phase transition was first presented by de Gennes[2] for the case of a magnetic field, and all the experimental evidence has supported this theory.

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Qualitatively the theory can be understood as follows: starting with a uniform helical structure of positive diamagnetic anisotropy with a magnetic field applied perpendicular to the helical axes, at low fields the structure is in a low energy configuration. At higher field strengths, however, a lower energy structure can be achieved by unwinding the helix so that at every point the molecular axis is parallel to the field. Since this requires overcoming elastic forces between the molecules, and therefore costs energy, it does not occur until a critical high field is reached. But at intermediate fields, a distortion of the helix minimizing the sum of elastic and field energies should occur.

In treating the nature of the distortion of the helical array, it has been pointed out[3] that the general form of the perturbed helix is

$$\mathbf{L} = (\cos\theta, \sin\theta, 0), \quad \theta = tZ + f(Z)$$

where $\mathbf{L}$ is the unit vector parallel to the axis of molecular alignment, $\theta$ the angle between the local director vector and the x axis for a helix of pitch $Z_0 = 2\pi/t$, and $f$ has the period $\pi/t$. This implies that the angle between successive layers in the helical array may be non-uniform when a field is applied. Studies of the field induced distortion have focused on the observation of the optical properties of cholesteric structures in the planar texture.[4-7] In these measurements, it would be difficult to distinguish between uniform and non-uniform distortions, since one effectively measures an average pitch or average bending angle. It should be possible to probe the pitch distributions by examining the appearance of higher-order bands in the reflection spectrum,[8,9] but studies of this type have not as yet been reported. Other evidence dealing with the general validity of the de Gennes[2] and Meyer[3] prediction of the form of the magnetic field induced distortion has been obtained by studying the transition using an electron spin resonance technique,[10] and by measuring the diamagnetic susceptibility through the transition.[11]

The purpose of this study was to focus on boundary conditions where non-uniform distortions might be very pronounced, namely on thin samples where walls couple very strongly with the first few molecular layers. Dreher[9] has recently considered this boundary problem and points out that when a cholesteric is located between fixed boundaries, the spatial period of the helical structure is not necessarily altered when a field is applied (although within that spatial period the molecular alignment is indeed altered). In order to allow direct optical observations of the nature of the pitch distortions, we chose to align the helix axis parallel to the walls and apply an electric field perpendicular to the helix axis.

Such an alignment allows one to observe the so-called "fingerprint"[12] or domain[13] texture and to follow the field induced distortion directly by microscopic observation. This type of experiment has been reported by Meyer[4] for a magnetic field distortion of a p-azoxyanisole (PAA) sample doped with cholesteryl acetate (CA). In the experiments described below, we report the electric field and thermal pitch distortions of a compensated mixture of cholesteryl chloride (CC) and cholesteryl myristate (CM).[6,14]



## EXPERIMENTAL

The cholesteric liquid crystal employed in this study was a 1.75:1 mixture of CC and CM whose preparation and properties have been previously described.[6,14] When glass or quartz is treated with a silane coupling agent[15] such as NN-dimethyl-N-octadecyl-3-aminopropyltrimethoxysilyl chloride (DMOAP) or with lecithin, homeotropically aligned nematic or cholesteric thin films can be achieved. In this study, tin oxide-coated quartz discs treated with lecithin allowed the preparation of homeotropically aligned CC:CM films at thickness up to 50 $\mu$. In such an alignment, the helical axis is parallel to the substrate, and the molecular axis twists from a perpendicular to a parallel orientation. The net result is a "fingerprint" pattern such as that shown in Figure 1. The fingerprints are explained as follows:[13] viewed normal to the glass plates, the optic axis of the molecule precesses periodically from a direction parallel to the direction of viewing to a direction normal to the direction of viewing. Thus, between crossed polarizers, one observes a series of bright and dark lines forming fingerprints whose spacing is precisely one-half the pitch of the helical array. These fingerprints can be observed and photographed under a polarizing microscope; variation of electric fields and temperature are then studied in an apparatus previously described.[16]

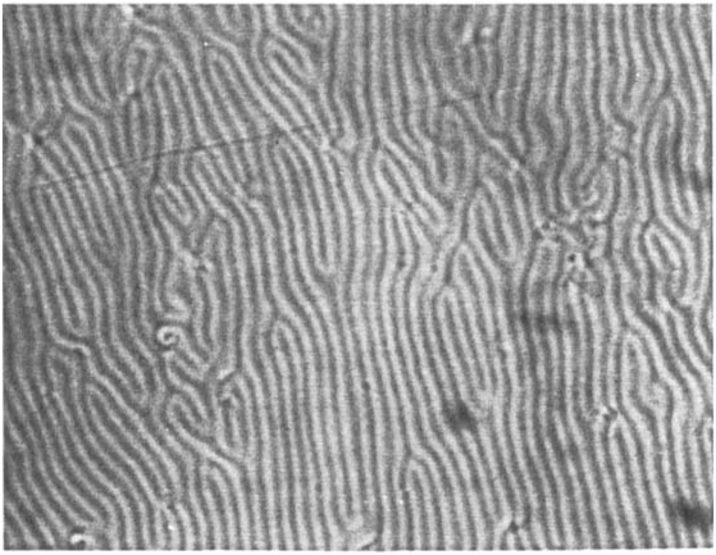
## RESULTS

### Thermal variation of pitch

Figure 1 indicates the variation of pitch with temperature which is observed by following the fingerprint pattern. The pitch-temperature dependence is plotted in Figure 2 for samples of several different thickness and compared with pitch measurements performed on CC-CM in the planar texture by measuring optical rotatory power.[6]

The results of measurements on homeotropically aligned and planar texture CC:CM are in good qualitative agreement – pitch increases continuously with temperature up to a "compensation temperature"[6] – usually called $T_{nematic}$ where the pitch is infinite and then decreases again until the isotropic transition temperature is reached. The quantitative agreement is poor, the apparent pitch in the homeotropically aligned samples being higher than in the planar texture.

The problem of the accuracy of pitch determinations from the fingerprint



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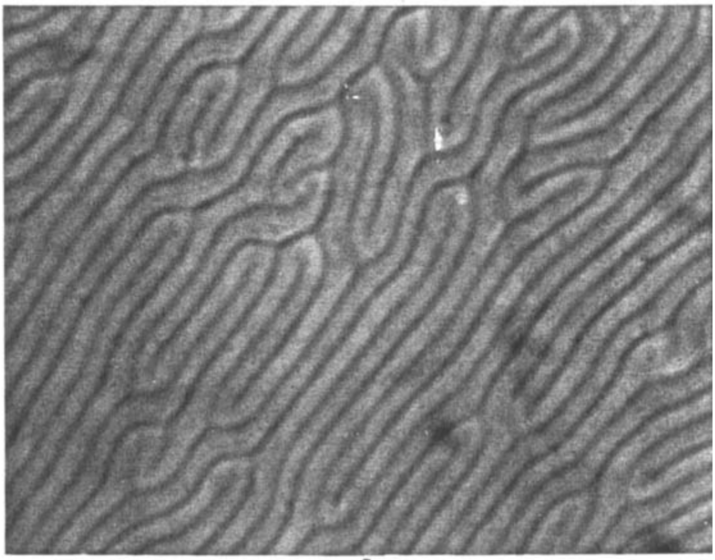

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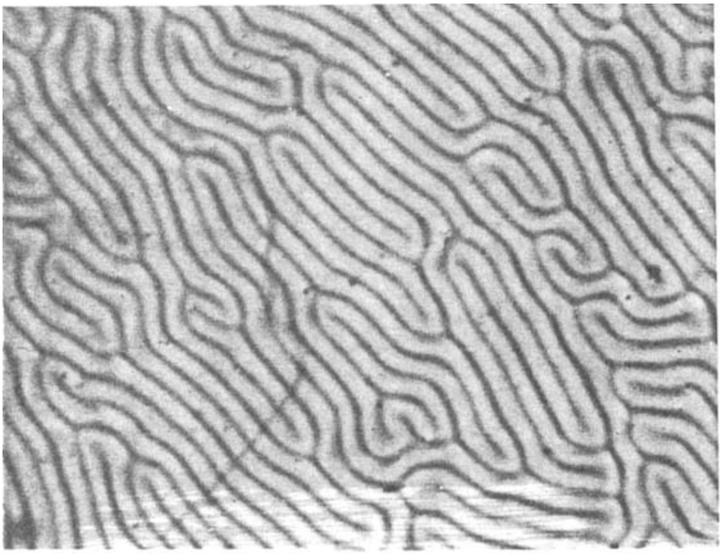
FIGURE 1 Temperature variation of the fingerprint pattern of a 12.7 $\mu$ thick sample of CC:CM: (a) 32°, Z = 14 $\mu$; (b) 36°, Z = 32 $\mu$; (c) 43°, Z = 28 $\mu$; (d) 48.5°, Z = 18 $\mu$.



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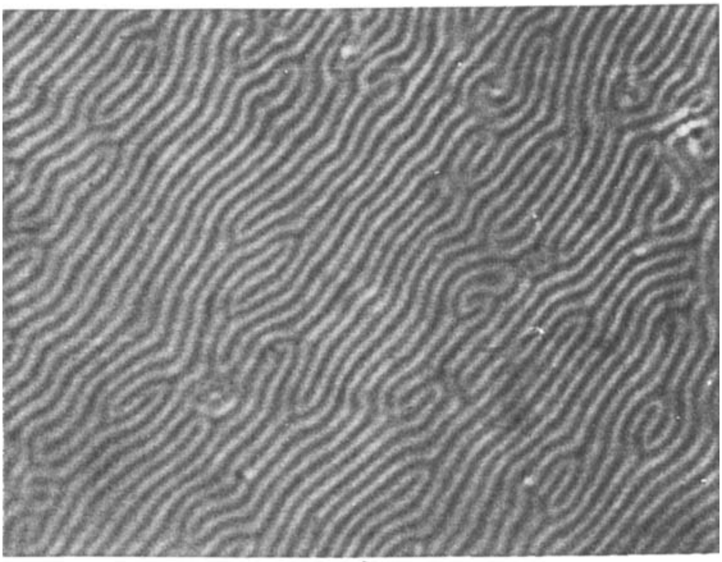

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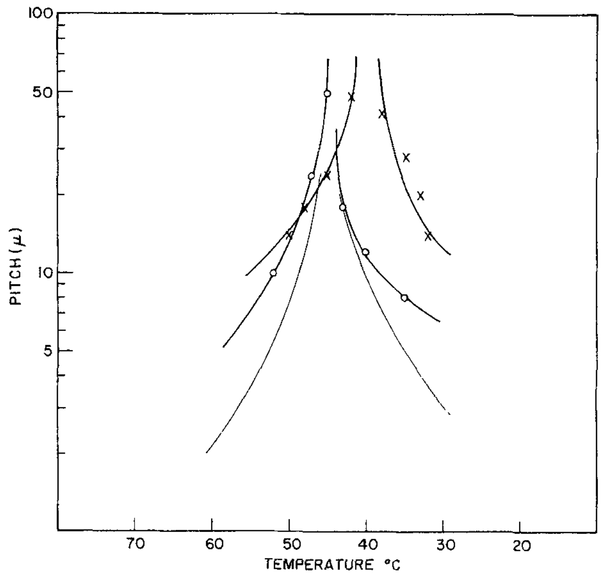



FIGURE 2 Temperature dependence of the pitch of CC:CM. Measurements from fingerprint pattern: o, 50 μ sample; X, 12.7 μ sample. The solid line represents measurements of pitch by the optical rotatory power method[6] on a 12.7 μ planar textured sample.

patterns has been examined by Cladis and Kleman.[13] Difficulties are known to arise when sample thickness and pitch are approximately the same. Further difficulties can arise if the helix axis is tilted at some angle with respect to the walls. In spite of these quantitative difficulties, this experiment confirms that the qualitative nature of helix expansion and contraction in the thermal process can be *directly* observed by microscopic observation of the fingerprint pattern.

### Electric field variation of pitch

Figures 3 and 4 present the variation in the fingerprint pattern when a 1 kHz ac electric field is applied perpendicular to the helix axis. When the thickness of the sample is much greater than the pitch (Figure 3), the results are in good agreement with the thermal variation behavior – pitch increases continuously until the electric field exceeds a critical value $F_c$ (Table, 1); above $F_c$ the sample appears black between crossed polarizers, as one would expect for a homeotropically aligned nematic. The results are also in agreement with Meyer's observation[4] on a 130 μ thick PAA-CA sample.

However, when the thickness is comparable to the pitch (Figure 4), in distinct contrast to the thermal variation behavior, the distortion appears non-uniform. Below $F_c$, the distance between the centers of adjacent fingers remains constant, but a portion of the finger is aligned homeotropically and the rest appears undisturbed. The amount which is aligned depends on the field strength.

One can then estimate an effective pitch Z of the helix from the contribution of the two regions – the "destroyed" and "undestroyed" regions (Table 2). If A is the distance occupied by one bright and one black region, and B is the increase in the width of one black line when the field is applied, then $A/(A\text{-}B) = Z/Z_0$.

If one then plots $Z/Z_0$ vs $F/F_c$ where $Z_0$ is the pitch of the undisturbed helix (Figure 5), the results on these thin samples indicate pitch distortions start at a value of $F/F_c$ considerably lower than those observed in planar texture samples[6] or in the 50 $\mu$ thick samples studied in this work.

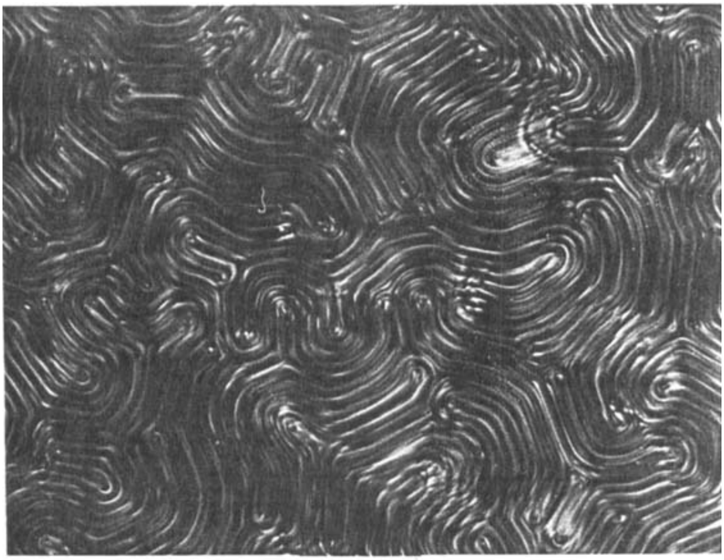
## DISCUSSION

The results above represent, to the best of our knowledge, the first direct experimental evidence for a non-uniform distortion of a cholesteric array undergoing a field induced nematic transition in which the spatial period of the helical structure is not altered. The effect is clearly observed when the sample thickness is comparable to the pitch. Microscopic observations of the destruction and growth of the cholesteric array indicate interesting nucleation and growth phenomena around the disclinations and alignment inversion walls existing in the samples. The destruction of the helix starts at disclinations as can be clearly seen in Figure 4, where several $\tau$ type disclinations[13] can be seen to enlarge as the field approaches $F_c$.

Meyer's published observations[4] on the effect of magnetic fields $H$ on a PAA-CA sample are further elaborated in his thesis;[17] indeed, he argues that in a real sample with the unit vector **L** rigidly defined at the surface, the dynamics of the distortion must be non-uniform. He cites the presence of a few alignment inversion walls in samples where H is slightly greater than the critical field as evidence for this non-uniformity, and as evidence that the cholesteric-nematic transition must be viewed as a nucleation and growth process.

Thus, it is not surprising that samples strongly interacting with the walls would allow the observation of non-uniform distortions. In fact, indirect evidence has recently appeared for non-linear distortions near the walls for a twisted nematic liquid crystal.[18] In order to explain the difference between capacitatively and optically measured magnetic thresholds for untwisting, van Doorn calculates the orientation of the director in a twisted nematic and predicts for thin layers a marked non-linearity.

It is anticipated that such non-linear distortions also occur in planar-texture cholesterics when thickness is comparable to or some small multiple of pitch, a case which is of course very similar to the twisted nematic case. The comparison



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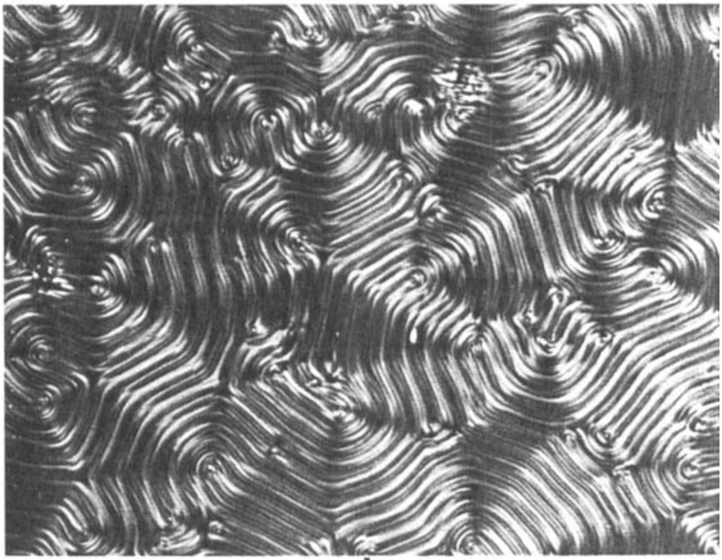

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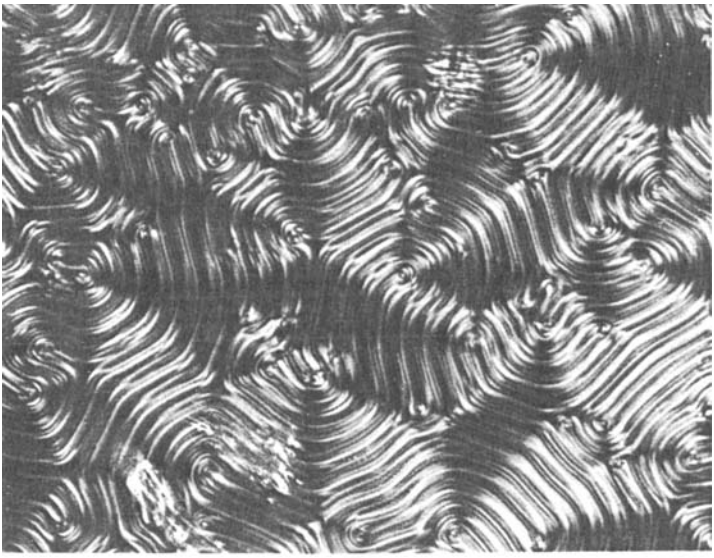
FIGURE 3 Electric field effect on the fingerprint pattern of a 50 μ thick sample of CC:CM at 47°: (a) $F = 1$ kV cm$^{-1}$; (b) $F = 2.9$ kV cm$^{-1}$; (c) $F = 3.3$ kV cm$^{-1}$; (d) $F = 4.2$ kV cm$^{-1}$.



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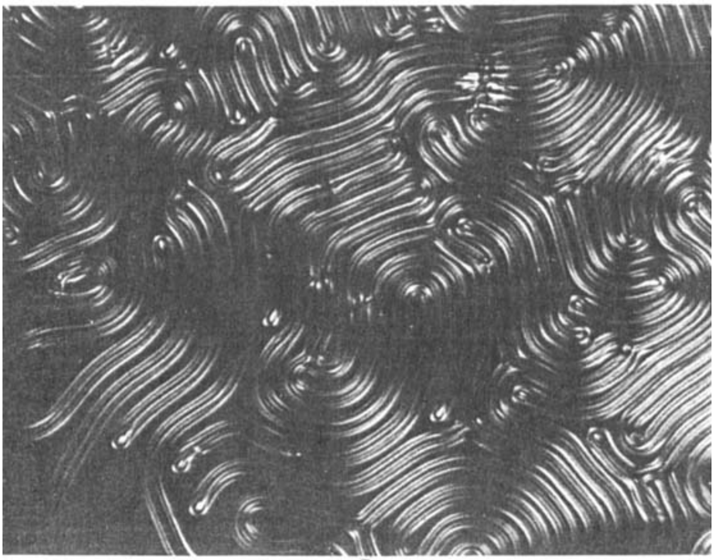

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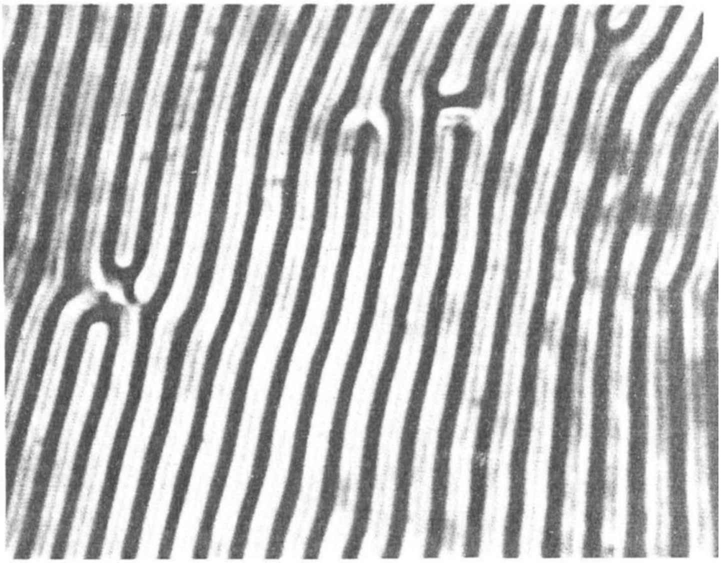


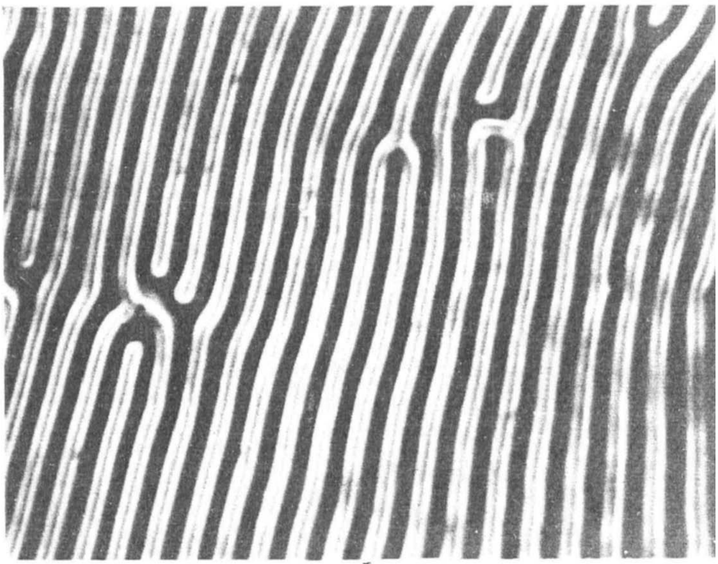
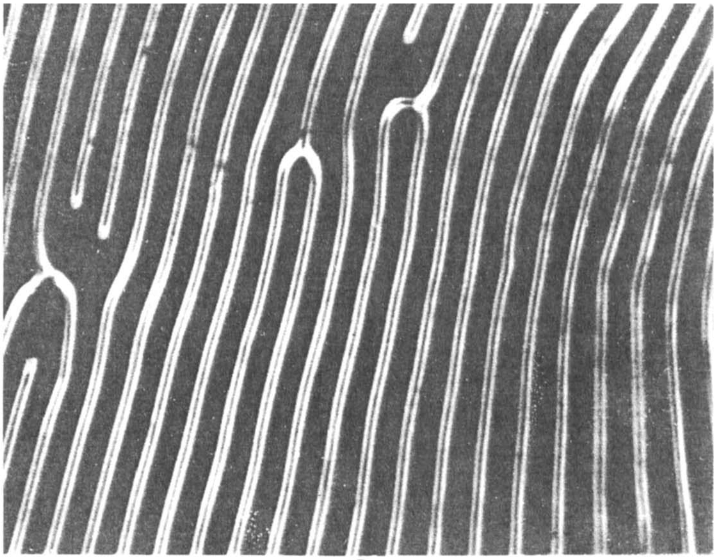




FIGURE 4 Electric field effect on the fingerprint pattern of a 6.3 $\mu$ thick sample of CC:CM at 46.5°: (a) F = 0; (b) F = 1.6 kV $cm^{-1}$; (c) F = 4.8 kV $cm^{-1}$; (d) F = 7.9 kV $cm^{-1}$.



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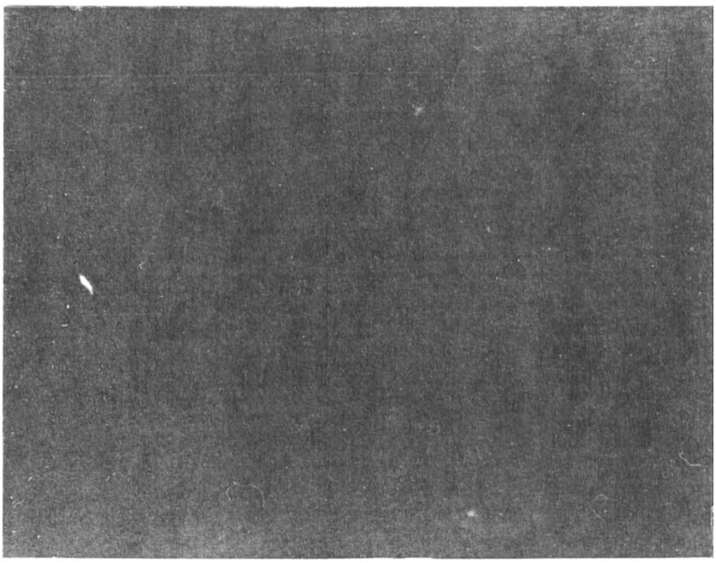

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TABLE 1
Critical field as a function of initial pitch ($Z_0$) for a 50 μ thick homeotropically aligned sample of CC:CM

| Temperature (°C) | $Z_0$ (μ) | $F_c$ (kV cm$^{-1}$) |
|---|---|---|
| 35 | 8 | 15.8 |
| 40 | 12 | 13.8 |
| 43 | 18 | 6.9 |
| 45 | 50 | 2.4 |
| 47 | 24 | 4.4 |
| 53 | 9 | 14.8 |

TABLE 2
$Z/Z_0$ of Non-uniformly distorted 6.3 μ homeotropically aligned Samples of CC:CM at 38.5° as a function of electric field

| $F$ (kV cm$^{-1}$) | $A$ | $B$ | $\frac{A}{A-B} = \frac{Z}{Z_0}$ | $F/F_c$ |
|---|---|---|---|---|
| 0.0 | 2.3 | 0.0 | 1.0 | 0.0 |
| 4.8 | 2.3 | 0.2 | 1.1 | 0.43 |
| 6.3 | 2.4 | 0.4 | 1.2 | 0.52 |
| 7.9 | 2.3 | 0.6 | 1.3 | 0.71 |
| 11.1 | 2.3 | 2.3 | ∞ | 1.0 |

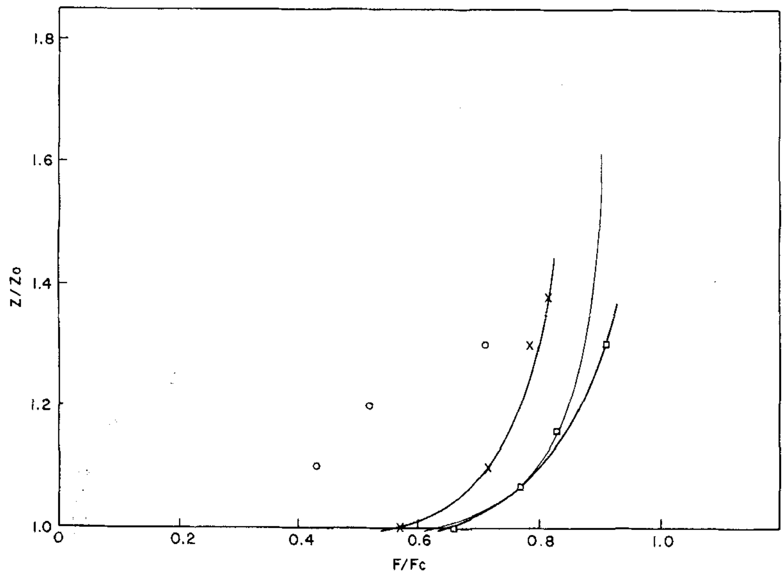


FIGURE 5 $Z/Z_0$ vs $F/F_c$ for CC:CM samples. Solid line, results from previous determination on a planar texture sample;[6] □, data from Figure 3; ×, data from fingerprint pattern on a 50 μ thick sample at 40°; ○, data from fingerprint pattern on a 6.3 μ sample at 38.5° (taken from Table 2).

of capacitative and optical thresholds for field induced distortions of planar textured cholesterics has not as yet been achieved, nor has a study of the higher order bands in the reflection spectra been conducted.[8,9] Such experiments will be attempted in our future work.

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