³J. J. Thomson and G. Benford, to be published.

⁴S. Chandrasekhar, Rev. Mod. Phys. <u>15</u>, 1 (1943).

⁵F. L. Hinton and C. Oberman, Phys. Fluids <u>11</u>, 1982 (1968). Our Eq. (7) is their Eq. (23) rewritten in one dimension.

⁶J. Weinstock, Phys. Fluids <u>11</u>, 1977 (1968).

 7 C. T. Dum and T. H. Dupree, Phys. Fluids <u>13</u>, 2064 (1970). They actually assume a Gaussian probability distribution without mentioning its relation to the random-phase approximation.

⁸In previous work (Refs. 1 and 2), for mathematical simplicity the initial $\delta(v - v_0)$ in Eq. (1) is not allowed to spread. The approximation does not affect D(v), since it only depends on $\langle R'^2 \rangle$, but it does affect $\epsilon(k, \omega)$. Dupree obtains Eq. (14) without the $\langle v'^2 \rangle$ term, as a consequence. Thus his diffusive term is $-\frac{1}{3}k^3D\tau^3$. Substituting $\langle v'^2 \rangle = 2D\tau$ into Eq. (14), we obtain

 $\epsilon(k,\omega) = 1 - \omega_{p}^{2} \int \tau d\tau \exp(i\omega \tau - \frac{1}{2}R^{2}v_{t}^{2} - \frac{1}{3}k^{2}D\tau^{3} - k^{2}D\tau^{3}),$

so our diffusive term $-\frac{4}{3}k^2D\tau^3$ is 4 times as large as Dupree's.

Electric and Magnetic Field Investigations of the Periodic Gridlike Deformation of a Cholesteric Liquid Crystal

T. J. Scheffer

Institut für Angewandte Festkörperphysik der Fraunhofer-Gesellschaft, 78 Frieburg in Breisgau, West Germany (Received 6 December 1971)

> We have observed a two-dimensional, gridlike deformation of a cholesteric liquid crystal in a magnetic field, proving that this periodic structure does not require space charge or fluid flow. The frequency response of the threshold voltage in an electric field shows that there is a continuous transformation from the pure dielectric to the electrohydrodynamic regime. Investigations near Grandjean-Cano disclinations show the considerable effect of torsional strain on the threshold values.

Several kinds of electric- and magnetic-fieldinduced distortions are known to occur in cholesteric liquid crystals. A transformation from the cholesteric to the nematic structure was first observed by Wysocki, Adams, and Haas¹ in electric fields and by Sackmann, Meiboom, and Snyder² in magnetic fields. The helical unwinding predicted by de Gennes³ and Meyer⁴ for fields perpendicular to the cholesteric helical axis has been experimentally verified for both electric^{5,6} and magnetic^{7,8} fields. No distortion is expected when fields are applied parallel to the helix axis and the susceptibility anisotropy is negative. Either a 90° rotation of the helical axis or a conical deformation with pitch contraction, depending on the boundary forces, would be expected when the susceptibility anisotropy is positive.⁴ The susceptibility anisotropy is defined to be $\Delta \chi = \chi_{\parallel} - \chi_{\perp}$, where χ_{\parallel} and χ_{\perp} are the diamagnetic or dielectric susceptibilities parallel and perpendicular to the local optic axis, respectively. The 90° rotation, making the helical axis perpendicular to the field, happens before helical unwinding and the nematic transformation. This 90° rotation has been observed in electric fields^{6,9} and magnetic fields,¹⁰

and the conical deformation has been observed in electric fields.⁵ Helfrich^{11,12} has discussed the possibility of a periodic, one-dimensional deformation that can occur at even lower fields than the 90° rotation. He theorizes that such deformations could be produced by an electrohydrodynamic process or a purely dielectric process. In the former case, the conductivity anisotropy $\sigma_{\parallel} - \sigma_{\perp}$ must be positive, but the dielectric anisotropy $\epsilon_{\parallel} - \epsilon_{\perp}$ can have either sign. In the latter process the dielectric anisotropy must always be positive. The purely dielectric case should have a magnetic analog. A two-dimensional periodic deformation has been observed by Gerritsma and Van Zanten¹³ and by Rondelez and Arnould.¹⁴ In the latter study the dielectric anisotropy is negative, clearly indicating the electrohydrodynamic process. The dielectric process is probably functioning in the former case.

In this Letter we report the observation of a two-dimensional periodic pattern in a magnetic field (Fig. 1), proving that material flow or electrical conduction plays no role in the deformation. The threshold field data presented help verify Helfrich's predictions for the dielectric process.



FIG. 1. Dark-field observation of $50-\mu$ m periodic grid deformation produced by a 3850-Oe magnetic field perpendicular to the film. The upper edges of the two bands are single Grandjean-Cano disclinations spaced 0.95 mm apart. There are 10.5, 11.0, and 11.5 turns of the cholesteric helix in the lower, middle, and upper sections of the picture, respectively.

We use a three-component mixture. The major constituent is *p*-methoxybenzylidine, *p*-*n* butylaniline (MBBA). 4.89% by weight of nematic *p*-(*p*-butoxybenzylidene)-aminobenzonitrile (nematic point 62°C, cholesteric point 106°C) is added to give a positive dielectric anisotropy, and 1.23% cholesteryl benzoate gives the mixture a cholesteric twist. The pitch, or distance, for one complete turn of the cholesteric helix is $10.6 \pm 0.1 \ \mu$ m, and the isotropic transition temperature is 32.3° C.

The liquid crystal in its cholesteric phase is allowed to creep between two unidirectionally rubbed, SnO_2 -coated glass plates that form a wedge-shaped cell. The wedge angle is typically 20 min of arc. A tangle of threads is produced, but after several hours it resolves itself into a series of single, parallel, equally spaced Grandjean-Cano disclinations. These disclinations serve as useful reference points for computing sample thickness and torsional strain throughout the cell. The magnetic field is applied normal to the face of the cell, or alternatively, an electric potential is applied to the SnO_2 -coated plates.

The threshold voltage for the grid deformation is virtually independent of frequency from 2 to 300 kHz, the upper limit of our equipment. As the frequency is lowered below 2 kHz, the threshold voltage smoothly decreases and approaches a new value about half as large at 30 Hz. We interpret this frequency response as indicating a continuous transformation between the pure dielectric and the electrohydrodynamic regimes.

As expected, the observed characteristics of the grid pattern in a magnetic field appear identi-



FIG. 2. Thickness dependence of the electric and magnetic threshold values. Circles and triangles indicate measurements at single Grandjean-Cano disclinations, the triangles for the wide side and the circles for the narrow side. Open symbols are magnetic field data and shaded symbols are 2-kHz electric field data. Squares are electric field values at double Grandjean-Cano disclinations. The solid lines indicate the appropriate threshold field for the unstrained helix. All lines have a slope of 0.5.

cal to those seen in an electric field, if the frequency is high enough to put the process in the dielectric regime. Our magnetic field was limited to about 7 kOe so it was not possible to observe the higher field distortions such as the 90° tilt of the helical axis,^{6,9} helical unwinding,^{5,6} and the cholesteric-to-nematic transformation.9 All of these other distortions, however, were observed in a 2-kHz electric field. One new observation is worth mentioning here. Several hours after removing the voltage to a wedge-shaped sample that had been transformed to the nematic state, the original single Grandjean-Cano disclinations reformed into double disclinations with a discontinuity of one full helical turn. This doubling was observed in every sample studied, regardless of cell thickness or angle.

The considerable influence of torsional strain on the threshold values is seen in Fig. 2. The parallel lines indicate that a threshold disconuity exists across a Grandjean-Cano disclination, where the ratio of the threshold values on both sides of the disclination is a constant, independent of sample thickness. For single disclinations this ratio is 1.35 ± 0.05 and 1.37 ± 0.03 in a magnetic and a 2-kHz electric field, respectively, and for double disclinations the ratio is 2.00 ± 0.02 . The threshold value is always lower on the narrow side of the disclination where the helix is expanded from its equilibrium value.

We observe the frontier of the grid deformation to pass linearly with field across the region bounded by two Granjean-Cano disclinations. The threshold field for the unstrained cholesteric is therefore taken as the average of the two extreme values. This is indicated by the solid lines in Fig. 2. Anticipating the theoretical result, the best lines going through the data points in Fig. 2 were drawn with the restriction that they have a slope of 0.5. Within experimental scatter, our data are in essential agreement with this slope.

The width/length ratio of a cell in the grid pattern departs from unity in the vicinity of a Grandjean-Cano disclination. In one sample this ratio is 1.25 on the narrow side and 0.80 on the wide side of the disclination. The length dimension (measured parallel to the disclination line) is continuous across the disclination, but the width dimension shows a large discontinuity. We believe that the intrinsic distortions of a Granjean-Cano disclination, rather than simple torsional strain, are responsible for this effect, because the square pattern is strongly distorted only over a region comparable to the specimen thickness. A logarithmic plot of the grid length versus sample thickness L is shown in Fig. 3. The measured slope of 0.53 ± 0.02 is in good agreement with the theoretical value of 0.5 which is indicated in the figure.

The expression for the period λ of Helfrich's assumed one-dimensional striped deformation for both the dielectric and conduction processes is^{11, 12} $\lambda = (2k_{33}/k_{22})^{1/4}(PL)^{1/2}$, where k_{33} and k_{22} are the respective bend and twist elastic constants, and P is the pitch. We have verified the $L^{1/2}$ dependence for the grid deformation. From the measurements we compute $(2k_{33}/k_{22})^{1/4} = 1.45 \pm 0.04$. Direct measurement of the elastic constants of a comparable system gives a value of 1.47 ± 0.07 for this quantity.¹⁴

In the dielectric regime the threshold voltage U_0 for the one-dimensional pattern is¹²

$$U_{0} = 2\pi \left[\frac{2\pi (2k_{22}k_{33})^{1/2} (\epsilon_{\parallel} + \epsilon_{\perp})L}{\epsilon_{\perp}(\epsilon_{\parallel} - \epsilon_{\perp})P} \right]^{1/2}$$
(1)

Our measurements verify the $L^{1/2}$ dependence. We know approximately the dielectric anisotropy from the threshold voltage V_0 for the normal deformation of the nematic mixture before the cholesterol benzoate was added to the system. It can be computed from $V_0 = 2\pi [\pi k_{11}/(\epsilon_{\parallel} - \epsilon_{\perp})]^{1/2}$.¹⁵ All our measurements were taken at $23.5 \pm 0.5^{\circ}$ C,



FIG. 3. Thickness dependence of grid period measured parallel to the disclination, shaded trianges for the wide side and shaded circles for the narrow side. The data are for a 2-kHz electric field, but grid spacing is the same for magnetic fields. The open circle is from a double disclination. The line has a slope of 0.5.

corresponding to a reduced temperature T/T_c = 0.971. $k_{11} = 3.88 \times 10^{-7}$ dyn for pure MBBA at this reduced temperature.¹⁶ We measure $V_0 = 2.2$ ± 0.1 V and therefore $\epsilon_{\parallel} - \epsilon_{\perp} = 0.90 \pm 0.08$. ϵ_{\perp} for our mixture should be very close to 5.17 measured for pure MBBA at $T/T_c = 0.971.^{17}$ With the experimental threshold data for the unstrained helix, we compute from Eq. (1) $k_{22}k_{33} = (2.68 \pm 0.37)$ $\times 10^{-13}$ dyn². This elastic-constant product $k_{22}k_{33}$ can also be computed directly. For pure MBBA at $T/T_c = 0.971$, $k_{33} = 4.66 \times 10^{-7}$ dyn.¹⁶ This value and the ratio k_{33}/k_{22} obtained from the grid-period calculation gives $k_{22}k_{33} = (0.99 \pm 0.10) \times 10^{-13}$ dyn². If the directly computed value of $k_{22}k_{33}$ is substituted into Eq. (1), a threshold value is obtained that is a factor of 1.28 lower than that actually measured. This is fair agreement in view of the assumptions we have made.

In the case of magnetic fields, Eq. (1) simplifies to

$$H_0 = 2\pi \left[\left(2k_{22}k_{33} \right)^{1/2} / (\chi_{\parallel} - \chi_{\perp}) PL \right]^{1/2}, \qquad (2)$$

where H_0 is the magnetic field threshold for the striped pattern. The diamagnetic anisotropy $\chi_{\parallel} - \chi_{\perp} = 1.04 \times 10^{-7}$ (cgs units) for pure MBBA at $T/T_c = 0.971.^{18}$ The threshold-field data for the unstrained helix give $k_{22}k_{33} = (1.04 \pm 0.10) \times 10^{-13}$ dyn². The good agreement in this simpler case with the direct value of $k_{22}k_{33}$ helps support the validity of Eq. (2). This agreement also supports Helfrich's idea that the grid deformation may be a superposition of two one-dimensional striped patterns that are independent of each other for small deformations.

Our low-frequency ac measurements make it possible to partly verify the expression for the threshold voltage of the striped pattern in the electrohydrodynamic regime.¹² According to theory, the ratio of the threshold voltage in the conduction regime to that in the dielectric regime is

$$\frac{U_{0}(\text{hydr})}{U_{0}(\text{diel})} = \left[\frac{(\sigma_{\parallel} + \sigma_{\perp})(\epsilon_{\parallel} - \epsilon_{\perp})}{(\sigma_{\parallel} - \sigma_{\perp})(\epsilon_{\parallel} + \epsilon_{\perp})}\right]^{1/2}.$$

The conductivity anisotropy of our cholesteric mixture should be close to that of pure MBBA where, at $T/T_c = 0.971$, $\sigma_{\parallel}/\sigma_{\perp} = 1.48 \pm 0.01$.¹⁷ We therefore compute U_0 (hydr)/ U_0 (diel) = 0.64 ± 0.04 . This ratio is in qualitative agreement with 0.5, the ratio of the threshold voltages that we measured at 30 Hz and at high frequencies.

The considerable effect of torsional strain on the threshold field remains to be explained. A naive substitution of the nonequilibrium pitch present on either side of a single disclination into Eq. (1) gives a threshold ratio that is 1.069 and 1.011 for L/P = 3.75 and 23.75, respectively. This ratio is much too small when compared with the thickness-independent measured value of 1.37. The observed threshold ratio is comparable in magnitude to the ratio which could be expected from the threshold fields of torsionally strained nematics,¹⁹ except that in our case we observe that equal positive and negative torsional strains give different threshold values. It is expected that the threshold for the grid deformation is lower on the side of the disclination where the helix is expanded since, in the same geometry, conical deformations⁵ are known to occur which cause the pitch to decrease from its equilibrium value.

Productive discussions with Mr. H. Gruler and Dr. G. Meier are gratefully acknowledged.

¹J. Wysocki, J. Adams, and W. Haas, Phys. Rev. Lett. 20, 1024 (1968).

²E. Sackmann, S. Meiboom, and L. C. Snyder, J. Amer. Chem. Soc. 89, 5981 (1967).

³P. G. de Gennes, Solid State Commun. <u>6</u>, 163 (1968).

⁴R. B. Meyer, Appl. Phys. Lett. <u>12</u>, 281 (1968).

⁵H. Baessler, T. Laronge, and M. Labes, J. Chem. Phys. 51, 3213 (1969).

⁶F. J. Kahn, Phys. Rev. Lett. 24, 209 (1970).

⁷G. Durand, L. Leger, F. Rondelez, and M. Veyssie, Phys. Rev. Lett. 22, 227 (1969).

⁸R. B. Meyer, Appl. Phys. Lett. 14, 208 (1969).

⁹J. Wysocki, J. Adams, and W. Haas, Mol. Cryst. Liquid Cryst. 8, 471 (1969).

¹⁰I. Rault and P. Cladis, Mol. Cryst. Liquid Cryst. <u>15</u>, 1 (1971).

¹¹W. Helfrich, Appl. Phys. Lett. <u>17</u>, 531 (1970).

¹²W. Helfrich, J. Chem. Phys. <u>55</u>, 839 (1971).

¹³C. Gerritsma and P. Van Zanten, Phys. Lett. <u>37A</u>, 47 (1971).

¹⁴F. Rondelez and H. Arnould, C. R. Acad. Sci., Ser. B 273, 549 (1971).

¹⁵H. Gruler and G. Meier, to be published.

¹⁶H. Gruler, unpublished; I. Haller, unpublished.

¹⁷D. Diguet, F. Rondelez, and G. Durand, C. R. Acad. Sci., Ser. B <u>271</u>, 954 (1970).

¹⁸H. Gasparoux, B. Regaya, and J. Prost, C. R. Acad. Sci., Ser. B 272, 1168 (1971).

¹⁹M. Schadt and W. Helfrich, Appl. Phys. Lett. <u>18</u>, 127 (1971).

Spin-Orbit Effects in the Index-of-Refraction Formalism for Polarized Neutrons

Peter H. Handel*

Department of Physics, University of Missouri—St. Louis, St. Louis, Missouri 63121 (Received 20 December 1971)

Previously unexplained asymmetries observed in neutron guides are described by modifying the neutron index-of-refraction formalism through the inclusion of the spin-orbit interaction. The latter is due to the magnetic moment of the neutron in the electrostatic dipole field at the surface of the neutron mirror. For a single reflection, this effect may be of about 1% per volt of surface dipole, and it provides us with a direct method of measuring the surface dipole barrier.

Total reflection of neutrons of wave vector k is presently described by defining the index of refraction for neutrons in terms of the coherent forward scattering amplitude f and concentration N of the nuclei in the mirror,^{1,2}

$$n = [1 + 4\pi N f / k^2]^{1/2}.$$
 (1)

Generally f and n are complex numbers since we



FIG. 1. Dark-field observation of $50-\mu$ m periodic grid deformation produced by a 3850-Oe magnetic field perpendicular to the film. The upper edges of the two bands are single Grandjean-Cano disclinations spaced 0.95 mm apart. There are 10.5, 11.0, and 11.5 turns of the cholesteric helix in the lower, middle, and upper sections of the picture, respectively.