[1]V. E. Pogorelov, A. I. Lizengevich, I. I. Kondilenko, and G. P. Buyan, Usp. Fiz. Nauk 127, 683 (1979) [Sov. Phys. Usp. 22, 270 (1979)].

[2]D. J. Diestler, Top. Appl. Phys. 15, 169 (1976).

[3]A. Laubereau and W. Kaiser, Ann. Rev. Phys. Chem. 26, 83 (1975).

[4]J. M. Wiesenfeld and B. C. Moore, J. Chem. Phys. 70, 930 (1979).

[5]L. E. Brus and V. E. Bondebey, J. Chem. Phys. 63, 3124 (1975); 65, 71 (1976).

[6]G. Korzeniewski and M. Metia, J. Chem. Phys. 70, 5174 (1979).

[7]M. Shugard, J. C. Tully, and A. Nitzan, J. Chem. Phys. 69, 336 (1978).

[8]D. J. Diestler, J. Chem. Phys. 60, 2092 (1974).

[9]A. A. Ovchinnikov and N. S. Érikhman, Zh. Eksp. Teor. Fiz. 69, 266 (1975) [Sov. Phys. JETP 42, 137 (1975)].

[10]A. Nitzan, S. Mukamel, and J. Jortner, J. Chem. Phys. 60, 3929 (1974); 63, 200 (1975).

[11]V. P. Sakun, Fiz. Tverd. Tela (Leningrad) 18, 2517 (1976) [Sov. Phys. Solid State 18, 1470 (1976)].

[12]R. Kubo and Y. Toyozawa, Prog. Theor. Phys. 13, 160 (1953).

[13]Yu. E. Perlin, Usp. Fiz. Nauk 80, 553 (1963) [Sov. Phys. Usp. (1963)].

[14]P. K. Davis and I. Oppenheim, J. Chem. Phys. 57, 505 (1972).

[15]S. A. Adelman and J. D. Doll, J. Chem. Phys. 64, 2375 (1976).

[16]M. Shugard, J. C. Tully, and A. Nitzan, J. Chem. Phys. 66, 2534 (1977).

[17]S. Fisher, Chem. Phys. Lett. 11, 579 (1971).

[18]B. I. Makshantsev and I. P. Perstnev, Opt. Spektrosk. 30, 371 (1970); 31, 881 (1971).

[19]A. A. Ovchinnikov and V. G. Plotnikov, Chem. Phys. 21, 349 (1977).

[20]E. E. Nikitin and A. I. Osipov, Kinetika i Analiz (Kinetics and Analysis), Vol. 4, VINITI, 1977.

[21]R. T. Allen and P. Fener, J. Chem. Phys. 40, 2810 (1964).

[22]J. A. Beswick and J. Jortner, J. Chem. Phys. 68, 2278 (1978).

[23]K. C. Janda, J. M. Steed, S. E. Novick, and W. Klemperer, J. Chem. Phys. 67, 5162 (1977).

[24]S. L. Holmgren, M. Waldman, and W. Klemperer, J. Chem. Phys. 67, 4414 (1977).

[25]S. A. Adelman and B. J. Garrison, J. Chem. Phys. 65, 3751 (1976).

[26]H. D. Ladouceur and D. J. Diestler, J. Chem. Phys. 70, 2620 (1979).

[27]R. Rossetti and L. E. Brus, J. Chem. Phys. 71, 3963 (1979).

Translated by J. G. Adashko

Structure of chiral smectics in an electric field

V. E. Dmitrienko and V. A. Belyakov

All-Union Research Institute of Physicotechnical and Radiotechnical Measurements
(Submitted 25 September 1979)
Zh. Eksp. Teor. Fiz. 78, 1568–1578 (April 1980)

We consider the distortion and the untwisting of the spiral structure of chiral smectic liquid crystals (SLQ*) in a uniform electric field perpendicular to the spiral axis. Account is taken of the joint action of two molecule-orientation mechanisms in the field: dielectric, due to the presence of anisotropy of the dielectric constant ε_a, and ferroelectric, due to the presence of spontaneous polarization P in the SLQ*. The equations that determine the dependence of the pitch of the spiral on the field at arbitrary ε_a and P are determined. The dependence of the spiral-untwisting helical field on the SLQ* parameters is found. It is shown that domains with two different molecule orientations can exist in a field-untwisted SLQ*. The energy of the wall that separates such domains and the distribution of the molecule orientation in the wall are calculated.

PACS numbers: 61.30.Gd

It was recently observed that spontaneous polarization can exist in chiral smectic liquid crystals (SLC*), and it was shown that when an electric field is applied to such ferroelectric SLC* the spiral structure of the SLC* is distorted and can disappear completely.[1-4] The cause of this distortion and of the untwisting of the spiral structure is that the presence of the spontaneous polarization causes the dipole moments of the molecules to tend to become oriented along the field. In addition, the usual molecule-orientation in a field, the same as in nematic and cholesteric crystals, is present in the SLC*. This mechanism is quadratic in the field, and in fact determines the molecule orientation in sufficiently strong fields.

We consider below the change of the SLC* structure in an external electric field in the presence of both orientation mechanisms. A case of particular interest is that of positive anisotropy of the dielectric properties. In this case the cited molecule-orientation mechanism compete with each other and tend to rotate the molecules in opposite directions; this leads to certain signularities in the structure of the SLC* in the field, and in particular to the possibility of existence of domains with different molecule orientations in the SLC*.

1. FUNDAMENTAL EQUATIONS

We determine the change of the SLC* structure under the influence of a field in the manner used for cholesteric crystals[1-4] and for helicoidal magnets.[5] We consider an SLC* placed in a uniform electric field E perpendicular to the spiral axis. The expression for the free energy of the SLC* in an electric field is

$$F = \frac{B_3}{2}\left(\frac{d\varphi}{dz} - q_0\right)^2 + EP\cos\varphi + \frac{\varepsilon_a E^2}{16\pi}\cos 2\varphi, \tag{1}$$

where B_3 is the elastic modulus, φ is the azimuthal angle of molecule orientation $q_0 = 2\pi/p_0$ (p_0 is the pitch of the spiral in the absence of a field), P is the spontaneous polarization, $\varepsilon_\alpha = (\varepsilon_\parallel - \varepsilon_\perp)\sin^2\theta$ (θ is the angle of the molecules in the field relative to the z axis, $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the longitudinal and transverse components of the dielectric constant). In the derivation of (1) it was taken into account that the direction of P in an SLC* is determined uniquely by symmetry considerations[1-4] and is perpendicular to the z axis and to the local direction of the director. The field E is perpendicular to the spiral axis z and is parallel to y. In the derivation of (1) it was assumed, just as in Ref. 6, where the effect of a magnetic field on an SLC* was considered, that the angle φ varies only along the z axis, the smectic layers are not bent, and the angle θ and the distance between the layers remain unchanged (the conditions under which it is possible to neglect the influence of the changes of θ in the field on the $\varphi(z)$ dependence were recently investigated in detail in Ref. 6).

The dependence of the director orientation $\varphi(z)$ on the coordinate is obtained in the usual manner from the condition that the free energy of the SLC* be a minimum[1-5] and can be expressed in terms of elliptic functions (Refs. 8, 9).[1] It is more convenient, however, to use the inverse function $z(\varphi)$ which is determined by the following expression (see the Appendix):

$$z(\varphi) = \frac{p_0}{2\pi} \int_0^\varphi \left[C + R\left(\frac{E}{E_0}\cos\varphi' + 1\right)^2 \right]^{-\frac{1}{2}} d\varphi', \qquad (2)$$

where $E_0 = 4\pi P/\varepsilon_a$, $R = P^2 p_0^2/(\pi\varepsilon_a B_3)$. The physical meaning of the field E_0 is that when E is of the order of E_0 the spontaneous and induced polarizations turn out to be of the same order. The dimensionless parameter R determines the spiral-untwisting regime: the ferroelectric regime is realized at $|R| \gg 1$, the dielectric at $|R| \ll 1$, and the intermediate one at $|R| \sim 1$. The quantity C in (2) is determined from the condition that the free energy of the SLC* be a minimum. This leads to the following equation for C (see the Appendix):

$$\int_0^\pi \left\{ \left[C + R\left(\frac{E}{E_0}\cos\varphi + 1\right)^2 \right]^{\frac{1}{2}} - 1 \right\} d\varphi = 0. \qquad (3)$$

In the general case this equation can be solved only numerically.

Since the integrand of (3) is monotonic with respect to C, Eq. (3) has, depending on the external field E, either a single solution for C, which when substituted in (2) determines uniquely the structure of the distorted SLC*, or has no solutions at all at energies higher than a certain critical E_c. The latter means that at the corresponding fields $E \geq E_c$ the SLC* helix is completely untwisted.

At $E = 0$ we obtain from (3) $C + R = 1$. After substitution in (2) this yields $\varphi = q_0 z$, as should be the case in the absence of a field.

Expressions (2) and (3), which determine the structure of the distorted SLC*, make it possible to obtain various structural characteristics of the SLC*. In particular, the pitch p_E of the spiral in the field
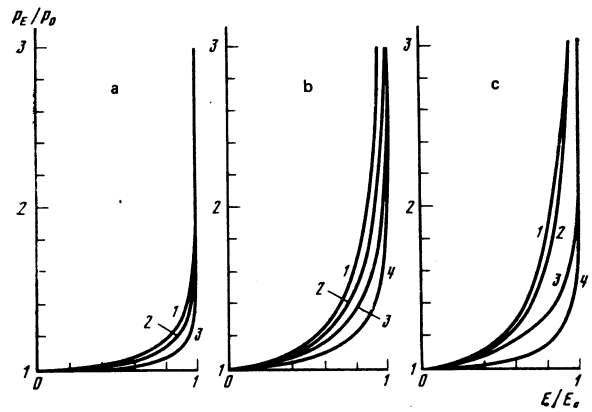

FIG. 1. Dependence of the pitch of the helix p_E on the field E for different values of the critical field E_c: a) $\varepsilon_a < 0$ (curves $1 - E_c = 0.2E_0$, $2 - E_c = E_0$, $3 - E_c = 10E_0$); b) $\varepsilon_a > 0$, $R \geq 1$ (curves $1 - E_c = E_0$, $2 - E_c = 0.9E_0$, $3 - E_c = 0.7E_0$, $4 - E_c = 0.1E_0$); c) $\varepsilon_a > 0$, $R \leq 1$ (curves $1 - E_c = 1.1E_0$, $2 - E_c = E_0$, $3 - E_c = 2E_0$, $4 - E_c = 5E_0$).

corresponds to a change of the azimuthal angle by 2π, and we obtain from (2)

$$p_E = \frac{p_0}{2\pi} \int_0^{2\pi} \left[C + R\left(\frac{E}{E_0}\cos\varphi + 1\right)^2 \right]^{-\frac{1}{2}} d\varphi. \qquad (4)$$

We note that the integrals in (2)–(4) can be expressed in terms of elliptic integrals (see the Appendix), but these expressions are quite cumbersome. It can be shown that in the limiting cases $|R| \gg 1$ and $|R| \ll 1$ Eqs. (2) and (4) go over into the known expressions for the ferroelectric[3] and dielectric[2,3] regimes of untwisting of the spiral, respectively.

The spiral pitch calculated from (4) with account taken of the condition (3) are shown as functions of the field in Fig. 1. It follows from this that in fields not too close to the critical field E_c the most noticeable change of the spiral pitch takes place in the intermediate case $R \sim 1$. The case $R = 1$ admits of a simple analytic solution, which will be considered in the next section.

2. ANALYTIC SOLUTION

We consider now the effect of an external SLC* at $R = 1$ (intermediate case). It is easy to verify by direct substitution that in this case Eq. (3) is satisfied (at $E < E$) if $C = 0$, and can thus be solved exactly.

For the dependence of the pitch on the field we obtain from (4)

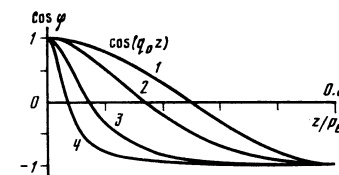$$p_E = p_0/[1 - (E/E_0)^2]^{\frac{1}{2}}. \qquad (5)$$

FIG. 2. Change of azimuthal angle of the director along the spiral axis in field-distorted SLC*, $R = 1$ (curves $1 - E = 0$, $2 - E = 0.5E_0$, $3 - E = 0.9E_0$, $4 - E = 0.98E_0$).

It is seen therefore that p_E increases quite rapidly with increasing E and the pitch $p_E \to \infty$ as $E \to E_0$, i.e., the spiral is untwisted (see Fig. 1b). Consequently in this case it is E_0 which is the critical field E_c, and as E tends to E_c the pitch diverges logarithmically.

We determine now the dependence of the director orientation (of the azimuthal angle φ) and z. From (2) we get

$$z(\varphi) = \frac{p_0}{2\pi} \int_0^\varphi \frac{d\varphi'}{1 + (E/E_0)\cos\varphi'}$$

$$= \frac{p_0}{\pi[1-(E/E_0)^2]^{1/2}} \operatorname{arctg}\left[\left(\frac{E_0-E}{E_0+E}\right)^{1/2} \operatorname{tg}\frac{\varphi}{2}\right], \qquad (6)$$

whence

$$\cos\varphi(z) = \frac{\cos(2\pi z/p_E) - E/E_0}{1 - (E/E_0)\cos(2\pi z/p_E)}. \qquad (7)$$

The function $\varphi(z)$ obtained from (7) is shown for different values of the field in Fig. 2. It is seen that as $E \to E_0$ the majority of the molecules are so oriented perpendicular to the field that $\varphi \approx \pi$.

3. THE CRITICAL FIELD IN THE GENERAL CASE

To determine the dependence of the spiral pitch in the field it is necessary, generally speaking, to solve Eq. (3) or (A.7). This is not necessary, however, for the determination of the critical field. It is clear that the untwisting of the spiral takes place at the field value $E = E_c$ at which the mean free energy of the SLC* in the field $F = \frac{1}{2}B_3(q_0^2 - C_1) - \varepsilon_a E^2/16\pi$ [see (A.8)] becomes comparable with the energy of the field-untwisted sample; this energy is given by

$$F_\infty = \frac{B_3}{2} q_0^2 + EP\cos\varphi_0 + \frac{\varepsilon_a E^2}{16\pi}\cos 2\varphi_0, \qquad (8)$$

where φ_0 is the molecule orientation angle in the untwisted sample and is independent of z. This angle is determined from the condition that (8) be a minimum:

$$-EP\sin\varphi_0 - \frac{\varepsilon_a E^2}{8\pi}\sin 2\varphi_0 = 0. \qquad (9)$$

If $\varepsilon_a < 0$, then the minimum corresponds to $\varphi_0 = \pi$, and the molecules are oriented perpendicular to the field. If $\varepsilon_a > 0$, then in fields $E \leq E_0 = 4\pi P/\varepsilon_a$ the molecules are oriented perpendicular to the field; in fields $E > E_0$ two orientations on the same footing are possible, $\varphi_{01} = \arccos(-E_0/E)$ and $\varphi_{02} = -\varphi_{01}$.

From the untwisting condition $F = (F_\infty)_{\min}$ we obtain for the critical field E_c

$$-\frac{C_1 B_3}{2} = \begin{cases} -E_c P + \varepsilon_a E_c^2/8\pi, & \text{if } \varepsilon_a < 0 \\ -E_c P + \varepsilon_a E_c^2/8\pi, & \text{if } \varepsilon_a > 0 \text{ and } E_c < E_0. \\ -2\pi P^2/\varepsilon_a, & \text{if } \varepsilon_a > 0 \text{ and } E_c > E_0 \end{cases} \qquad (10)$$

It is easy to verify directly from (4) that under the condition (10) the pitch $p_E \to \infty$. Substituting C_1 from (10) in (3) or in (A.7) we obtain after integration the following relations, which determine in parametric form the dependence of the critical field on the SLC* parameters; at $\varepsilon_a < 0$

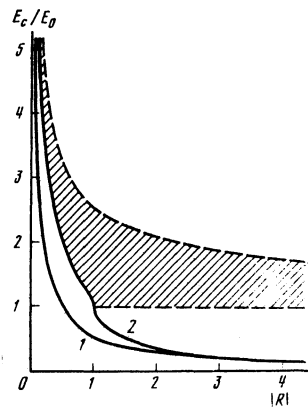$$E_c = E_0 \operatorname{sh}^2\psi, \quad R = -\pi^2/(2\psi + \operatorname{sh} 2\psi)^2; \qquad (11)$$

at $\varepsilon_a > 0$ and $E_c \leq E_0(R \geq 1)$

$$E_c = E_0 \sin^2\psi, \quad R = \pi^2/(2\psi + \sin 2\psi)^2, \qquad (12)$$

and at $\varepsilon_a > 0$ and $E_c \geq E_0(0 < R \leq 1)$

$$E_c = E_0/\sin\psi, \quad R = \pi^2/(2\psi + 2\operatorname{ctg}\psi)^2. \qquad (13)$$

It follows from (11)–(13) that in all three cases the dependence of E_c on the SLC* parameters is of the form $E_c = E_0 f(R)$. This dependence is shown in Fig. 3.

In cases (11) and (12) the molecules in the SLC* are oriented immediately after the untwisting perpendicular to the field, while in case (13) they make an angle φ_{01} and φ_{02} with the field. It is easy to ascertain from (11)–(13) the variation of E_c with the changes of the parameters of the SLC*. In particular, E_c increases with increasing B_3 and decreases with increasing p_0. At $\varepsilon_a < 0$ the field E_c decreases with increasing P and $|\varepsilon_a|$. At $\varepsilon_a > 0$ the dependence of E_c on P and ε_a is more interesting. Thus, at $E_c < E_0$ the field E_c decreases with increasing P, while at $E_c > E_0$, conversely, it increases; at $E_c < E_1(E_1$ is defined by the relations $E_1 = E_0/\sin\psi_1$ and $\psi_1 = \operatorname{cot}\psi_1)$ increases with increasing ε_a, and decreases at $E_c > E_1$.

It is also of interest to note that at $\varepsilon_a > 0$ the derivatives of E_c with respect to all the parameters (p_0, $\beta_{3,1}P$, and ε_a) become infinite at $E_c = E_0$, so that small changes of the parameters lead to considerable changes of E_c.

4. DOMAIN WALLS

As already noted, it follows from the expression for the free energy of a field-untwisted SLC* it follows that at $\varepsilon_a > 0$ and $E > E_0$ two orientations of the molecules in the field are possible, at angles $\varphi_{01} = \arccos(-E_0/E)$ and $\varphi_{02} = -\varphi_{01}$.

We consider now a situation in which the molecule orientation corresponds to φ_{01} in one part of an unbounded SLC*, and to φ_{02} in another, and determine the change of the molecule orientation in the intermediate region. This change can occur either jumpwise, in which case it corresponds to a defect in the SLC*, or continuously. We consider below the second case, i.e., a problem similar to that of domain walls in magnets.

V. E. Dmitrienko and V. A. Belyakov

In particular, we determine the energy of such a domain wall in an SLC* and the molecule-orientation distribution in it.

The expressions for the SLC* energy density and the Euler equation take in this situation the same forms (1) and (A.1) as above (under the assumption that φ depends only on z, i.e., that the wall is perpendicular to z). Now, however, we seek nonperiodic solutions, in which the molecule orientation angle changes from φ_{01} to φ_{02}, and accordingly, in expression (A.3) we have $\varphi \to \varphi_{\pm\infty}$ as $z \to \pm\infty$, where φ_{∞} and $\varphi_{-\infty}$ coincide either with φ_{01} or with φ_{02}.

The free energy of the wall, which is equal by definition to the energy difference between the SLC* with the considered molecule orientation and the single-domain SLC* (with all the molecules in the volume identically oriented), is given by the equation

$$\Delta F = \int_{-\infty}^{\infty} [F-(F_{\infty})_{min}]dz = \int_{\varphi_{-\infty}}^{\varphi_{\infty}} \frac{F-(F_{\infty})_{min}}{d\varphi/dz}d\varphi$$

$$= \int_{\varphi_{-\infty}}^{\varphi_{\infty}} \left[\frac{(B_3 C_1/2)+(\varepsilon_a E^2/4\pi)\cos^2\varphi+2EP\cos\varphi+2\pi P^2/\varepsilon_a}{\pm[C_1+(\varepsilon_a E^2/4\pi B_3)\cos^2\varphi+(2EP/B_3)\cos\varphi]^{1/2}} - q_0 B_3 \right] d\varphi. \quad (14)$$

Expression (14) describes four types of domain wall with different distribution of the molecule orientation in them. In fact, four possibilities are realized, in accord with the different values of the limits of φ_{∞} and $\varphi_{-\infty}$ and the two possible signs in (14) and in (A.3). We shall call a wall proper if the direction of rotation of the molecule axes with changing coordinate z coincides with the direction 0 molecule rotation in the undistorted sample, and improper in the opposite case. Walls in which the molecules are rotated through an angle less than π will be called N walls, and those with rotation through an angle larger than π W walls. The constants C_1 in the expressions (14) for the free energy of the walls are determined from the condition that the corresponding energy be a minimum with respect to C_1. From the form of the integrand in (14) it follows that ΔF is a monotonically increasing function of C_1. Therefore ΔF reaches a minimum at the lower limit of the permissible values of C_1, which is determined by the condition that ΔF be real, namely $C_1 = 4\pi P^2/\varepsilon_a B_3$. That the obtained C_1 is correct is confirmed by the fact that when φ tends to φ_{01} or φ_{02} we have $z \to \pm\infty$. Taking into account the obtained value of C_1, we get from (14) for the free energies of the various types of wall:

a) for an N wall

$$\Delta F_N^{\pm} = 2B_3 q_0 [\sqrt{R}(\varphi_{01}-\pi-\mathrm{tg}\,\varphi_{01})\pm\varphi_{01}\mp\pi]; \quad (15)$$

b) for a W wall

$$\Delta F_W^{\pm} = 2B_3 q_0 [\sqrt{R}(\varphi_{01}-\mathrm{tg}\,\varphi_{01})\mp\varphi_{01}]. \quad (16)$$

The upper and lower signs in (15) and (16) pertain to proper and improper walls, respectively.

It follows from (15) and (16) that the W-wall energy $\Delta F_W^{\pm}$ is always larger than zero and it is larger than the N-wall energy $(\Delta F_W^{\pm} \geq \Delta F_N^{\pm})$. The energy of the improper N wall ΔF_N^{-} is also larger than zero, and the

energy of the proper N wall is $\Delta F_N^{+} < \Delta F_N^{-}$ and its sign depends on the value of the field. In some field interval the improper N wall turns out to be energywise favored over the one-domain sample. The corresponding interval of the field E at which $\Delta F_N^{+} < 0$ is determined by a relation derived from (15):

$$max\{E_c, E_0\} < E < E_2, \quad (17)$$

where $E_2 = E_0/\cos\varphi_2$, and φ_2 obtained from the equation $(\sqrt{R}+1)\varphi_2 = \mathrm{tg}\,\varphi_2$; the range of values of E corresponding to the inequality (17) is shown shaded in Fig. 3.

It is physically quite understandable why the proper N wall is energywise favored in fields somewhat higher than critical (at $E_c > E_0$). In fact, at E somewhat smaller than E_c but larger than E_0 the twisted sample can be represented as a sequence of domains separated by alternating proper N and W walls. Untwisting takes place in a field E_e such that this sequence of domains becomes unprofitable, i.e., at $\Delta F_N^{+} + \Delta F_N^{+} = 0$. Since it is physically obvious that $\Delta F_N^{+} < \Delta F_W^{+}$, it follows hence that S at $E = E_c$ and in a certain region $E > E_0$ (shown shaded in Fig. 3).

We determine now the variation of the angle φ with the coordinate z in the wall. From (A.3) we obtain after integration:

a) for the N wall $(\varphi_{01} < \varphi < 2\pi - \varphi_{01})$

$$\mathrm{tg}\,\frac{\varphi}{2} = \mathrm{tg}\,\frac{\varphi_{01}}{2}\,\mathrm{cth}\left(\pm\frac{z\pi\sqrt{R}}{p_0\,\mathrm{ctg}\,\varphi_{01}}\right), \quad (18)$$

b) for the W wall $(-\varphi_{01} < \varphi < \varphi_{01})$

$$\mathrm{tg}\,\frac{\varphi}{2} = \mathrm{tg}\,\frac{\varphi_{01}}{2}\,\mathrm{th}\left(\mp\frac{z\pi\sqrt{R}}{p_0\,\mathrm{ctg}\,\varphi_{01}}\right), \quad (19)$$

where the upper and lower signs pertain to proper and improper walls.

It is interesting to note that the character of the variation of the angle φ in the wall, and particularly the characteristic wall dimension Δz, are described by an expression that does not contain the spiral pitch p_0 in the absence of a field:

$$\Delta z = \frac{p_0|\mathrm{ctg}\,\varphi_{01}|}{\pi\sqrt{R}} = \left(\frac{\varepsilon_a B_3}{\pi}\right)^{1/2}\frac{|\mathrm{ctg}\,\varphi_{01}|}{P}. \quad (20)$$

We see therefore that the wall dimensions depend on the field E. Two cases can be realized here. If $E_c < E_0$, then with increasing applied field E the following state become energywise most favored: a spiral structure in the interval $0 < E < E_0$, an untwisted single domain structure at $E_c < E < E_0$, a structure with proper N wall, in the interval $E_0 < E < E_2$, and an untwisted single-domain sample at $E > E_2$. At $E = E_0$ the wall dimension is infinite and decreases further with increasing E, reaching a value

$$(\Delta z)_{min} = \left(\frac{\varepsilon_a B_3}{\pi}\right)^{1/2}\frac{\mathrm{ctg}\,\varphi_2}{P}$$

at $E = E_2$.

In the case $E_c > E_0$ and with increasing E, a state with proper N wall becomes energywise more favored immediately past the spiral structure all the way to the field E_2. In this case the wall dimension in the interval

790   Sov. Phys. JETP 51(4), April 1980

V. E. Dmitrienko and V. A. Belyakov   790

$E_c < E_0 < E_2$ remains finite and decreases with increasing E. The maximum wall dimension at $E = E_c$ is

$$(\Delta z)_{max} = \left(\frac{\varepsilon_a B_3 E_0^2}{\pi P^2 (E_c^2 - E_0^2)} \right)^{1/2}$$

and the minimum wall is the same as in the preceding case. Thus, when the parameters of the SLC* and of the external field change, the wall dimension Δz can vary in a very wide range.

To conclude this section we note that in a slightly inhomogeneous (along the z axis) field the walls can move through the sample (along z). If the field is such that $\partial(\Delta F)/\partial E < 0$, then it is "profitable" for the wall to move into the strong-field region, and into the weaker-field region at $\partial(\Delta F)/\partial E > 0$. From (15) and (16) we obtain the possible signs of the derivatives:

$$\frac{\partial(\Delta F_N^\pm)}{\partial E} = \frac{\partial(\Delta F_W^\mp)}{\partial E} = 2B_3 q_0 [-R^{1/2} \operatorname{tg}^2 \varphi_{01} \pm 1] \frac{\partial \varphi_{01}}{\partial E}$$

$$\sim R^{1/2} \left[\left(\frac{E}{E_0} \right)^2 - 1 \right] \mp 1 \tag{21}$$

(we have taken in account here the fact that $\partial \varphi_{01}/\partial E < 0$). It follows from (21) that $\partial(\Delta F_N^-)/\partial E = \partial(\Delta F_W^+)/\partial E > 0$, whereas $\partial(\Delta F_N^+)/\partial E$ and $\partial(\Delta F_W^-)/\partial E$ can be positive as well as negative. This behavior of the domain walls makes it possible, in principle, to move them along the sample by varying the field and its inhomogeneity.

CONCLUSION

It follows from the foregoing analysis that the behavior of the SLC* in an electric field is on the whole similar to but much more diverse than the behavior of cholesteric crystals. The reason is that two molecule-orientation mechanisms, due to spontaneous and to induced dipoles, play a role in the SLC*. This behavior can be in fact even more complicated, since we have disregarded above the possibility of bending and inclination of the smectic layers relative to the field direction, the change of the angle of inclination of the molecules in the layer, and a number of other deformations. But even without this the obtained picture is quite complicated and cannot always be interpreted finally. Of particular interest in this connection is the presence of an analytically solvable case at a definite relation between the SLC* parameters.

It is useful to bear in mind that the various relations between the ferroelectric and dielectric mechanisms of the action of the external field on the SLC* can be obtained not only by selecting SLC* with different parameters, but also by turning on an additional magnetic or alternating electric field of sufficiently high frequency ω, which can lead to effective enhancement of the dielectric mechanism of the untwisting of the SLC* spiral. In this way it is also possible to vary the critical field E_c so as to obtain optimal experimental conditions.

Another peculiarity of the SLC* is the presence of two possible molecule orientations in the field-untwisted SLC*. We note that the state of the SLC* with a domain wall in an external field may be of interest because the width of the domain wall and its position can be controlled with the aid of the external field.

The expressions obtained in this paper for the variation of the SLC* structure in a field in the general case can be of use also for the description of the electro-optical properties of the SLC*, in particular the diffraction of light by a field-distorted structure (an analogous problem for the purely ferroelectric regimes was investigated in Ref. 10).

We note also that the foregoing analysis can be directly applied in some cases to a description of the behavior, in a magnetic field, of helicoidal magnets in which two competing helicoid-distortion mechanisms can appear.[5]

APPENDIX

We determine the distortion of a helical SLC* structure under the influence of the field. From the condition that (1) be a minimum we obtain the Euler equation

$$B_3 \frac{d^2\varphi}{dz^2} + EP \sin \varphi + \frac{\varepsilon_a E^2}{8\pi} \sin 2\varphi = 0, \tag{A.1}$$

which has a first integral

$$\left(\frac{d\varphi}{dz} \right)^2 - \frac{2EP}{B_3} \cos \varphi - \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi = C_1 = \text{const.} \tag{A.2}$$

From (A.2) we get

$$\frac{d\varphi}{dz} = \pm \left[C_1 + \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi + \frac{2EP}{B_3} \cos \varphi \right]^{1/2}.$$

Integrating (A.2), we obtain the following implicit expression for φ as a function of z:

$$z = \pm \int_0^\varphi \left[C_1 + \frac{2EP}{B_3} \cos \varphi' + \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi' \right]^{-1/2} d\varphi'. \tag{A.3}$$

The plus and minus signs correspond here to the same and opposite directions of rotation, respectively, of the molecule axes with changing z, as in the undistorted sample. Since it is obvious that the former case is energywise favored, we retain the plus sign everywhere, except in Sec. 4. Taking this into account we obtain from (A.3), after introducing a new constant $C = C_1/q_0^2 - R$, expression (2). The constants C or C_1 in (2), (A.2), and (A.3) are determined from the condition that the average free energy of the sample $\bar{F}$ be a minimum:

$$\bar{F} = \frac{1}{p_E} \int_0^{p_E} F \, dz = \frac{1}{p_E} \int_0^{2\pi} \frac{F \, d\varphi}{d\varphi/dz}. \tag{A.4}$$

Using expression (A.2) for $d\varphi/dz$, we get

$$\bar{F} = \frac{B_3}{p_E} \left\{ \int_0^{2\pi} \left[\left(C_1 + \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi + \frac{2EP}{B_3} \cos \varphi \right)^{1/2} - q_0 \right] d\varphi \right\}$$
$$+ \frac{B_3}{2} (q_0^2 - C_1) - \frac{\varepsilon_a E^2}{16\pi}. \tag{A.5}$$

Differentiating $\bar{F}$ with respect to C_1 we obtain the condition for the minimum of $\bar{F}$:

$$\frac{\partial \bar{F}}{\partial C_1} = -\frac{B_3}{p_E^2} \frac{\partial p_E}{\partial C_1} \left\{ \int_0^{2\pi} \left(C_1 + \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi + \frac{2EP}{B_3} \cos \varphi \right)^{1/2} d\varphi - 2\pi q_0 \right\} = 0. \tag{A.6}$$

It is seen from (4) that $\partial p_E/\partial C_1 \neq 0$ and consequently satisfaction of (A.6) calls for

$$\int_0^{2\pi} \left(C_1 + \frac{\varepsilon_a E^2}{4\pi B_3} \cos^2 \varphi + \frac{2EP}{B_3} \cos \varphi \right)^{1/2} d\varphi = 2\pi q_0. \tag{A.7}$$

V. E. Dmitrienko and V. A. Belyakov

After transforming the constant we obtain (3) from (A.7). From (A.5) and (A.7) we obtain for $\bar{F}$

$$\bar{F} = \frac{B_3}{2}(q_0{}^2 - C_1) - \frac{\varepsilon_a E^2}{16\pi}. \qquad (A.8)$$

The integrals in (2)–(4), (A.3), and (A.7) are expressed in terms of elliptic integrals (see Ref. 8). These expressions are different in the cases $\varepsilon_a < 0$ and $\varepsilon_a > 0$.

A. At $\varepsilon_a < 0$

$$\cos \varphi(z) = \frac{1 + (E_0/E)[1 + (-C/R)^{1/2}][2\,\mathrm{cn}^2(2zK(k)/p_E) - 1]}{(E_0/E)[1 + (-C/R)^{1/2}] - 1 + 2\,\mathrm{cn}^2(2zK(k)/p_E)},$$

$$p_E = 2p_0 K(k)/\{\pi[R + (C^{1/2} + E(-R)^{1/2}/E_0)^2]^{1/2}\}, \qquad (A.9)$$

where C is determined from an equation obtained from (A.7) or (3)

$$[E(k) - (1 + k^2/n)K(k) + (2 + n + k^2/n)\Pi(n, k)][R \\ + (C^{1/2} + E(-R)^{1/2}/E_0)^2]^{1/2} = \pi. \qquad (A.10)$$

$\mathrm{cn}(u)$ in (A.9) is the elliptic cosine, and $K(k)$, $E(k)$, and $\pi(n, kj)$ in (A.9) and (A.10) are complete elliptic integrals, with k and n defined by the expressions

$$k^2 = 4E_0 E(-CR)^{1/2}/\{E_0{}^2 R + [E(-R)^{1/2} + E_0 C^{1/2}]^2\},$$

$$n = 2E/[E_0 - (-C/R)^{1/2}E_0 - E]. \qquad (A.11)$$

B. At $\varepsilon_a > 0$ and $R > 1$

$$\cos \varphi(z) = \frac{(E_0/E)[1 + (-C/R)^{1/2}][2\,\mathrm{cn}^2(2zK(k)/p_E) - 1] - 1}{(E_0/E)[1 + (-C/R)^{1/2}] + 1 - 2\,\mathrm{cn}^2(2zK(k)/p_E)},$$

$$p_E = 2p_0 K(k)/\{\pi[R - ((-C)^{1/2} - ER^{1/2}/E_0)^2]^{1/2}\},$$

$$k^2 = 4E_0 E(-RC)^{1/2}/[E_0{}^2 R - (E_0(-C)^{1/2} - ER^{1/2})^2],$$

$$n = 2E/[E_0 + E_0(-C/R)^{1/2} - E], \qquad (A.12)$$

and C is determined from the equation

$$[E(k) - (1 + k^2/n)K(k) + (2 + n + k^2/n)\Pi(n, k)][R \\ - ((-C)^{1/2} - ER^{1/2}/E_0)^2]^{1/2} = \pi. \qquad (A.13)$$

C. At $\varepsilon_a > 0$ and $R < 1$

$$\mathrm{tg}\frac{\varphi(z)}{2} = \left(\frac{A}{B}\right)\frac{1 - \mathrm{cn}(4zK(k)/p_E)}{1 + \mathrm{cn}(4zK(k)/p_E)}, \qquad p_E = \frac{2K(k)p_0}{\pi(AB)^{1/2}},$$

$$k^2 = [(E/E_0)^2 R - (A - B)^2/4]/(AB), \qquad (A.14)$$

$$A = [C + R(1 + E/E_0)^2]^{1/2}, \quad B = [C + R(1 - E/E_0)^2]^{1/2}.$$

The constant C in (A.14) is determined from the equation

$$2(AB)^{1/2}\left[E(k) - K(k) + \frac{(A+B)^2}{4AB}\Pi\left(\frac{(A-B)^2}{4AB}, k\right)\right] = \pi. \qquad (A.15)$$

In the limiting cases $|R| \gg 1$ and $|R| \ll 1$ we obtain from (A.9)–(A.15) the known expressions for the ferroelectric[1-4] and dielectric[2,3] cases of spiral untwisting, respectively.

[1] The corresponding expressions are given in the Appendix.

[1] R. B. Meyer, Mol. Cryst. Liq. Cryst. 40, 33 (1977).

[2] P. de Gennes, The Physics of Liquid Crystals, Oxford, 1974.

[3] L. M. Blinov, Élektro- i magnitooptika zhidkikh kristallov (Electro- and Magneto-optics of Liquid Crystals), Nauka, 1978.

[4] S. A. Pikin and V. L. Indenbom, Usp. Fiz. Nauk 125, 251 (1978) [Sov. Phys. Usp. 21, 487 (1978)].

[5] I. A. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. 47, 992 (1964) [Sov. Phys. JETP 20, 665 (1964)].

[6] A. Rapini, J. Phys. (Paris) 33, 237 (1972).

[7] B. I. Ostrovskiĭ, S. A. Pikin, and V. G. Chigrinov, Zh. Eksp. Teor. Fiz. 77, 1615 (1979) [Sov. Phys. JETP 50, 811 (1979)].

[8] I. S. Gradshtein and I. M. Ryzhik, Tablitsy integralov, summ, ryadov i proizvedenii (Tables of Integrals, Sums Series, and Products), Fizmatgiz, 1962 [Academic, 1965].

[9] P. F. Byrd and M. D. Friedman, Handbook of Elliptic Integrals for Engineers and Physicists, Berlin, Springer, 1954.

[10] V. A. Belyakov and V. E. Dmitrienko, Light Scattering in Solids (Proc. of the Second USA–USSR Symposium on Light Scattering in Condensed Matter), New York, Plenum Press, 1979, p. 377.

Translated by J. G. Adashko