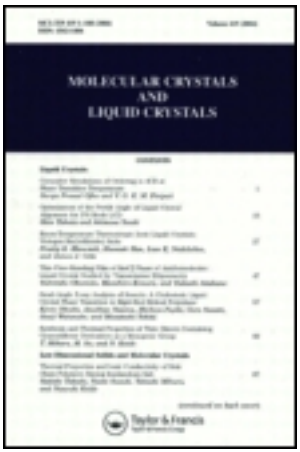

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Ph. Martinot-lagarde [a]
[a] Laboratoire de Phylique des Solides, Université de Paris-Sud, 91405, Orsay, France
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Unwinding of the Helical Texture of a Smectic C* Liquid Crystal, Through Ferroelectric and Dielectric Anisotropic Coupling With an Applied Field†

Ph. MARTINOT-LAGARDE

Laboratoire de Phy)ique des Solides, Université de Paris-Sud – 91405 Orsay, France.

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A large enough DC or low frequency AC field couples to the permanent polarization P and unwinds the helical texture of a smectic C* liquid crystal.[1] From the measurement of the unwinding critical field, one can deduce the value of P. We present here a general model to compute the unwinding critical field; this model takes into account both the effects of P, and of the dielectric anisotropy. This point will be useful to an understanding of the AC high frequency regime.[2]

THEORETICAL CALCULATION

We use here the method used by de Gennes to obtain the critical field necessary to unwind cholesterics and extended to the smectic C* by Meyer.[3]

The method is the following: the difference between the free energies of an infinite sample in two particular states (the unwound state, and the one turn state) is calculated from the value of the applied field E. Annuling this difference gives the value of E which allows unwinding of the last turn of the helix. This value of E is taken as the critical field E_c to unwind the whole helical structure.

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If a uniform electric field **E** in the plane of the layers is applied and ϕ is the angle of the permanent polarization $\mathbf{P} = \mathbf{P}_0\theta$ with respect to **E**, the ϕ dependent part of the free energy of a whole sample of infinite length along **z** is:

$$F = \int_{-\infty}^{+\infty} dz \left[\frac{1}{2} K_0 \theta^2 \left(\frac{d\phi}{dz} - q_0 \right)^2 + \left(\frac{\Delta\varepsilon}{8\pi} \right) E^2 \theta^2 \cos^2\phi - P_0 \theta E \cos\phi \right] \quad (1)$$

where $\Delta\varepsilon$ is the dielectric anisotropy and q_0 is the helical wave vector. We have neglected the effects of flexoelectricity, which have been considered by other authors.[4]

Minimizing F versus $\phi(z)$ gives the equilibrium equation:

$$\frac{d^2\theta}{dz^2} = -\frac{\Delta\varepsilon}{4\pi} \frac{E^2}{K_0} \cos\phi \sin\phi + \frac{P_0 E}{K_0 \theta} \sin\phi \quad (2)$$

Hence, by an integration:

$$\left(\frac{1}{2} \frac{d\phi}{dz} \right)^2 = 2 \sin^2 \frac{\phi}{2} \left(-\frac{\Delta\varepsilon}{4\pi} \frac{E^2}{K_0} \cos^2 \frac{\phi}{2} + P_0 \frac{E}{K_0 \theta} \right) + \tau \quad (3)$$

The integration constant τ corresponds to a uniform torsion due to the boundary conditions.

CASE OF NEGATIVE ANISOTROPY($\Delta\varepsilon < 0$)

The lower energy unwound state corresponds to $\tau = 0$ in (3). For this value of τ, Eqs. (2) and (3) have four solutions:

the two unwound states: $\dfrac{d\phi}{dz} = 0 \qquad \phi = 0 \qquad \phi = \pi$

the two one turn states: $z = \pm \displaystyle\int \frac{d\phi}{\sqrt{(d\phi/dz)^2}}$

The difference $F_1 - F_0$ between the free energies of the lower energy one turn and unwound states is finite for an infinite sample, because these two states correspond to the same value of τ:

$$F_1 - F_0 = \int_0^{2\pi} K_0 \theta^2 \left\{ \left(\frac{d\phi}{dz} \right)_1 - q_0 \right\} d\phi \quad (4)$$

Annuling, after integration, $F_1 - F_0$, gives the equation which allows us to obtain E_c.

$$\psi_- + \frac{1}{2}\sinh^2\psi_- = \frac{\pi}{2}\sqrt{K_0\theta^2 q_0^2 \times \frac{-\Delta\varepsilon}{4\pi P_0^2}} \quad \text{and} \quad E_c = \frac{4\pi}{-\Delta\varepsilon\theta} P_0 \sinh^2\psi_- \quad (5)$$

for small θ: $E_c = (\pi^4/4)(K_0\theta/P_0 Z^2)$; for large θ: $E_c = (\pi^2/Z)\sqrt{4\pi K_0/(-\Delta\varepsilon)}$.

CASE OF POSITIVE ANISOTROPY ($\Delta\varepsilon > 0$)

If the polarization induced by the critical field is lower than the permanent polarization, or more precisely, if $K_0\theta^2 q_0^2(\Delta\varepsilon)/(4\pi P_0^2) \leqslant 1$, the calculation is almost the same as in the previous case. It gives the same unwound state and almost the same one turn states.

The equation giving E_c is:

$$\psi_+ + \frac{1}{2}\sin 2\psi_+ = \frac{\pi}{2}\sqrt{K_0\theta^2 q_0^2 \frac{\Delta\varepsilon}{4\pi P_0^2}} \quad \text{and} \quad E_c = \frac{4\pi}{\Delta\varepsilon\theta} P_0 \sin^2\psi_+ \quad (6)$$

for small θ: $E_c = (\pi^4/4)(K_0\theta/P_0 Z^2)$.

If the permanent polarization is smaller than the induced polarization, we can call $\cos\phi_0$ the rate of these two polarizations (if $K_0\theta^2 q_0^2(\Delta\varepsilon)/(4\pi P_0^2) \geqslant 1$)

$$\cos\phi_0 = \frac{4\pi P_0\theta}{\Delta\varepsilon\theta^2 E_c}$$

The lower energy unwound states correspond to $\phi = \phi_0$ and

$$\tau_0 = \frac{\Delta\varepsilon E^2}{8\pi K_0}(1 - \cos\phi_0)^2$$

For this value of τ, there exist six solutions for the Eqs. (2) and (3): two unwound states: $\phi = \pm\phi_0$, two right and two left "part-of-a-turn" states:

$$z = E\sqrt{\frac{\Delta\varepsilon}{4\pi K_0}} \int \frac{d\phi}{\pm(\cos\phi - \cos\phi_0)}$$

with $-\phi_0 \leqslant \phi \leqslant \phi_0$, or $+\phi_0 \leqslant \phi \leqslant 2\pi - \phi_0$.

The one turn state is defined as the solution of Eq. (3), when $\tau \to \tau_0$ by higher values. It is equivalent to the two right "part-of-a-turn" states, one following the other, and its free energy F_1 is the sum of the energies of these two states. Annuling the difference $F_1 - F_0$ gives the equation which allows

us to calculate E_c. Here, when $E > E_c$ the sample can be either in one of the two unwound states or in the lower energy "part-of-a-turn" state, ($-\phi_0 < \phi < +\phi_0$), but all complete turns are unwound.

$$tg\phi_0 - \phi_0 = \frac{\pi}{2}\left(\sqrt{K_0\theta^2 q_0^2 \frac{\Delta\varepsilon}{4\pi P_0^2}} - 1\right) \quad \text{and} \quad E_c = \frac{4\pi P_0}{\Delta\varepsilon\theta\cos\phi_0} \tag{7}$$

CASE OF LOW INDUCED POLARIZATION

If the induced polarization is very small compared with the permanent polarization, the two Eqs. (5) and (6) can be solved approximately and give the single result:

$$E_c = \frac{\pi^2}{16}\frac{K_0\theta q_0^2}{P_0}\left[1 + \frac{\pi^2}{48}\frac{K_0\theta^2 q_0^2}{4\pi P_0^2}\frac{\Delta\varepsilon}{}\right] \tag{8}$$

which reduces to the threshold computed by Meyer for $\Delta\varepsilon = 0$.

CASE OF LOW PERMANENT POLARIZATION

If the permanent polarization (in the non ferroelectric limit) is very low compared with the induced polarization, Eqs. (5) and (7) also give the well known result for cholesterics

$$E_c = \frac{\pi^2}{Z}\sqrt{\frac{4\pi K_0}{|\Delta\varepsilon|}} \tag{9}$$

even if the two unwound states are different. If the dielectric anisotropy is positive, the long axis of the molecules is in the plane defined by the applied field and the normal to the layers ($\phi = \pm\pi/2$). If the anisotropy is negative, the long axis of the molecules is in the plane perpendicular to the applied field ($\phi = 0$ or π).

CONCLUSION

The originality of our calculation is the explicit dependence of E_c on P_0, K_0, θ and $\Delta\varepsilon$.

— *If the anisotropy* $\Delta\varepsilon$ *is negative*, the induced polarization helps to unwind the helix and gives the same unwound state as the permanent polarization ($\phi = 0$). Near the A $\rightarrow$ C* transition, the critical field is slightly smaller proportionally to θq_0^2, and increases continuously to reach the value for large θ which is the value obtained for cholesterics (proportional to q_0).

— *If the anisotropy is positive*, the induced polarization hinders the unwinding due to the permanent polarization. Near the transition, the critical field is slightly larger than proportional to θq_0^2. Under a given temperature T defined by $\theta^2(T)q_0^2(T)(\Delta\varepsilon)/(4\pi P_0^2) = 1$ the unwound state no longer corresponds to $\phi = 0$, but to $\phi = \phi_0$ with $\cos\phi_0 = 4\pi P_0\theta/\Delta\varepsilon\theta^2 E$. The orientation is a compromise between dielectric and ferroelectric alignment. So if $E > E_c$, the helix is unwound and the direction of the molecules changes with the amount of the applied field. However our assumption of constant θ may not always be valid.

From our knowledge of θ and q_0, the measurement of E_c and $\Delta\varepsilon$ should allow us to determine P_0.

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