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The influence of finite dimensions on the static ordering of the S_C^* phase in an electric field

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A ferroelectric liquid-crystalline sample of a finite length along the helical axis is studied in an external electric field applied perpendicular to the helical axis. By taking into account the linear coupling to the field, the equilibrium state is found for the case of free boundary conditions. Just below the critical field which induces the transition into the homogeneous S_C^* phase the domain-like structure appears as in the case of an infinite sample. The helical period in a finite sample is not a continuous function of the field, but it increases in finite jumps. We show that the finiteness of the sample has also an influence on the dielectric response at zero field.

1. Introduction

In the S_C^* phase the molecular director precesses as we proceed from one smectic layer to another so that the helical structure is formed with the period of about 10^3 smectic layers. An external electric or magnetic field applied perpendicular to the helical axis deforms the helical structure. If the field exceeds the critical value, the phase transition into the homogeneous S_C^* phase takes place. Both critical electric and critical magnetic field strongly depend on temperature [1–3]. By excluding a narrow temperature region below the S_A–S_C^* transition temperature, $T_c - T < 1$ K, we can describe the S_C^*–S_C^* phase transition induced by an external field within the constant amplitude approximation [4, 5] (CAA).

The helical unwinding caused by an external electric field has been studied already for a sample of an infinite length along the helical axis [6–8]. The static susceptibility has been shown [6] to diverge logarithmically at the critical field, which is in contradiction with the experimental results [9–12].

It has been pointed out [8] that two processes contribute to the dielectric response in an electric field: local reorientation of the in-plane polarization and the change of the helical pitch which is important especially near the critical electric field. The fluctuations of the helical pitch which contribute to the dielectric response at a bias electric field are of a long wavelength, so that the corresponding frequency should be very low. In a dielectric experiment where the applied frequencies are higher than a few 10's of Hz this process cannot contribute to the dielectric response. We expect that the relative contribution of this process to the dielectric response is more pronounced in samples of a finite dimension along the helical axis.

In the present paper the S_C^*–S_C^* phase transition in an electric field is considered for a sample of a finite length along the helical axis, taking into account the linear coupling to an electric field. We assume that the molecular director at both boundaries is free to rotate. In § 2 the mathematical procedure is described, that leads to the equilibrium

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solution in a static electric field. The results of the static solution are presented in § 3. In § 4 the influence of finite lengths on the dielectric response at zero bias field is discussed and in the Conclusions final remarks are given.

2. Formulation of the problem

Consider a sample of a finite length L along the helical axis which coincides with the z axis. We assume that the molecular director at both boundaries, defined by $z=0$ and by $z=L$, rotates freely on the smectic cone. In general a finite sample is polarized even in the absence of an external field in contrast to an infinite sample. The polarization appears whenever there is not exactly an integer number of helical periods in the sample. Let us denote this polarization by P_0. The orientation of the polarization P_0 in the smectic plane is arbitrary in the absence of the field. Any static electric field breaks this axial symmetry and turns the whole sample in such a way that the total polarization is parallel to the field.

The problem is studied on the basis of the Landau model [13] within the CAA. To describe the state of the system two order parameters are used, the tilt ξ which is a projection of the director $\mathbf{n}$ into the smectic plane xy, and in the in-plane polarization $\mathbf{P}$. In the CAA only the phase Φ of the tilt ξ is allowed to vary, so that one can express both order parameters in terms of their magnitudes Θ and P and the phase Φ, $\xi=\Theta(\cos\Phi, \sin\Phi)$ and $\mathbf{P}=P(-\sin\Phi, \cos\Phi)$. The part of the free energy which depends on the phase Φ can be written [8]

$$F=\int_0^L\left[\tfrac{1}{2}K_3\Theta^2\left(\frac{d\Phi}{dz}\right)^2-K_3\Theta^2 q_0\frac{d\Phi}{dz}-EP\cos\Phi\right]dz, \quad (1)$$

where K_3 is an elastic constant and q_0 is the wave vector related to the helical period p_0 at zero field, $q_0=2\pi/p_0$. The electric field points along the y axis, $\mathbf{E}=(0,E)$. The minimization of the free energy (1) leads to the sine-Gordon equation

$$K_3\Theta^2\frac{d^2\Phi}{dz^2}-EP\sin\Phi=0, \quad (2)$$

and to the boundary conditions

$$\frac{d\Phi}{dz}(z=0)=\frac{d\Phi}{dz}(z=L)=q_0, \quad (3)$$

which means that no external torque acts on the director at the boundaries. Let us denote by Φ_0 the value of the phase Φ at one boundary $\Phi(z=0)=\Phi_0$. We can then find the value of the phase at the other boundary, $\Phi(z=L)=n2\pi\pm\Phi_0$, where Φ_0 can in general take any value from the interval $[-\pi, +\pi]$. The integer number n and the phase Φ_0 are still to be determined. At a given field E and at a given length L there is a relation between the integer n and the phase Φ_0,

$$L=\int_{\Phi_0}^{n2\pi-\Phi_0}\frac{d\Phi}{d\Phi/dz}=\frac{8\kappa}{\pi q_0\sqrt{(E/E_c)}}[(n+1)K(\kappa)-F(\phi,\kappa)], \quad (4)$$

expressed by the complete $K(\kappa)$ and the incomplete $F(\phi,\kappa)$ elliptic integrals [14] of the first kind. The angle ϕ is equal to $\phi=(\Phi_0+\pi)/2$. The solution depends on the critical electric field $E_c=(\pi/4)^2\ (K_3\Theta^2 q_0^2/P)$ and $\kappa=2\sqrt{E}/\sqrt{(4/\pi)^2E_c+4E\cos^2\ (\Phi_0/2)]}$ is a dimensionless modulus, $\kappa\in[0,1]$. In deriving equation (4) we take into account the first integral of equation (2) and the boundary conditions (see equation (3)). At several n_i-s

we can solve numerically equation (4) for Φ_{0i} and obtain pairs (n_i, Φ_{0i}) which determine the state of the system. The equilibrium state is the one that minimizes the free energy which is, expressed explicitly in a dimensionless form, equal to

$$\frac{F}{E_c PL}=\frac{8\sqrt{(E/E_c)}}{\kappa}[(n+1)E(\kappa)-E(\phi,\kappa)]-\frac{8}{\pi}(n\pi-\Phi_0)-\frac{E}{E_c}\cos\Phi_0, \quad (5)$$

where $E(\kappa)$ and $E(\phi, \kappa)$ are the complete and the incomplete elliptic integrals [14] of the second kind, respectively. This minimization can only be done numerically. Once we find the integer number n which measures the number of helical periods in the sample and the initial phase Φ_0, the solution of equation (2) with boundary conditions (see equation (3)) can be expressed by the jacobian elliptic function cn

$$\sin\frac{\Phi}{2}=-\mathrm{cn}\left[\frac{\pi\sqrt{(E/E_c)}}{4\kappa}q_0(z+z_0),\kappa\right], \quad (6a)$$

where the constant z_0 is related to the initial phase Φ_0

$$z_0=\frac{4\kappa}{\pi q_0\sqrt{(E/E_c)}}F\left(\frac{\Phi_0+\pi}{2},\kappa\right). \quad (6b)$$

3. Results

A static solution $\Phi(z)$ of the sine-Gordon equation (2) is presented in figure 1 for different values of the reduced field E/E_c. The chosen length L of the sample is $L=4p_0$, where p_0 is the helical period at zero field. As seen from the figure the initial phase $\Phi_0=\Phi(z=0)$ changes with the field in such a way that the slope of the phase Φ is fixed and equal at both boundaries. It turns out that the free energy is minimal if the phase at $z=L$ is equal to $\Phi(z=L)=n2\pi-\Phi_0$. That means that the function $\Phi(z)-\Phi(L/2)$ is an odd function of the coordinate z with respect to the middle of the sample, $z=L/2$. In order for the total polarization to be parallel to the field, the initial phase Φ_0 must be

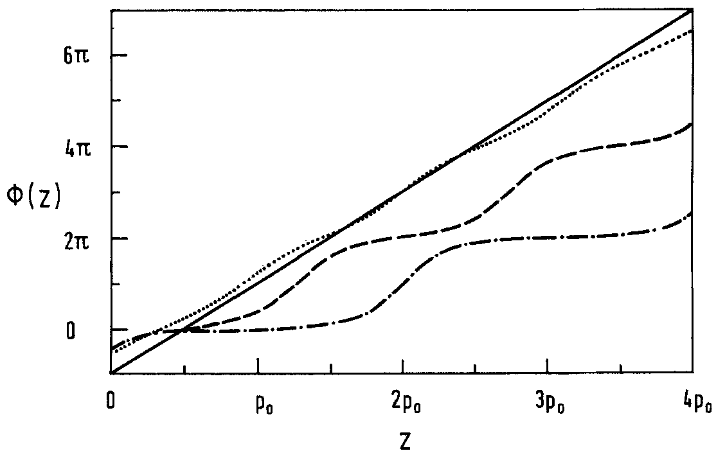


Figure 1. The static solution $\Phi(z)$ of the sine-Gordon equation with free boundary conditions on a finite interval of the length $L=4p_0$, where p_0 is the period of the helical structure at zero field. Different curves are related to different reduced fields E/E_c. At higher fields the solution lattice is found. ——, $E/E_c=0$; …., $E/E_c=0{\cdot}4$; – – –, $E/E_c=0{\cdot}85$; –·–·–, $E/E_c=0{\cdot}99$.

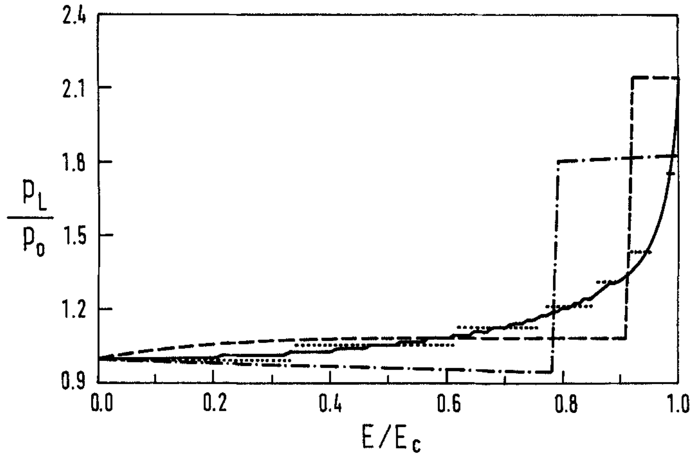


Figure 2. The period p_L of the helix in a finite sample as a function of the reduced electric field E/E_c. Different curves correspond to different values of the length L. —·—·—, $L=\sqrt{2}p_0$; – – –, $L=\sqrt{3}p_0$; ...., $L=15{\cdot}4p_0$; ——, $L=80{\cdot}7p_0$.

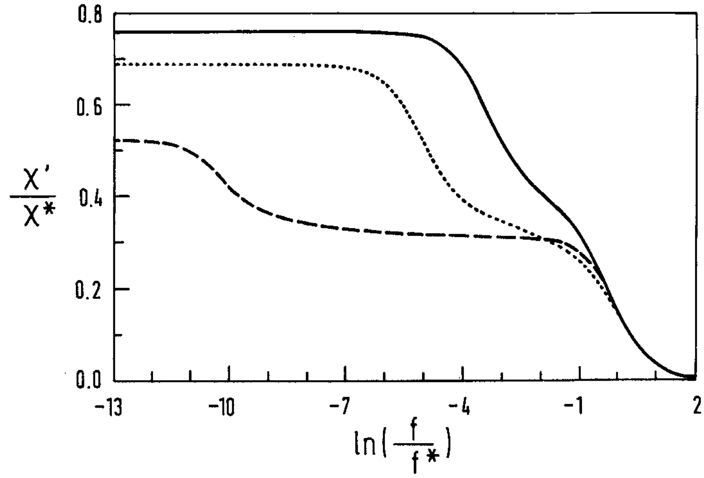
negative, $\Phi_0 \in [-\pi, 0]$. The deformation of the phase $\Phi(z)$ is small at lower fields. As the field approaches its critical value, the domain structure is formed. Within each domain the in-plane polarization is roughly parallel to the field.

It is worth mentioning that a pure homogeneous S_C^* phase, described by $\Phi=0$, strictly speaking, never takes place in a finite sample, since $\Phi=0$ is not a solution of equation (2) with the boundary conditions (see equation (3)).

The helical period p_L in a finite sample, defined by $\Phi(z+p_L)=2\pi+\Phi(z)$, is in thin samples considerably different from the pitch of an infinite sample. The pitch p_L is shown in figure 2 in dependence on the reduced field E/E_c. Different curves correspond to different values of the length L of the sample. One can notice jumps at certain field values, which diminish as we proceed to higher lengths. As shown in the figure the period p_L at small fields changes linearly with the field, either it increases or decreases, whereas in an infinite sample the helical period at small fields grows quadratically with the field.

4. Dielectric response at zero field

A dielectric response at zero field is, in a finite sample, more complicated than the response of an infinite sample. In an infinite sample the response does not depend on the orientation of the measuring field in the smectic plane. In a finite sample this is no longer the case due to the presence of the polarization P_0 in zero field. The response is different if the measuring field is parallel or if it is perpendicular to the polarization P_0. Here we consider only the case where the measuring field is parallel to this polarization. In this case the dielectric response is as shown in figure 3 where the real and the imaginary parts of the dielectric susceptibility are depicted in dependence on $\ln(f/f^*)$. A frequency f is the frequency of the measuring field. $f^* = \gamma^{-1} K_3 q_0^2$ is the phason frequency at $q=0$ and γ is a rotational viscosity. Different curves correspond to different sample lengths L. The response is significant at two frequencies, at $f_1 = \tau_L^{-1}$ and at $f_2 = \tau_F^{-1}$. The contribution at f_2 occurs due to the phason relaxation at $q=0$ and it is present also in the response of an infinite sample. The corresponding eigenmode has inverse relaxation time $\tau_F^{-1} \approx \gamma^{-1} K_3 q_0^2$. The contribution to the dielectric spectrum at



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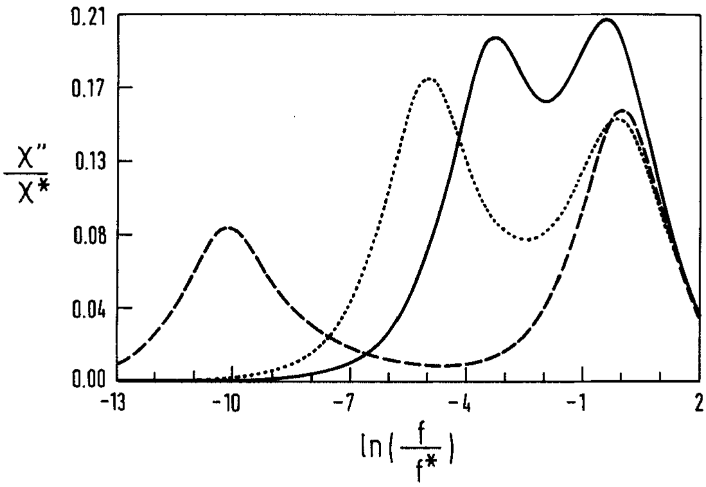

(*b*)

Figure 3. The analytical results for the real part χ'/χ^* and the imaginary part χ''/χ^* of the dielectric susceptibility, plotted in units $\chi^* = P/E_c$, in dependence on $\ln(f/f^*)$ where $f^* = \gamma^{-1} K_3 q_0^2$ and f is the frequency of the measuring field, evaluated at different lengths of the sample. The values of the length L for the curves are: ——, $L = 2{\cdot}8p_0$; ...., $L = 6{\cdot}2p_0$; – – –, $L = 80{\cdot}7p_0$.

the frequency f_1 appears due to the linear response of the helical pitch to small fields and it is present in finite samples only. In an infinite sample the helical period responds quadratically to small fields, so that this mode is absent from the dielectric spectrum.

Let us determine the relaxation time τ_L of the mode related to the variation of the helical period, which contributes to the dielectric response. The phason relaxation mode which causes the change of the helical period and for which the minimal energy is needed is the phason mode at $q=q_0 \pm q_m$, where $q_m = \pi/L$. The corresponding inverse relaxation time is equal to $\tau_L^{-1} = \gamma^{-1} K_3 q_m^2$ and it is thus inversely proportional to the square of the sample length L along the helical axis. This relaxation is referred to as a slow relaxation since the corresponding relaxation time is usually three or four orders of magnitude larger that the relaxation time τ_F of the fast mode. Although the relaxation time τ_L of this slow relaxation mode is rather low (less than about a second), it can be in principle detected in a dielectric experiment.

There is some experimental evidence for the existence of the slow relaxation process in ferroelectric liquid crystals. Studying the dynamic response of second harmonic generation in a ferroelectric liquid crystalline system, Ozaki and Yoshino [15] have detected two response times, one of the order of about a second and the other one of the order of about a millisecond. Recently also the theoretical work has been done by Hornreich and Shtrikman [16] on the dynamic response in the helicoidal cholesteric phase. Two relaxations have been predicted with well separated relaxation times, $\tau_F/\tau_L \approx 10^{-4}$. The long relaxation time τ_L is found to depend on the sample length quadratically, just as in our case.

5. Conclusions

A sample of a finite length along the helical axis has been considered in an external electric field applied perpendicular to the helical axis. The static solution has been found assuming free director rotation at both boundaries. Since a finite sample in general carries the polarization P_0 even in the absence of the field, the orientation of the helical structure in the field is nonarbitrary even if the field is very small. The total polarization of the sample is at any field parallel to the field and this is expressed by the initial phase Φ_0 which can in equilibrium only take values from the interval $[-\pi, 0]$. In the equilibrium state the function $\Phi(z) - \Phi(L/2)$ is an odd function with respect to the middle of the sample, $L/2$. The two parameters, the initial phase Φ_0 and the number of domains n, which at a certain field and at a certain sample length describe the state, are determined numerically by minimization of the free energy. The static solution is expressed by the jacobian elliptic function, similar as in the sample of an infinite length. The helical period p_L of a finite sample is found to be a noncontinuous function of the field. At small fields it depends linearly on the field in contrast to the helical period of an infinite sample.

The finite dimension of the sample along the helical axis influences the dielectric response at zero field. In addition to the response at the frequency $f_2 = \gamma^{-1} K_3 q_0^2$, being the only response in an infinite sample, the response at the frequency $f_1 = \gamma^{-1} K_3(\pi/L)^2$ appears in a finite sample, corresponding to the variation of the helical period. This frequency is rather low, since it is inversely proportional to the square of the length of the sample. It is about two or three orders of magnitude lower than the phason relaxation frequency at $q=0, f_2$. The contribution of the low frequency mode to the dielectric response might explain contradictions between the theoretically predicted behaviour [6] of the static dielectric susceptibility in a bias electric field and the experimental results [9–12].

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