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# Molecular Crystals and Liquid Crystals

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### Temperature and Field Hysteresis of the Pitch Variations in Thin Planar Layers of Cholesterics

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## TEMPERATURE AND FIELD HYSTERESIS OF THE PITCH VARIATIONS IN THIN PLANAR LAYERS OF CHOLESTERICS

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Temperature hysteresis of the cholesteric pitch in planar cholesteric layers was observed in [1,2]. A similar hysteresis of the cholesteric pitch in planar layers has to take place in the applied electric (magnetic) field. The restriction on the sample characteristics and strength of the surface anchoring potential are found which ensure defectless mechanism of the pitch field and temperature variations (jumps in the pitch value). The equations describing temperature and field pitch variations are presented and analysed. For weak field an universal character of the pitch variations, similar to those for the temperature variations and dependent on the value of some parameter only, is revealed. In the frame of Rapini model anchoring potential the field and temperature dependencies of the pitch variations and the hysteresis in the pitch jumps for specific values of the mentioned parameter are calculated. Transmission optical spectra of the layer for normal incidence of light are calculated at the points of pitch jumps.

Keywords: cholesterics; temperature and field pitch hysteresis

#### INTRODUCTION

Detailed studies of chiral liquid crystals become now a point of great interest. Especially promising are studies of the chiral LC in restricted geometries. The temperature variations of the cholesteric pitch in thin

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planar cholesteric liquid crystals (CLC) layers were investigated in [1,2] by means of the optical transmission spectroscopy. The optical transmission spectra were interpreted in the framework of the well-developed theory of the chiral LC optics [3,4] in the terms of temperature variations of the cholesteric parameters in the layer.

The observed unusual temperature behaviour of transmission spectra for the wavelength close to the cholesteric pitch value and temperature hysteresis of the jump-like changes of the pitch were attributed to the director deviations at the sample surface from the alignment direction in the potential well of surface anchoring and to the jump-like transitions of cholesteric helix in the layer between the helix configurations differing by one in the number of half turns at the layer thickness. Because the pitch jumps reveal themselves in many fine studies [5,6] the problem demands further investigation. The theoretical approach developed in [3,4] failed to describe the hysteresis mechanism relevant to the experimentally studied samples. In the paper [7] the restriction on the sample characteristics and strength of the surface anchoring potential are found which ensure defectless mechanism of the temperature pitch variations (jumps in the pitch value). It is found that critical value of the parameter  $S_d = K_{22}/(dW)$  exists (where K<sub>22</sub> is the twist constant, W is the height of the anchoring potential and d is the sample thickness) which ensures applicability to the problem of the pitch jump mechanism connected with overcoming by the director of the anchoring barrier at surface of the layer.

A similar behaviour of the director configuration of planar CLC and smectic layers in electric (or magnetic) field was predicted in recent paper [8]. Below we restrict the analysis of the temperature and field variations of the director configuration by a planar CLC layer assuming that the pitch jump mechanism is connected with overcoming by the director of the anchoring barrier at the surface.

#### PRINCIPLE EQUATIONS

We present below an analysis of the hysteresis phenomena for temperature variations of the CLC pitch in planar layers based on the continuum theory of elasticity taking into account of molecular anchoring forces at the surface. The conditions found in [1,2] for a jump in the CLC pitch, resulting in a restructuring of the director configuration in the layer from the CLC helix with N half-turns to the helix with N+1 half-turns explain qualitatively the observed hysteresis. However there was a lack of quantitative agreement between the theory and experiment.

We shall concentrate below the main attention to the transitions between N and N+1 half-turns in the layer which proceed without strong local disturbances of the director configuration.

We start by finding the temperature behavior of  $\varphi$ , the angle of deviation of the director orientation at the surface of the cholesteric layer from the alignment direction. Following [9] we write the free energy of the layer in the form

$$F(T) = 2W_s(\phi) + (K_{22}d/2)(2\pi/p_d(T) - 2\pi/p(T))^2, \tag{1}$$

where  $W_s(\phi)$  is the surface anchoring potential, p(T) is the equilibrium value of the pitch in a bulk CLC,  $p_d(T)$  is the corresponding value in the layer.

Because the pitch value in the layer  $p_d(T)$  is determined by the angle  $\varphi$  and the equilibrium pitch in a bulk CLC p(T) may be expressed via the angle  $\varphi_0(T)$ , which gives the value of deviation from the alignment direction at the surface in the absence of the anchoring forces the expression (1) for the free energy may be presented as a function of these angles as the following:

$$F(T) = 2W_s(\varphi) + (2K_{22}/d)(\varphi - \varphi_0(T))^2$$
 (2)

The angle  $\varphi$  may be found from the conditions of minimum of the free energy (2) what gives the following equation for  $\varphi$ :

$$\partial W_s(\varphi)/\partial \varphi + (2K_{22}/d)(\varphi - \varphi_0(T)) = 0$$
 (3)

Note, that after dividing (2,3) by W, one gets an equation containing parameter  $S_d$ . It means that the dependence of  $\varphi$  and the free energy F on the temperature at constant  $S_d$  is universal, i.e. independent on the number of half-turns of the helix in the layer.

A smooth changing of the director deviation angle  $\varphi$  is possible while  $\varphi$  is less than the critical angle  $\varphi_c$ . Upon achieving by  $\varphi(T)$  of  $\varphi_c$  a jumplike change of the pitch occurs and the transition to a new configuration of the helix differing by one in the number of half-turns occurs. The pitch just before the jump is expressed via  $\varphi_c$  (which is determined by the shape of the anchoring potential).

$$p_d(T_c) = 2d/(N + 2\varphi_c/\pi), p(T_c) = 2d/(N + 2\varphi_0(T_c)/\pi),$$
 (4)

where N is the number of half-turns at the layer thickness in the initial equilibrium configuration and  $T_c$  is the jump temperature. The angle  $\varphi_0(T_c)$  is given by the formula:

$$\varphi_0(T_c) = \varphi_c + (\partial W_c(\varphi)/\partial \varphi)_{\varphi =} \varphi_c/(2WS_d). \tag{5}$$

So if the shape of the anchoring potential and the temperature variations of the free pitch (in bulk cholesteric) are known the given above expressions determine the behavior of the pitch in the layer. The same expressions may be used for solution of the reciprocal problem, i.e. for reconstruction of the anchoring potential from the experimentally found temperature behavior of the pitch in the layer.

#### CRITICAL THICKNESS OF LAYER

Let us discuss the conditions of realisation of the pitch jump mechanism which is under the consideration. Because a jump with changing by one the number of half-turns in the layer may result in transition to nonequilibrium (metastable) configuration it is reasonable to introduce such critical parameter  $S_{dc}$  (or critical thickness of the layer) that if  $S_d > S_{dc}$  the transition proceeds to the stable configuration. The critical value of the parameter  $S_d$  and the critical layer thickness are determined by (3) if one puts  $\varphi = \varphi_c$  and  $2(\varphi_c - \varphi_0(T)) = -\pi$ . This substitution results in the relationship:

$$(\partial W_s(\varphi)/\partial \varphi)_{\varphi=}\varphi_c - \pi K_{22}/d = 0$$
 (6)

Here and below the critical parameters relevant to Rapini anchoring potential (see [10,11]) will be used:  $W_s(\varphi) = -(W/2)\cos^2\varphi$  for which  $\varphi_c = \pi/4$ . For the accepted assumptions the critical thickness  $d_c$  and critical parameter  $S_{dc}$  are determined by the formulas:

$$d_c = 2\pi K_{22}/W, \qquad S_{dc} = K_{22}/Wd_c = 1/2\pi$$
 (7)

Summarising the discussion of this section note that for the sample thickness smaller than  $d_c$  or  $S_d > S_{dc}$  the metastable configurations of the helix are not involved in the pitch jump transitions. In the following, we shall assume that the value of  $S_d$  is larger than  $1/2\pi$  in order to ensure that only one director configuration with the number of director half-turns differing from N by 1 may have a free energy below that of the initial configuration. This assumption allows us to not consider pitch jumps with  $\Delta N$  equal to  $\pm 2$  and more, which sometimes occur in the jump-wise changes of the director field [12].

#### PITCH VARIATIONS FOR RAPINI POTENTIAL

To make quantitative theoretical predictions it is necessary to know the explicit form of the anchoring potential. In what follows, the general expressions of the preceding sections are employed to describe the hysteresis for Rapini model of the anchoring potential.

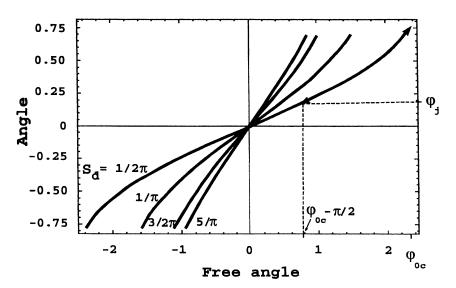
The results of calculations of the temperature dependence of the deviation angle of the director from the alignment direction at the surface are presented in Figure 1. Figure 2 presents the hysteresis loop for the deviation of the half-turns number in the layer from the integer number corresponding to the director alignment at the surface along the alignment direction.

#### FIELD HYSTERESIS OF PITCH IN PLANAR LAYERS

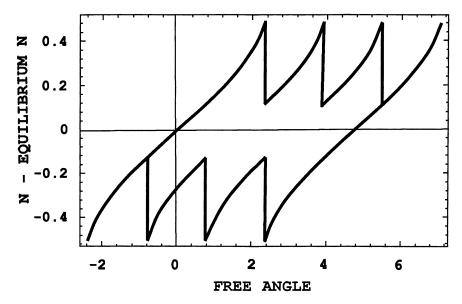
Another interesting problem is related to the pitch behavior in a layer subjected to action of external electric or magnetic field. This problem is more complicated than the considered above [7]. So below it will be considered the simplest situation of the CLC layer in an external field applied perpendicular to the helix axis.

In this case the density of the bulk free energy is of more complicated form due to the fact that it includes the external field and the cholesteric helix is distorted by the field. So the free energy of the layer in the field is given by the following expression.

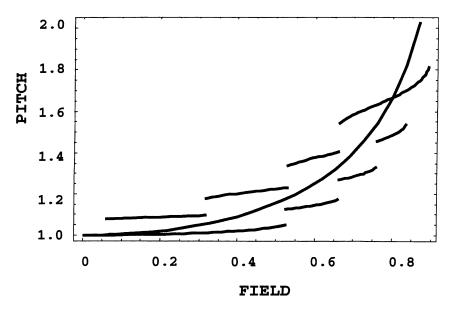
$$F(E) = F_s + \int F_v(E)dv, \tag{8}$$



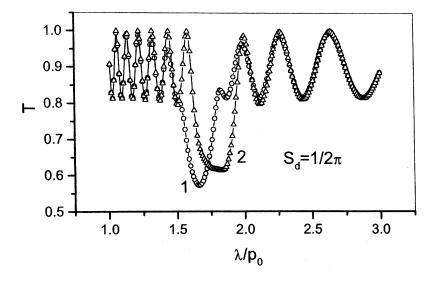
**FIGURE 1** Calculated deviation of the director (radians) at the surface versus the free deviation angle for several values of  $S_d$ . At one of the curve changes of the deviation angle at the jump are shown.

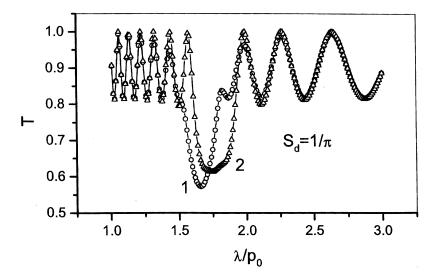


**FIGURE 2** Calculated deviation of the number of half-pitches from the integer number versus the free deviation angle (radians) for  $S_d=1/2\pi$ .

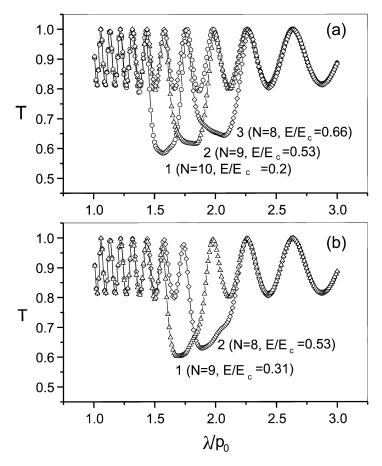


**FIGURE 3** Calculated changes of the pitch (divided by the value of pitch in zero field) versus the applied field (divided by the critical field) at increasing and decreasing of the field for  $S_d=1/2\pi$ .





**FIGURE 4** Calculated transmission spectra before (curves 1) and after (curves 2) the pitch jump from N=10 to N=9 at applied field (top graph  $E/E_c=0.527$ ) for the  $5\,\mu m$  cell thickness (bottom graph  $E/E_c=0.436$ ).



**FIGURE 5** Calculated transmission spectra at the pitch jumps for increasing applied field (a) (1, 2 correspond respectively to  $E/E_c=0.20$  and  $E/E_c=0.53$  for the jumps from N=10 to N=9 and 3 correspond to  $E/E_c=0.66$  for the jump from N=9 to N=8) and for decreasing applied field (b) (1, 2 correspond respectively to  $E/E_c=0.31$  and  $E/E_c=0.53$  for the jumps from N=8 to N=9 and the jumps from N=7 to N=8).

where  $F_v(E)$  is the density of the bulk free energy in the field  $E,\,F_s$  is the surface free energy.

$$F_{v}(E) = (K_{22}d/2)(d\varphi/dz - 2\pi/p_0)^2 + (\varepsilon_a E^2/16\pi)\cos 2\varphi,$$
 (9)

where in (9)  $\varphi$  describes local orientation of the director in the bulk of layer relative to the field direction,  $\varepsilon_a$  is the dielectric anisotropy and z is the coordinate along the cholesteric axis.

In the general case solution of this problem is complicated [7,13]. However in the case of strong anchoring and sufficiently large number of the pitch at the layer thickness the solution of the problem is similar to the case of varying temperature. Under the accepted assumptions the deviation of director at the layer surface in the field is described by (1) if p(T) is substituted by p(E), where p(E) is the value of the CLC pitch in field E in a bulk sample.

The results of calculations of the dependencies on the field of the pitch value in the layer are presented at Figure 3. Similar to the case of the temperature changes the variations of the pitch along with smooth changes experience jumps in varying field. The points of jumps are different for opposite directions of the field changes, i.e. there are hysteresis phenomena. One has to pay attention to the fact that the final value of the pitch in absence of the field is different from the initial values in absence of the field. All above calculations were performed for the value of parameter  $Sd = 1/2\pi$ . It was assumed also, that the number of the pitches at the sample thickness without field is 5 and the director directions at the layer surfaces coincide with the alignment direction. The pitch (divided by the value for zero field) is plotted versus the field (divided by the value for critical field of unwinding). The behavior of the pitch versus field in absence of the anchoring (smooth line) is taken from the paper [13].

The calculations of the optical transmission spectra at the points of jumps (Figures. 4 and 5) performed in the same approach as in [14] demonstrate how these jumps and hysteresis phenomena reveal themselves in the CLC optics. At the calculations the 5CB refractive indices were accepted  $(n_{0(\lambda\,=\,514)}=1.544,\,n_{e(\lambda\,=\,514)}=1.736,\,n_{0(\lambda\,=\,630)}=1.531,\,n_{e(\lambda\,=\,630)}=1.706)$  and normal dispersion low are taken into account.

### **CONCLUSION**

The presented investigation of the temperature and field variations of the CLC pitch in a layer showed that if the pitch jumps are described by the slipping mechanism (i.e.  $S_{\rm d} > S_{\rm dc}$ ) a sufficiently universal picture of the pitch changes takes place. The theory gives accessible to the experimental investigations predictions, for example, on the values of the pitch at the jump point. The approach allows obtaining qualitative information on the surface anchoring forces if the shape of the anchoring potential is known. However not less urgent is the reciprocal problem, i.e. the reconstruction of the real anchoring potential. It, in principle, can be solved with the help of the formulas obtained in the present work. What is concerned of hysteresis phenomenon in CLC layers and more general in layers of chiral LC it is quite promising for the applications.

And finally, mention that the temperature and field variations of the pitch in a layer are influenced by the director orientation fluctuations [7]. So the measured temperature and field dependencies can be used for studying of the fluctuations (see, for example [15,16]).

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