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TEMPERATURE PITCH VARIATIONS IN PLANAR CHOLESTERIC LAYERS: THE ROLE OF FLUCTUATIONS AND SURFACE ANCHORING

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The influence of thermodynamic fluctuations on temperature pitch variations in planar cholesteric samples with a finite surface anchoring energy is theoretically investigated in the framework of the continuum theory of liquid crystals. It is shown that taking fluctuations into account allows explaining experimental observations, namely, the absence of a temperature pitch jump hysteresis in sufficiently thick samples and its existence in thin ones. A description of fluctuations, including two phenomenological parameters, is proposed. It allows us to predict temperature points at which the pitch jumps in the sample between two configurations with the numbers of director half-turns differing by one, as a function of the anchoring energy, Frank elastic modulus, sample thickness, and temperture (or fluctuation energy). It is shown that performing precise measurements of the pitch versus the temperature in well-controled samples should allow determining the phenomenological constants and then predicting the influence of fluctuations on pitch jump parameters in samples of an arbitrary thickness and (or) surface anchoring energy. The corresponding calculations are performed using the Rapini–Popoular anchoring potential. It is shown that the influence of fluctuations on the pitch variation is only negligible in sufficiently thin layers. It is also noted that the results obtained could be useful for investigating pitch jump dynamics in the future.

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1. INTRODUCTION

Temperature unwinding of the helical structure of cholesteric liquid crystal layers of a finite thickness and surface anchoring energy has not yet been investigated sufficiently to clarify the physics of this phenomenom and to ensure optimal applications of the corresponding effects. Only cholesteric liquid crystal layers of a finite thickness down to monomolecular layers [7] have been investigated intensively and have revealed some interesting phenomena that are in particular related to the molecular anchoring at the layer surfaces. The effects observed in the cholesteric liquid crystal layers and their influence on the optical properties of the layer are of a great applied value because just the electrooptics of liquid crystal layers forms a basis of numerous efficient applications of liquid crystals in displays and information processing devices.

As has been known since long ago, the temperature evolution of the cholesteric liquid crystal structure [1, 2] in samples with a finite surface anchoring energy can be continuous at some ranges of the temperature with jumpwise changes at certain temperature points, with a strong hysteresis occurring when the temperature is changed in opposite directions [2, 3]. This problem was recently investigated in Ref. [4]. In this theoretical paper, a simple model for temperature variations of the

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pitch was developed in the framework of the Frank elasticity theory by taking the surface anchoring forces into account.

However, recent experimental investigations [2,3] show that the simple model in Ref. [4], where the pitch jump mechanism is related to the sliding of the director at layer surfaces through the anchoring potential barrier, is not directly applicable. The most probable cause of the discrepancy between the theory and experiments is that the theory neglects liquid crystal thermal fluctuations in the layer. It turns out that this assumption can be justified for sufficiently thin layers only. In what follows, we therefore present the same model with thermal fluctuations in the liquid crystal layer additionally taken into account. We show that taking thermal fluctuations into account allows explaining the tendencies observed in the experiments, predicting some new effects accessible experimentally, and determining the range of the parameters where the simple model is valid.

In general our primary aim in this paper is to give a qualitative an a semi-qualitative interpretation of the avalable experimental data and to propose a model for pitch variations.

2. ELASTIC MODEL WITHOUT FLUCTUATIONS

We first summarize the main results of the simple model without thermal fluctuations [4]. We examine the cholesteric liquid crystal helix unwinding as the temperature changes.

We consider a perfect planar layer of cholesteric liquid crystal and assume that the anchoring energies and the alignment directions are identical at both surfaces. The pitch variations due to temperature changes are determined by minimizing the free energy [1]

$$F(T) = 2W_s(\varphi) + \frac{K_{22}d}{2}\left(\frac{2\pi}{p_d(T)} - \frac{2\pi}{p(T)}\right)^2, \qquad (1)$$

where K_{22} is the twist Frank modulus, $W_s(\varphi)$ is the surface anchoring potential, d is the sample thickness, $p(T)$ is the equilibrium pitch at temperature T in a bulk cholesteric sample, $p_d(T)$ is the pitch measured at the same temperature in the layer, and φ is the deviation angle of the director with respect to the alignment direction at the surface. Because the pitch value $p_d(T)$ in the layer is determined by the angle φ and the equilibrium pitch $p(T)$ is determined by the angle $\varphi_0(T)$ that corresponds to a free deviation of the director from the alignment direction at the surface (in the absence of anchoring), free energy (1) can also be expressed as a function of these angles. As a result, the φ variations due to pitch (temperature) changes can be described by the equation [4]

$$\frac{\partial W_s(\varphi)}{\partial \varphi} + \frac{2K_{22}}{d}\left[\varphi - \varphi_0(T)\right] = 0. \qquad (2)$$

The pitch jumps occur when the angle φ reaches some critical value φ_c that depends on the shape of the anchoring potential $W_s(\varphi)$. The value of the free rotation angle $\varphi_0(T)$ at the jump point (or in other words, the corresponding value of the pitch in a bulk cholesteric liquid crystal) is related to the surface anchoring potential by

$$\varphi_0(T_j) = \varphi_c + \left(\frac{\partial W_s(\varphi)}{\partial \varphi}\right)_{\varphi=\varphi_c} \frac{1}{2WS_d}, \qquad (3)$$

where T_j is the jump temperature and $S_d = K_{22}/dW$ is a dimensionless parameter (with W being the depth of the surface potential).

Some results of this model, especially related to the hysteresis phenomena, are presented in [4]. In particular, formulas are given for the height of the anchoring barrier B between two director configurations in which the numbers N of director half-turns differ by 1 in the layer thickness.

In this paper, we give some additional results related to this simple model.

First of all, we examine the director deviation angle for the temperature points of a special physical interest. All our calculations are done using the Rapini–Popoular anchoring potential [1,4,5]

$$W_s(\varphi) = -(W/2)\cos^2\varphi,$$

for which the critical angle is $\varphi_c = \pi/4$.

Figure 1 shows the director deviation angle φ_e (from the rubbing direction) as a function of the parameter S_d at the temperature corresponding to equal free energies of the configurations with N and $N+1$ director half-turns in the layer thickness. The corresponding equation determining φ_e follows from (2) and is given by

$$\sin(2\varphi_e) + 4S_d[\varphi_e - \pi/4] = 0. \qquad (4)$$

Figure 2 presents the director deviation angle (from the rubbing direction) as a function of the layer thickness for the temperature corresponding to equal free energies of the configurations with N and $N+1$ director half-turns in the layer thickness. We note that in thick samples, the deviation angle φ_e approaches zero, while in thin ones, it becomes larger, reaching $\pi/4$ (which is the value of the critical angle φ_c) at zero thickness.

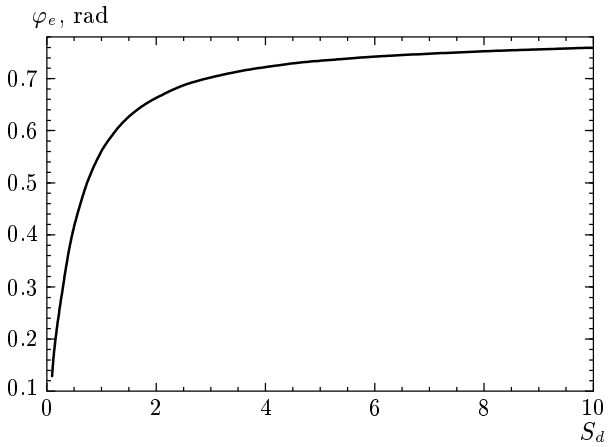

Fig. 1. The calculated director deviation angle (from the rubbing direction) φ_e as a function of the parameter S_d for the temperature corresponding to equal free energies of configurations with N and $N+1$ director half-turns in the layer thickness

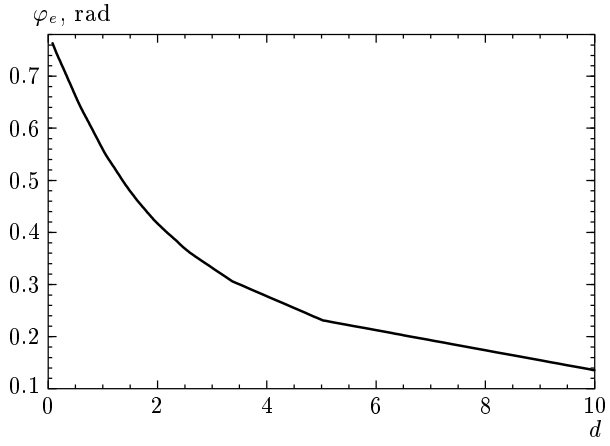

Fig. 2. The calculated director deviation angle φ_e versus the sample thickness normalized by the penetration length K_{22}/W (other conditions are the same as in Fig. 1)

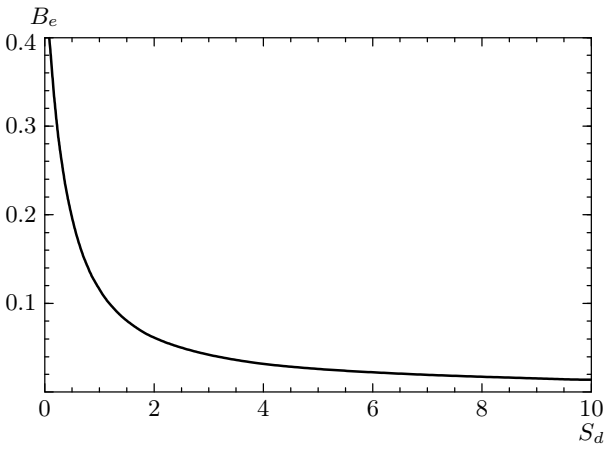

Fig. 3. The calculated height of the barrier B_e between director configurations in the layer differing by 1 in the number of the director half-turns N in the layer thickness as a function of the parameter S_d for the temperature corresponding to equal free energies for N and $N+1$ configurations

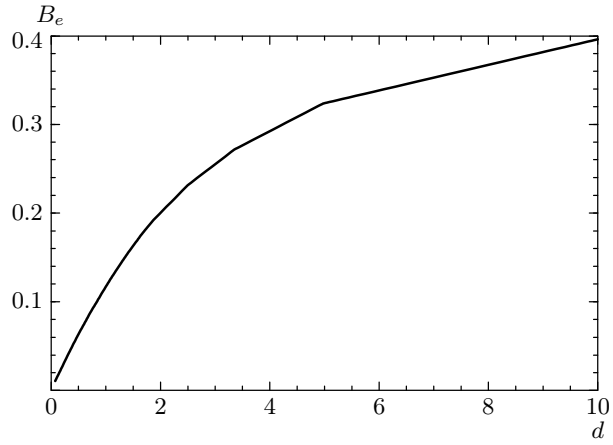

Fig. 4. The calculated height of the barrier B_e versus the sample thickness normalized by the penetration length K_{22}/W (other conditions are the same as in Fig. 1)

In Figs. 3 and 4, we show the results of the calculation of the barrier B_e between two director configurations with the numbers N of director half-turns differing by 1 in the layer thickness as a function of the parameter S_d (or the sample thickness) at the temperature corresponding to equal free energies for N and $N+1$ configurations. We note that the expression for B_e normalized by W can be found from Eq. (14) in [4], which reduces to

$$B_e = \cos^2(2\varphi_e) - \frac{\sin^2(2\varphi_e)}{8S_d} - \frac{1}{2}, \qquad (5)$$

where φ_e is the director deviation angle from the rubbing direction at the surface for the free director rotation angle $\varphi_0 = \pi/4$ (see Figs. 1 and 2 for the calculated values of φ_e).

It is useful to note that the limit of B_e at $S_d = 0$, or infinite thickness, is 0. The opposite limit at infinite S_d, or zero thickness, is $W/2$.

Figures 5 and 6 present the energy difference between N and $N+1$ configurations versus φ and the free rotation angle φ_0 (director deviation angle from

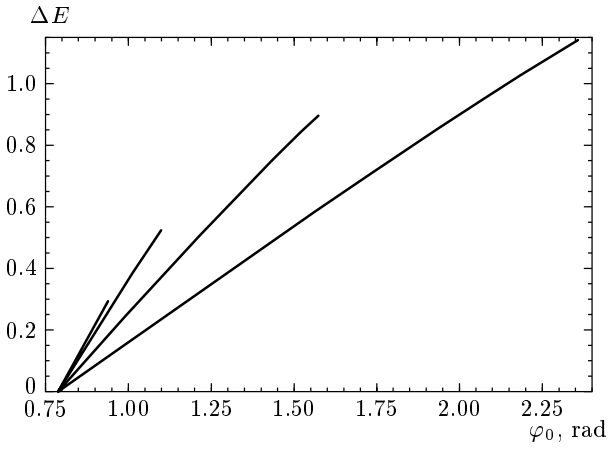

Fig. 5. The calculated difference of the free energy of configurations with N and $N+1$ director half-turns versus the free rotation angle φ_0; the calculations have been performed (from the bottom to top curves) for $S_d = 1/2\pi,\ 1/\pi,\ 5/2\pi,\ 5/\pi$

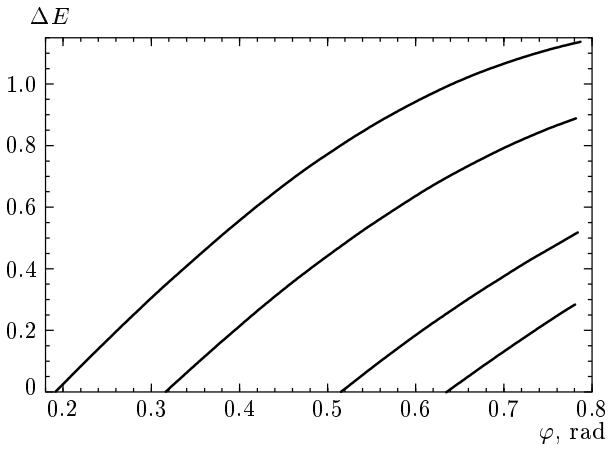

Fig. 6. The calculated difference of the free energy of configurations with N and $N+1$ director half-turns versus the director deviation angle from their alignment direction φ; the calculations have been performed (from the left to right hand side curves) for $S_d = 1/2\pi,\ 1/\pi,\ 5/2\pi,\ 5/\pi$

the alignment direction) beginning at the point where

$$F(N) = F(N+1).$$

Calculations have been performed using the formula deduced from Eq. (1),

$$\Delta E = \frac{F(N,\varphi_0) - F(N+1,\varphi_0 - \pi/2)}{W} = \\ = \frac{\sin^2[2\varphi(\varphi_0)]}{8S_d} - \cos^2[\varphi(\varphi_0)] - \\ - \frac{\sin^2[2\varphi(\varphi_0 - \pi/2)]}{8S_d} + \cos^2[\varphi(\varphi_0 - \pi/2)], \quad (6)$$

for $S_d = 1/2\pi,\ 1/\pi,\ 5/2\pi$, and $5/\pi$, where the argument of φ indicates that φ is a function of φ_0. In what follows, we 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 can have a free energy below that of the initial configuration. This assumption allows us to disregard pitch jumps with $\Delta N = \pm 2, \pm 3$, etc., which sometimes occur in jump-wise changes of the director field [10].

As mentioned above, this model must be improved by including the effects of liquid crystal thermal fluctuations in the bulk of the layer. The corresponding modification of the model is presented in the next section.

3. THE INFLUENCE OF FLUCTUATIONS ON PITCH CHANGES

The expressions given in the previous section relate thermodynamic equilibrium values of the parameters. But close to the points where the pitch jumps (transitions between N and $N+1$ configurations), bulk thermodynamic fluctuations can change the position of the transition points. For example, the hysteresis can decrease and even completely disappear because of fluctuations. In terms of the height of the surface anchoring potential between two configurations related to the transition, this implies that if in the simple model [4] the height of the barrier B must be equal to zero for the transition to occur, in a model taking fluctuations into account the transition can occur at $B \neq 0$, namely, for B differing from 0 by qk_BT, where T is the temperature, k_B is the Boltzmann constant, and q is some phenomenological coefficient to be determined experimentally. This means that the thermodynamic fluctuations of the energy of the liquid crystal allow the system to overcome the barrier, even if the equilibrium energy of the system is below the barrier.

It is now very essential to stress that the anchoring energy is proportional to the area of the layer surface and is independent of the thickness of the layer. In contrast, the bulk fluctuations of the energy, E_f, are proportional to the square root of the volume [6],

$$E_f = \sqrt{\langle \Delta E^2 \rangle} \sim qk_BT\sqrt{V} = qk_BT\sqrt{dS}, \quad (7)$$

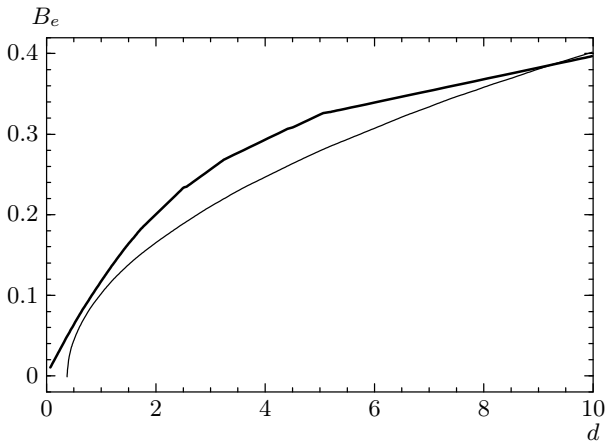

Fig. 7. The thickness d_f at which the hysteresis in the pitch jumps disappears is given at the intersection point between the two curves representing the barrier B_e and the fluctuation energy E_f as a function of the layer thickness normalized by the penetration length K_{22}/W (in the calculations, it was assumed that $d_0 = 0.4$ and $qk_BT/W = 0.13$)

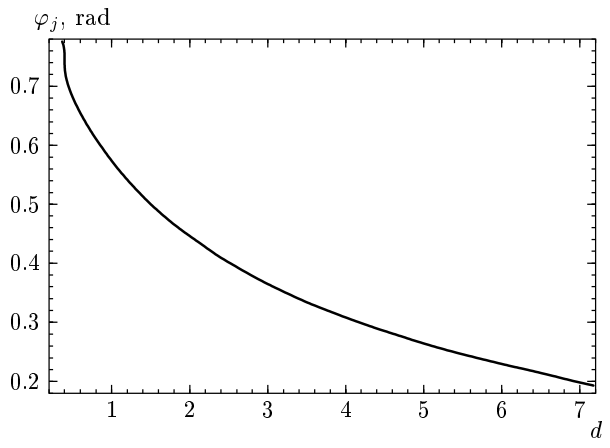

Fig. 8. The calculated jump angle φ_j for thicknesses $d < d_f$

where ΔE is the deviation of the energy from the equilibrium value due to fluctuations, V is the volume of the system, S and d are the surface area and the layer thickness, respectively. In what follows, we do not attempt to maintain numerical accuracy, but only indicate the form of the answers. Because the height of the anchoring barrier B is independent of d, the ratio $\sqrt{\langle \Delta E^2 \rangle}/W$ grows proportionally to $\sqrt{d}$, such that for some value of the layer thickness d, the fluctuation energy $\sqrt{\langle \Delta E^2 \rangle}$ becomes larger than the height of the surface anchoring potential barrier B.

Because fluctuations are reduced near the solid surfaces, we rewrite Eq. (7) as

$$E_f = \sqrt{\langle \Delta E^2 \rangle} \sim qk_BT\sqrt{(d - d_0)S}\,, \qquad (8)$$

where d_0 is some effective «surface thickness» in which fluctuations are suppressed. In principle, d_0 could be found from a microscopic theory, but we consider it as a new phenomenological parameter.

We note that the coefficient q in Eq. (8) is of the dimension $[L]^{-3/2}$. It can of course be made dimensionless by replacing qk_BT in Eq. (8) with $[L_p]^{-3/2}(qk_BT)$, where L_p is, for instance, the anchoring penetration length K_{22}/W.

To find the sample thickness for which the fluctuation energy becomes equal to the barrier B_e, we calculated the fluctuation energy E_f together with B_e as a function of the layer thickness (Fig. 7). Because the value of B_e given by Eq. (4) is the barrier height at the temperature point of equal free energies of two configurations differing by 1 in N, the intersection point of the curves for B_e and for $\sqrt{\langle \Delta E^2 \rangle}$ in Fig. 7 gives the thickness d_f for which the hysteresis disappears in the pitch jumps.

This is why the hysteresis in the pitch jump must disappear at layer thicknesses larger than d_f. The jump value of the director deviation angle φ_j then coincides with the director deviation angle φ_e corresponding to the temperature at which the configurations with N and $N + 1$ director half-turns in the layer thickness have equal free energies (see Fig. 2). This statement is confirmed by experimental observations resolving the hysteresis only for sufficiently thin samples [2, 3].

If the sample thickness is less than d_f, hysteresis occurs and the jump value of the deviation angle φ_j does not coincide with the deviation angle φ_e and exeeds it, while remaining smaller than the critical angle φ_c.

The physical reason why the fluctuation energy becomes larger than the surface anchoring barrier at some layer thickness is in the fact that the height of the surface anchoring potential is independent of the layer thickness, whereas the thermal fluctuation energy increases as the layer thickness increases.

The results of the calculations for the jump angle φ_j at thicknesses smaller than d_f and for the behavior of the jump angle φ_j at thicknesses both larger and smaller than d_f are presented in Figs. 8 and 9, respectively.

Figure 8 shows that the jump angle φ_j is essentially reduced by fluctuations, and the hysteresis is therefore also reduced. For the layer thicknesses $d > d_f$, the hysteresis completely disappears, and the jump angle φ_j is equal to φ_e. For $d < d_f$, the hysteresis reveals itself,

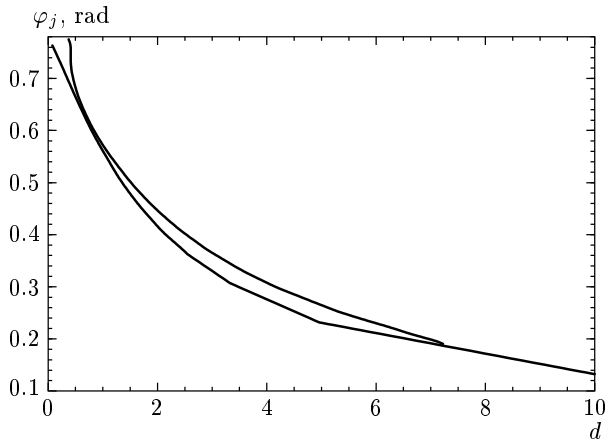

Fig. 9. The calculated jump angle φ_j at thicknesses both larger and smaller than d_f

but is less than in the model that does not take thermal fluctuations into account. This is why the jump angle φ_j is less than the critical angle φ_c that determines the jump angle in the model neglecting thermal fluctuations. It is only in sufficiently thin samples that the jump angle φ_j approaches the critical angle φ_c (which is $\pi/4$ for the Rapini anchoring potential), when fluctuations may be neglected.

4. TRANSITION IN A LIMITED AREA OF THE LAYER

It was tacitly assumed above that the $N \to N+1$ transitions driven by fluctuations occur over the entire surface area of the layer simultaneously. We now examine the role of the surface area S of the layer that is subjected to the transition. The difference between the anchoring barrier and the fluctuation energy can be estimated as

$$E_f - B \sim \sqrt{(d-d_0)S} - bS, \qquad (9)$$

where b is some coefficient. If we assume that the layer thickness d is fixed, the maximum of expression (8) is reached for the surface area

$$S^* = \frac{d-d_0}{4b^2}.$$

This is the surface area of the sample in which the transition is most favorable.

If the surface area of the sample is larger than S^*, we must analyze the situation where only a part of the layer experiences a fluctuation-induced $N \to N+1$ transition. The question then arises about the energy cost of the defect separating this region from the rest of the layer. The answer can be found using an estimate similar to (9) where we add the line energy of the defect line. The corresponding estimate allows us to determine the minimal surface area S_{min} of the region of the layer subjected to the $N \to N+1$ transition, which does not collapse and spontaneously increases in size after nucleation.

It is known [1] that in a wedge Cano–Grandjean structure, regions with N and $N+1$ half-pitches are separated by linear defects (χ disclination lines). We therefore also assume that the region induced by fluctuations with $N+1$ half-turns of the director is separated from the rest by a linear defect of the same type.

Consequently, we must now find the maximum of an expression of the type

$$E_f - B - E_d \sim \sqrt{(d-d_0)S} - bS - t\sqrt{S}, \qquad (10)$$

where the last term represents the energy E_d of the linear defect, proportional to its length and its energy per unit length (which we assume to be independent of the layer thickness, even if we know that it varies as $\ln(d/r_c)$, where r_c is the core radius). Expression (10) passes through a maximum at

$$S = \frac{\left(\sqrt{d-d_0} - t\right)^2}{4b^2},$$

where it is assumed that

$$\sqrt{d-d_0} > t$$

(otherwise, the fluctuation energy is insufficient for creating a linear defect). It follows from Eq. (10) that the maximal possible surface area for the fluctuation-induced $N \to N+1$ transition is given by

$$S_{max} = \frac{\left(\sqrt{d-d_0} - t\right)^2}{b^2}. \qquad (11)$$

To ensure a further growth of the area with the $N+1$ configuration after the fluctuation transition, the condition

$$SF(N+1) + K_{22}\sqrt{S} < SF(N) \qquad (12)$$

must be satisfied, assuming that the energy of the defect is of the order of K_{22} per unite length. This condition gives the minimal surface area of the fluctuation that can grow,

$$S_{min} = \left[\frac{K_{22}}{F(N) - F(N+1)}\right]^2, \qquad (13)$$

where the corresponding differences $F(N) - F(N+1)$ are shown in Figs. 5 and 6 as a function of φ_{0j} (φ_j)

for some values of S_d. We note that after the pitch jump to the $N+1$ configuration, the angle φ differs from the initial angle at the jump point φ_j in the N configuration.

Finally, the condition $S_{max} > S_{min}$ must be satisfied for a fluctuation transition to the $N+1$ configuration to occur in the layer.

We note that expression (12), which gives the energy gain during the $N \to N+1$ transition, may also be useful in describing the dynamics of defects (associated with the pitch jumps); this has not yet been studied in detail, contrary to the case with homeotropic anchoring [8].

5. ON THE DETERMINATION OF THE PHENOMENOLOGICAL CONSTANTS

The phenomenological constants q and d_0 introduced above could in principle be found from the microscopic theory of liquid crystals. But the complexity of liquid crystals and many uncertainties in their parameters do not allow us to expect a good accuracy of the corresponding calculations. A more practical way to determine them is therefore to compare the present theory with experimental measurements.

As regards d_0, it has a clear physical meaning and can be estimated quite well. It must be of the order of the penetration length K_{22}/W of the anchoring, i.e., of a micrometer order with $K_{22} = 10^{-6}$ dyn and $W = 10^{-2}$ erg/cm^2 [1]. The parameter q cannot be estimated so easily (it can be found in a nonanalytical form in the framework of the rather sophisticated approach of fluctuations in liquid crystal in restricted geometries [9]).

We now analyze what measurements could be used to extract the information about the phenomenological parameters under discussion. Keeping in mind that the coefficients in expressions (9) and (10) are related to the introduced phenomenological parameters as

$$b = \frac{B_e}{qk_BT}, \quad t = \frac{K_{22}}{qk_BT}, \tag{14}$$

we can find their values. Indeed, in accordance with Eqs. (8) or (10) and (13), this can be done by first measuring the sample thickness at which the hysteresis in jumps of the pitch disappears and then by measuring the minimal surface area of the region where the $N \to N+1$ transition occurs and does not collapse (i.e., grows) in later time. We thus obtain two relations that allow finding the two parameters d_0 and q.

The corrersponding measurements would consist of measuring the director deviation angle φ as a function of the temperature for different sample thicknesses d (or strength of the anchoring W). From these measurements, one can extract the jump angle φ_j and the value of d (or W) at which the temperature hysteresis for jumps disappears. Another measurement could be performed at the temperature of the pitch jump and would consist in measuring the minimal surface area of the region that is subjected to the $N \to N+1$ transition due to the fluctuation and which begins to grow after nucleation.

6. CONCLUSION

The results of the previous sections show that the dependence of the temperature-induced pitch jump hysteresis on the sample thickness can be explained by taking thermal fluctuations into account. In addition, our phenomenological theory should allow us to make quantitative predictions, provided the introduced phenomenological constants are determined from experiments. In theory, it should be sufficient to determine d_0 and q in a sample of a given thickness d in order to be able to predict the angle of the pitch jump and the hysteresis value for any other values of d or the anchoring strenth W. Unfortunately, the experiments performed up to now do not allow us to determine these phenomenological constants. A specially designed experiment for studying hysteresis phenomena in cholesteric layers would therefore be desirable. We can nevertheless give a rough estimate of q from the experiments described in Ref. [2]. In this work, hysteresis was observed for the layer thickness $d = 4.8$ μm and was not observed for $d = 18$ μm. If we assume that hysteresis disappears for $d = 10$ μm and the area of fluctuation S is of the order 10^4 μm^2, the dimensionless $q = 0.2$ (by taking $L_p = 1$ μm). This value looks quite reasonable, because according to [6], this quantity can be estimated as $\sqrt{C_{ve}/C_v}$, where C_v and C_{ve} are the total specific heat of the substance and the part of specific heat related to liquid crystal elasticity, respectively. It should also be mentioned that the method chosen in [2, 3] for measuring the pitch with the help of spectral optical measurements in the region of the reflection band, which are then fitted to the theoretical curves (see, e.g., [11]), gives a very precise measurement of the pitch. Other methods can be also used for the same purpose. We note that in the situations where Mauguin approximation of the cholesteric optics is valid, measurements of the rotation of the polarization plane of the light crossing the layer should give

similar information about the changes of the pitch in the layer.

We finally emphasize that this work could be used as a starting point for studying dynamics of pitch jumps in cholesteric layers.

Although the accuracy of our results is open to debate and the results are mostly based on assumptions, we believe that the proposed model for specific pitch changes demonstrates the possibility of the scenario considered in our paper, and a reasonable agreement with experimental data shows that weare on the right track.

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