Influence of phase sequence on focal conic domains in smectic films

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We present a study of smectic focal conic domains in films of various liquid-crystal compounds on a solid substrate. Focal conic domains are generated as a result of the antagonistic anchoring conditions of the film surfaces, random planar at the substrate interface and homeotropic at the air interface. The curved arrangement of the smectic layers in focal conic domains leads to a depression in the film/air interface above each domain. Using atomic force microscopy, we determine the temperature dependence of the depth of the depressions in different smectic phases and at different phase transitions. In most cases, our results are not in accordance with the assumption of strictly incompressible smectic layers, and we discuss the observed behavior with respect to the known results on the layer compressibility modulus *B*.

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I. INTRODUCTION

Structural features of liquid-crystal (LC) phases comprise a wide range of length scales, ranging from few nanometers (smectic density wave) to hundreds of micrometers (cholesteric helix) and can be influenced in many ways, e.g., by external fields, spatial confinement, and anchoring on substrates. In recent years, the concept has emerged to use LCs, e.g., by employing their characteristic defect structures, as matrices or templates to control the self-assembly of new microsystems and nanosystems. For instance, the distortion of the director field $\mathbf{n}(\mathbf{r})$ (\mathbf{n} designating the preferred direction of the long axis of the rodlike molecules) in a nematic phase by small water droplets or solid particles leads to special colloidal interactions [1] which may be used for the design of new self-assembling colloidal systems [2–4].

Smectic phases are characterized by an arrangement of the LC molecules in layers which exist in addition to the nematiclike parallel ordering of the molecules. The elastic constant controlling the compression and dilation of the smectic layers is many orders of magnitude larger than those controlling deformations of the nematic director field. Thus, in a smectic phase, only defect structures compatible with a nearly constant layer thickness are possible. This constraint results in the formation of so-called focal conic domains (FCDs) [5,6] in which the smectic layers are wrapped around two singular lines, in general an ellipse and a hyperbola, which are localized in two perpendicular planes and pass through each others focal point.

FCDs can be generated by applying an external influence which would induce a bent configuration of the director field in the nematic phase. In the smectic phase, the frustration resulting from the incompressibility of the smectic layers (which prevents a bend of the director field) leads to the formation of FCDs. There are several ways to create this frustration, e.g., the application of an electric field [7] or the use of cells with hybrid alignment conditions [8–10]. A simple way, similar to the use of a hybrid cell, is the preparation of a μ m thick smectic film on a substrate with random

planar anchoring conditions (i.e., n is oriented parallel to the interface but there is no preferred in-plane direction.) At the air interface of the film, usually a strong homeotropic anchoring exists (**n** is oriented perpendicularly to the interface). The two antagonistic anchoring conditions of the film interfaces can only be reconciled by bending the smectic layers to form FCDs, which possess in this case a toric shape, i.e., the two singular lines adopt the shape of a circle (localized on the substrate surface) and a straight line (running from the circle center to the air interface). In these films, FCDs often self-organize in a regular two-dimensional lattice [11,12] and several studies have explored methods to control their size and arrangement, e.g., by confining the FCDs into microchannels [13-16] or anchoring patterns [17,18] on the substrate. It was also shown that FCDs may serve as matrices for the self-assembly of soft microsystems [15].

The presence of FCDs not only provides smectic films with a superstructure made up by a regular arrangement of defect lines and curved layers, but it also leads to a deformation or modulation of the film/air interface: the curved arrangement of the smectic layers results in a depression in the film/air interface above each FCD. If the smectic layers were completely incompressible, the depth of the FCD-induced surface depression should depend only on the diameter of the FDC and the thickness of the smectic film. On the other hand, atomic force microscopy (AFM) measurements have shown that the depth of the surface depression may exhibit a pronounced temperature dependence in the smectic-*A* phase [19]. Experimental studies of this behavior could help to clarify the detailed structure of FCDs and also contribute to the design of new functional soft matter systems.

In this paper, we present a comparative study of the FCDinduced surface depressions for several LC compounds showing a variety of phase sequences. We find that the depth of the surface depressions varies in a broad range from values close to those expected on the basis of completely incompressible layers to values which are considerably smaller. We discuss our results with respect to the known results on the layer compressibility modulus B. There are several qualitative observations which indicate that a small value of B leads to a decrease in the depth of the FCDinduced depressions (because a small value of B allows for a dilation of the smectic layers). Quantitatively, however, our

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Acronym and Structure		Transition Temperatures (°C)
	n = 8	Sm-A 33 N 41 iso
n CB	C_nH_{2n+1} \sim	Sm- A 51 iso
		Sm- A 59 iso
$\bar{8}.S.5$	C ₈ H ₁₇ O-COS-C ₅ H ₁₁	Sm-C 55 Sm-A 63 N 86 iso
A7	$C_7H_{15}O$ $OCO - CH(CI) - CH(CH_3)_2$	Sm-C 73 Sm-A 82 iso
C7	$C_7H_{15}O-OCO - CH(CI) - CH(CH_3) - C_2H_5$	Sm-C 55 Sm-A 62 iso

TABLE I. Liquid-crystal compounds under investigation.

results can be explained in a simple elastic model only if we assume that the static value of B (which is relevant for our experiment) is one or 2 orders magnitude smaller than the B values obtained from mechanical measurements carried out in the kHz range. This indicates that there a probably additional factors influencing the depth of the FCD-induced surface depressions.

II. EXPERIMENT

Table I shows the LC compounds of the present study and their phase sequences and transition temperatures. Compounds of the *n*CB series were obtained from Synthon Chemicals, Germany, all other were synthesized in the group of G. Heppke during the course of previous studies [20]. Besides the pure compounds, we studied binary mixtures of 8CB and 10CB. The mixtures were prepared by weighing appropriate amounts of both compounds in a small glass tube and subsequent stirring for 30 min in the temperature range of the isotropic phase.

In order to generate FCDs in our LC films we need substrates imposing a random planar anchoring on the LC molecules. As substrates we used silicon wafers which were first cleaned with piranha solution and then coated with the alkoxysilane compound *N*-methyl-3-aminopropyl-trimethoxysilane (MAP) [21]. MAP was obtained from ABCR company, Germany. A diluted aqueous solution $(x_{\text{MAP}} \approx 3 \times 10^{-3} \text{ mol/l})$ was prepared and the silicon wafer was immersed for 5 min in the silane solution, washed with pure water, dried, and then cured for 1 h at 110 °C resulting in a chemisorbed silane layer permanently bonded to the substrate [22].

LC films on the substrates were produced by placing at room temperature a tiny amount of LC on the substrate. The temperature was raised to the range of the nematic or isotropic phase where the LCs formed flat droplets which reached a quasistatic shape after a few minutes. The samples were then slowly (0.2 °C/min) cooled across the nematic to smectic-A or isotropic to smectic-A transition where the FCDs formed. Thus, our LC films are in fact flat droplets the thickness of which increases from the edge to the center; near the edge, the thickness increases typically by a value between 50 and 100 nm per each μ m distance from the edge; this slope decreases toward the central region of the droplet.

The size of the FCDs in the LC films depends on the film thickness [11,23], and we intended to compare the different LC compounds in samples with similar thickness values. Thus, all LC films or droplets were prepared such that their thickness in the central region was in the range from 40 to 45 μ m. The thickness was determined by confocal microscopy measurements using a Leica TCS SP2 confocal microscope system. The film/air and the substrate/film interfaces were discernible by means of their reflections and the film thickness was obtained from the difference of the corresponding z coordinates. A structural study of the FCDs by fluorescent confocal polarizing microscopy measurements [24] was beyond the scope of the present study. Thus, it was not necessary to dope the samples with a fluorescent dye. All AFM measurements described in the following were carried out in that region of the sample for which the local thickness has been previously determined by confocal microscopy.

The AFM studies were conducted using a Nanoscope IIIa system equipped with a Nanoscope heater and heater controller. The measurements were done in tapping mode. We used commercial silicon tips (Olympus OMCL-AC160TS-W2) with a spring constant of 66 N/m and a typical resonance frequency around 370 KHz. In tapping mode AFM, the cantilever oscillates with a frequency close to its resonance value and the damping of the oscillation amplitude due to the interaction with the sample surface serves as the feedback signal. In order to limit the interactions between the tip and the smectic surface to a minimum, it is important to work at a damped oscillation amplitude value which is not smaller than 70% of the free oscillation amplitude. The free amplitude value was usually set to 1 V (driving voltage \approx 120 mV). The working amplitude set point during the image recording was then set as high as possible (usually 0.7-0.8 V). The typical scanning rate was in the range between 0.2 and 0.4 Hz per line. Additional information on tapping mode AFM of liquid and liquid-crystal surfaces can be found in [25–27]. Figure 1 gives an example of an AFM image; the depth of the FCD-induced surface depressions is determined from the cross-section data obtained from the Nanoscope analysis software.



FIG. 1. (Color online) Top: AFM image of the surface of an 8CB film. The image shows an area of $36 \times 23 \ \mu\text{m}^2$ containing two FCDs. Bottom: cross sections along the line in the above AFM image demonstrating the temperature dependence of the FCD-induced surface depressions (solid line: $T - T_{NI} = -0.3 \ ^\circ\text{C}$, dashed line: $T - T_{NI} = -4.3 \ ^\circ\text{C}$, T_{NI} is the nematic–isotropic transition temperature).

III. RESULTS

Figure 2 shows a schematic cross section of a FCD with diameter 2r in a smectic film of thickness H. If we assume that the layers are completely incompressible (the lines representing the smectic layers in Fig. 2 are then parts of concentric circles), the depth h of the depression in the smectic/air interface is obtained by simple geometric arguments as

$$h = H - \sqrt{H^2 - r^2} \equiv h_0.$$
 (1)

In the following, we designate the values of h which have been calculated by Eq. (1) as h_0 . Typical values for our experimental systems are $H=40 \ \mu\text{m}$ and $r=8 \ \mu\text{m}$ which would result in $h_0=800 \ \text{nm}$. However, the actual h values measured in our experimental systems are in a broad range and in many cases considerably smaller than h_0 . Thus, we



FIG. 2. (Color online) Schematic cross section through a toric FCD with diameter 2r in a smectic film of thickness *H*. The bend deformation of the smectic layers leads to a depression in the film/ air interface with depth *h*. If the cross sections of the layer planes form concentric circles, the value of *h* is easily calculated according to the Pythagorean theorem (triangle indicated by the dashed lines).

TABLE II. Film thickness *H*, FCD diameter 2r, and the resulting calculated [Eq. (1)] depth h_0 of the FCD-induced surface depression for the six one-component samples and three binary 8CB/10CB mixtures, designated by the mol fraction x_{10CB} of 10CB, under investigation.

Sample	<i>H</i> (μm)	$2r$ (μ m)	h_0 (μ m)
A7	44	19	1.03
C7	43	16	0.75
12CB	42	10	0.31
10CB	41	11	0.35
$x_{10CB} = 0.75$	43	12	0.42
$x_{10CB} = 0.5$	42	13	0.52
$x_{10CB} = 0.25$	45	14	0.56
8CB	45	18	0.91
8.S.5	46	19	0.99

describe the experimental behavior by means of the ratio h/h_0 which can be seen as a measure of how close an experimental system is to the idealized structure of Fig. 2 for which $h/h_0=1$. Table II gives for each sample the values of film thickness *H*, domain diameter 2r, and calculated depth of the surface depression h_0 .

A. FCDs in the smectic-A phase

Figure 3 shows the temperature dependence of h/h_0 in the smectic-A phase for the six compounds under investigation. The data were collected with samples possessing approximately the same film thickness H (cf. Table II). Figure 3 seems to indicate that we can divide the LCs into two groups. In the four compounds possessing a direct



FIG. 3. Temperature dependence of the ratio h/h_0 [h: measured depth of the FCD-induced surface depressions and h_0 : calculated values using Eq. (1)] in ≈ 40 - μ m-thick films of the compounds under investigation. The four upper data sets belong to compounds possessing a smectic-A-isotropic transition: 12CB (\odot), A7 (\bigcirc), C7 (\blacksquare), and 10CB (\diamond). The two lower data sets belong to compounds possessing a smectic-A-nematic transition: $\bar{8}$. S.5 (\blacktriangle) and 8CB (\square). $T_{A \leftrightarrow N/I}$ designates the smectic-A-nematic or smectic-A-isotropic transition temperature (solid lines are just guides to the eyes).



FIG. 4. Temperature dependence of the ratio h/h_0 [h: measured depth of the FCD-induced surface depressions and h_0 : calculated values using Eq. (1)] in \approx 40- μ m-thick films of the compounds 10CB (\diamond), 8CB (\Box), and three 8CB/10CB mixtures with x_{10CB} equal to 0.25 (*), 0.5 (+), and 0.75 (\blacklozenge). $T_{A \leftrightarrow N/I}$ designates the smectic-*A*-nematic or smectic-*A*-isotropic transition temperature (solid lines are just guides to the eyes).

smectic-A-isotropic transition, h is not or only weakly dependent on temperature and the values of h/h_0 are in the range from 0.8 to 1. In contrast, the two compounds possessing a smectic-A-nematic transition exhibit a pronounced temperature dependence of h and considerably smaller values of h/h_0 in the range from ≈ 0 to 0.5.

The different behaviors of smectic-A-isotropic and smectic-A-nematic compounds have been already described by means of the compounds 8CB and 10CB [19]. The present study confirms this behavior also for nonpolar smectic compounds. It is reasonable to ascribe the temperature dependence of h in the smectic-A-nematic case to the second-order nature of the phase transition and the decrease in the smectic layer compressibility modulus B. We will discuss this point in Sec. III C.

In order to explore in more detail the different behaviors of compounds possessing smectic-A-isotropic and smectic-A-nematic transitions, we have studied mixtures of 8CB and 10CB. Figure 4 shows the temperature dependence of h/h_0 for three 8CB/10CB mixtures with a mol fraction of 10CB, x_{10CB} , equal to 0.25, 0.5, and 0.75. In the mixture with $x_{10CB} = 0.25$, the smectic-A-nematic transition is still secondorder but close to a tricritical point (located at $x_{10CB} \approx 0.33$ [28]). Compared to pure 8CB, the values of h/h_0 are considerably larger. The mixture with $x_{10CB}=0.5$ possesses a first order smectic-A-nematic transition; the values of h/h_0 , however, differ only slightly from those of the $x_{10CB}=0.25$ mixture. Finally, the mixture with $x_{10CB} = 0.75$ possesses, as pure 10CB, a direct smectic-A-isotropic transition; the values of h/h_0 are lower than in pure 10CB and the temperature dependence is close to the transition more pronounced than in 10CB. However, in general it seems difficult to conclude from the values of h/h_0 and their temperature dependence on the nature of the smectic-A-nematic transition or even to distinguish between smectic-A-nematic and smectic-Aisotropic transitions.

B. FCDs in the smectic-*C* phase

Three of the compounds under investigation possess a smectic-C phase below their smectic-A phase, and we have



FIG. 5. Temperature dependence of the ratio h/h_0 [*h*: measured depth of the FCD-induced surface depressions and h_0 : calculated values using Eq. (1)] in ≈ 40 - μ m-thick films of the compounds A7 (\bigcirc), C7 (\blacksquare), and $\overline{8}$.S.5 (\blacktriangle). $T_{C \leftrightarrow A}$ designates the smectic-*C*-smectic-*A* transition temperature (solid lines are just guides to the eyes).

measured the values of h/h_0 across the smectic-C-smectic-A transition (cf. Fig. 5). For the compound $\overline{8}$. S.5, we observe a pronounced decrease in the h/h_0 values which starts ≈ 0.5 °C above the transition and continues within a temperature range of 2 °C below. At still lower temperatures, the values remain essentially constant in the smectic-Cphase. The compound C7 shows a different behavior: at the transition from the smectic-A to the smectic-C phase, a small sharp decrease in h/h_0 is observed, then the values increase in the smectic-C phase with decreasing temperature until values close to 1 are reached. For the compound A7, the smectic-C-smectic-A transition itself seems to have no effect on the magnitude of h/h_0 , ≈ 1 °C below the transition the values start a slight increase to $h/h_0 \approx 1$. We should note that both compounds C7 and A7 possess a transition to a higherordered phase, probably crystal G, at a temperature 10 $^{\circ}$ C (C7) or 2.5 °C (A7) below their smectic-C-smectic-A transition. We discuss the behavior of h/h_0 in the following section.

C. Relation to the layer compression modulus B

The presence of the FCD-induced depressions increases the area of the film/air interface compared to a completely flat interface. In principle, this results in a dilative stress for the smectic layers below the depressions because an increase in the thickness of the smectic film in the central region of the FCDs would result in an energy gain $\sigma_{air}\Delta A$ (with σ_{air} being the film/air interface tension and ΔA the corresponding decrease in the interface area). Indeed we observe in the present study in several cases values of h/h_0 smaller than 1, indicating that the film thickness in the central region of the FCDs is larger than in the idealized structure shown in Fig. 2. One can think of two mechanisms which could lead to an increase in the local film thickness in the central region of the toric FCDs: the creation of new smectic layers, i.e., the generation of edge dislocations around the straight singular line of the FCDs, or a dilation of the smectic layers in this region.

The compounds 8CB and $\overline{8}$. S.5 show a pronounced temperature dependence of h/h_0 , which increases with decreasing temperature from a value of ≈ 0.05 close to the transition to the nematic phase to a value of 0.2 (8CB) or 0.5 ($\overline{8}$. S.5) at temperatures 5 °C below the transition. This behavior is reversible with respect to the direction of the temperature change. If the creation of edge dislocations would be responsible for the temperature dependence of h, edge dislocations would have to be continuously and reversibly generated and annihilated with increasing or decreasing temperature, respectively, which is an unlikely scenario. On the other hand, a complete cooling/heating cycle for one sample has in our experiment a duration of about 2 days. Given these long time periods, a contribution from a mechanism based on edge dislocations might not completely to be ruled out. However, because of the following arguments, we believe that the temperature dependence of h and the deviation of h/h_0 from 1 is mainly a result of the elastic properties controlling the compression or dilation of the smectic layers.

The local energy density f of an elastic distortion of a smectic-A phase can be written as [29]

$$f = \frac{1}{2}K_1c^2 + \frac{1}{2}Be^2.$$
 (2)

The first term describes the energy related to a splay of the director field $\mathbf{n}(\mathbf{r})$ (K_1 denoting the splay elastic constant and $c = \operatorname{div} \mathbf{n}$), and the second term describes the energy related to a compression or dilation of the smectic layers [*B* denoting the layer compression modulus and $e = (d-d_0)/d_0$ the deviation of the actual layer thickness *d* from its equilibrium value d_0].

A number of studies are concerned with measurements of B at the smectic-A-nematic transition: the studies are based either on light scattering measurements (yielding the ratio B/K_1 [30–34] or on mechanical techniques enabling the determination of absolute values of B[35-45]. The behavior of B has been studied also at transitions between the smectic-A phase and different types of smectic-C phases [46-48] and near a smectic-C-hexatic-I critical point [49]. When a smectic-A-nematic transition is approached from below, Bdecreases with increasing temperature to values close to zero at the transition. The behavior can be described either by a simple power law or by more complex models in which a small but finite value of B at the transition is assumed (the nature of the critical behavior of B is still under investigation, recent studies indicate, that the observation of a finite value of B at the transition might result from a dynamical effect [43,45]).

A small value of *B* allows for a dilation of the smectic layers in the central region of the FCDs leading to small values of h/h_0 as observed for 8CB and $\overline{8}$.S.5 on approaching the transition to the nematic phase. For both compounds,

measurements of the absolute value of *B* have been carried out [38,40,41]. At a given temperature difference to the smectic-*A*-nematic transition, the magnitude of *B* is in $\overline{8}$.S.5 approximately four times larger than in 8CB. Accordingly, the values of h/h_0 should be in $\overline{8}$.S.5 closer to 1 than in 8CB, as is indeed the case (cf. Fig. 3): the h/h_0 values are in $\overline{8}$.S.5 approximately two times larger than in 8CB. Unfortunately, we did not find any measurements of *B* for 10CB or 12CB (or any compound with a direct smectic-*A*-isotropic transition). The h/h_0 values of these compounds are close to 1, and we would expect considerably larger *B* values than in 8CB.

The behavior of B at smectic-A-smectic-C transitions has been studied for a number of compounds but not for 8.S.5, A7, or C7. In [47,48], a minimum of B at the transition temperature has been observed, which is followed by an increase in B with decreasing temperature within the smectic-Cphase. For 8.S.5, we find that the h/h_0 values decrease as the transition to the smectic-C phase is approached, but within the smectic-C phase, the values stay at a low constant level. For the compounds A7 and C7, we observe, if at all, only a minor effect of the transition to the smectic-C phase on the values of h/h_0 . Although the values of h/h_0 in these compounds are already close to 1, they increase slightly with decreasing temperature in the smectic-C phase. A pronounced increase in B in a smectic-C phase has been observed near a smectic-C-hexatic-I critical point [49]. The increase in h/h_0 in the smectic-C phase of A7 and C7 might also result from the development of hexatic order since these compounds show, in contrast to $\overline{8}$. S.5, a more ordered phase below their smectic-C phase.

In addition to the qualitative arguments described above, we tried a more quantitative test of the relation between h/h_0 and *B* by comparing the energy Δw_{surf} gained by a decrease in the surface area (by a decrease in *h*) and the energy Δw_{dil} to be paid for an increase in the film thickness (by a dilation of the smectic layers). If we approximate the shape of the FCD-induced surface depression by a right circular cone with axis length *h* and base diameter 2*r*, Δw_{surf} is given by

$$\Delta w_{surf} = \sigma_{air} \pi r (\sqrt{r^2 + h^2} - \sqrt{r^2 + h_0^2}).$$
(3)

In order to estimate the magnitude of Δw_{dil} , we treat a FCD in a rough approximation as a smectic cylinder with height *H*, diameter 2*r*, and elastic modulus *B*; we ignore the curved arrangement of the smectic layers and assume that the layers form a simple stack with the layer normal being everywhere parallel to the cylinder axis (a detailed theoretical description of the curvature energy of a FCD has been given in [50]). Δw_{dil} is then given by integrating the second term of Eq. (2) over the volume of the cylinder:

$$\Delta w_{dil} = \int_{V} \frac{1}{2} B e^2 dV. \tag{4}$$

We assume that the dilation *e* decreases linearly with radial distance ρ from the center of the FCD, i.e., at the center of the FCD (ρ =0), $e=(h_0-h)/H$, and e=0 at the perimeter (ρ =*r*):



FIG. 6. (Color online) Solid lines: values of h/h_0 as function of modulus *B*, calculated using Eqs. (3) and (6) with σ_{air} =0.03 N/m, *H*=45 μ m, and *r*=5 μ m or *r*=10 μ m. For the dashed lines, the intrinsic dilation sheath (see text) of the FCDs was additionally taken into account. Gray areas: experimental values of h/h_0 (this study) and *B* (determined with second sound measurements, from [40]) for the compounds 8CB and $\overline{8}$.S.5.

$$e = \frac{h_0 - h}{H} \left(1 - \frac{\rho}{r} \right). \tag{5}$$

The integration in Eq. (4) then gives

$$\Delta w_{dil} = \frac{\pi B (h_0 - h)^2 r^2}{12H}.$$
 (6)

From the equilibrium condition $\Delta w_{surf} + \Delta w_{dil} = 0$ we can calculate h/h_0 as a function of B for given values of r, H, and σ_{air} . Figure 6 shows two examples obtained with σ_{air} =0.03 N/m, H=45 μ m, and r=5 μ m or r=10 μ m, respectively. The positions of the experimental data for the two compounds 8.S.5 and 8CB, for which both B and h/h_0 are available, are also indicated in Fig. 6. It is obvious that there is a large discrepancy. According to our model, we should measure for 8CB and $\overline{8}$. S.5 either considerably larger values of h/h_0 or these two compounds should possess values of B 1 or 2 orders of magnitude smaller than reported. One reason for this discrepancy is certainly the simplicity of our model which neglects the curved smectic layer structure in a FCD. For instance, it was shown that the smectic layers in a FCD show an intrinsic dilation near the two singular lines of the FCD [51]. The magnitude Δh_i of the resulting change in h has been estimated as [19]

$$\Delta h_i = -\lambda \, \log \frac{H}{\lambda},\tag{7}$$

with $\lambda = \sqrt{K_1/B}$. However, taking this intrinsic dilation into account in our model (setting the splay elastic constant K_1 to a typical value of 10^{-11} N) results only in a minor modification of the predicted relation between h/h_0 and *B* (cf. dashed lines in Fig. 6).

A possible explanation of the discrepancy between the small B values resulting from our simple model and the larger values reported in the literature could consist in the dependence of B on the frequency used for its determination:

for most measurements of the absolute magnitude of B the second sound technique [36] was used which is based on a propagation mode supported by the smectic layers. These measurements are carried out in the kHz range, whereas our determination of h/h_0 is a purely static experiment. The frequency dependence of B at low frequencies has been studied in [45], where a pronounced decrease in B with decreasing frequency in the range from 10 to 1 Hz was observed. On the other hand, there are also low-frequency measurements of B[52] which yielded similar results as the second sound measurements. Thus, although there are indications that the static value of the modulus B is smaller than the values obtained from second sound measurements, it is not clear how large this difference is and if it is sufficient to explain the discrepancy between the small B values resulting from our simple model and the larger values determined by second sound measurements.

It is obvious that a more elaborated model, taking the arrangement of the smectic layers in a FCD into account, is needed for a complete description of our results. Furthermore, although the spontaneous generation of edge dislocations with increasing temperature is unlikely, there may be other mechanisms. In a recent study [53] on the shape of smectic droplets and their change with temperature a similar problem was encountered. In [53] it was shown that the radius of the droplet facet behaves approximately reversible with temperature although different mechanisms determine the change in the droplet shape during cooling and heating.

IV. CONCLUSION

Using AFM measurements, we have determined the temperature dependence of the depth h of the FCD-induced depressions in the surface of smectic films of several compounds possessing various phase sequences. Generally, we find that the values of h can vary in wide range from values close to h_0 (the value expected by geometric arguments for incompressible layers) down to values considerably smaller than h_0 . In the smectic-A phase of compounds showing a smectic-A-isotropic transition (10CB, 12CB, A7, and C7) h amounts to values relatively close to h_0 , i.e., $h/h_0 \approx 0.8$ to 1. The compounds possessing a second-order smectic-A-nematic transition (8CB and $\overline{8}$. S.5) show in their smectic-A phase considerably smaller values of h/h_0 ranging from 0.5 far below the transition to the nematic phase down to 0.05 near the transition. There is, however, no sharp boundary between these two groups of compounds: the study of a binary system of 8CB and 10CB shows that h/h_0 varies smoothly as one proceeds from second-order smectic-A-nematic over first-order smectic-A-nematic to smectic-A-isotropic systems.

Three of the compounds under investigation show a smectic-*C* phase below their smectic-*A* phase. In $\overline{8}$.S.5, h/h_0 decreases considerably at and below the transition to the smectic-*C* phase. In the other two compounds, A7 and C7, we observe a completely different behavior: in the smectic-*C* phase, h/h_0 increases to ≈ 1 with decreasing temperature.

We have discussed our experimental observations in relation to the layer compressibility modulus *B*. A small value of B would allow for a dilation of the smectic layers in the central region of the FCDs and thus result in a decrease in the value of h. This mechanism would qualitatively explain the decrease in h/h_0 on approaching a second-order smectic-A-nematic transition and also the smaller values of h/h_0 in 8CB compared to the values of 8.S.5 (since 8CB shows also smaller values of B compared to $\overline{8}$. S.5 [40]). A reason for the different behaviors in the smectic-C phase might consist in the development of hexatic order which was shown to increase the value of B considerably [49]. The compounds A7 and C7 show a crystal-G phase below their smectic-C phase indicating a possible tendency for the development of hexatic order in the smectic-C phase. The compound $\overline{8}$. S.5 does not show a more ordered hexatic or crystal phase below its smectic-C phase. However, we have no explanation for our finding that in 8.8.5 the h/h_0 values stay on a low level with decreasing temperature in the smectic-Cphase (whereas measurements of B have shown in several cases that *B* increases with decreasing temperature [48]).

For two of the compounds under investigation, 8CB and $\overline{8}$.S.5, absolute values of *B* in the smectic-*A* phase are avail-

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able. In a simple model, we have compared the energy which is gained by a decrease in the surface area resulting from a decrease in the depression depth h with the energy that is required for a corresponding dilation of the smectic layers. The two energies are of similar size only if we assume that the static B value of our experiment is 1–2 orders of magnitude smaller than the values obtained from second sound measurements in the kHz range. Although B was shown to decrease with decreasing frequency, this result shows that our model is too simple to draw accurate quantitative conclusions. Nevertheless, the qualitative indications of a relation between h/h_0 and B described above suggest that the depth of the FCD-induced surface depressions is mainly controlled by the layer elastic modulus of the smectic liquid crystal.

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