Stepwise transition of a topological defect from the smectic film to the boundary of a dipolar inclusion

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Cholesteric droplets accompanied by a topological defect are studied in free standing smectic C^* films. We observed a transition between two droplet-defect configurations with the defect in the film and on the droplet boundary. We found that the distance between the droplet surface and the topological defect decreases continuously with increasing temperature and above a certain critical temperature the defect jumps to the droplet boundary. We relate this stepwise change in the defect position to the change in the anchoring on the droplet boundary. This transformation leads to a decrease in the interparticle distances in self-organized chains from droplets. Our simple theory allows us to estimate the value of the anchoring energy.

I. INTRODUCTION

Liquid crystals are ideal materials for studying topological defects. Various inclusions in orientationally ordered liquid crystals typically produce defects and singularities of the director field easily visible by polarized light microscopy techniques. All this is related to the anchoring phenomenon, i.e., the tendency to orient liquid crystal molecules when they contact the surface of the inclusion. Investigations in this field became essentially actual in recent years due to the discovery of strong interaction between inclusions and their self-organization in liquid crystal media [1–11]. Self-organization refers to processes where pre-existing components (inclusions and companion topological defects in our case) assemble themselves without external intervention. The ability to reversibly manipulate the self-aggregation provides a basis to design microstructures that can be controlled. These wonderful effects are connected with formation of topological defects in the liquid crystal near inclusions. The simplest, infinitely strong anchoring assumption, that is, that the orientation of the liquid crystal is firmly fixed on the surface, means that the surface imposes boundary conditions for the orientational field. However it is clear that coupling for many realistic situations can be weak. For instance, finite anchoring was found in a smectic film at a layer step forming the boundary between areas of different thickness [7]. On a phenomenological level finite anchoring can be described by adding an appropriate surface potential W to the free energy of the system. However despite the vast amount of available literature on interaction and self-organization of inclusions [1–11] the topic of finite coupling on the inclusion surface is still full of challenging questions. Mostly two limited cases were considered theoretically in detail: (a) strong anchoring [2,12] when topological defects form near the inclusion, (b) weak anchoring [13,14] when topological defects are not formed. These limiting cases lead to essentially different behavior both for individual inclusions and their assembly. The aim of our paper is to clarify the behavior for temperature dependent defect positions at a finite anchoring at the surface of an inclusion with a topological defect in a thin ferroelectric smectic C^* (SmC^*) film, in which molecules tilt with respect to the layer normal [15].

This deceptively simple problem in fact is a quite complicated one. Even the implementation of numerical methods has serious problems because one has to treat with a comparable high accuracy two very different length scales (for the inclusion and the defect) [16,17]. In two-dimensional systems, like thin free standing films, additional complications occur. Unlike three-dimensional nematics where orientational ordering is characterized by a headless unit vector (director) **n** with a symmetry **n** → −**n**, orientational order in SmC films is defined in terms of so-called **c** director which is a conventional polar vector **c** [15]. It means that there is no **c** → −**c** symmetry, only combined symmetry of simultaneous inversions of **c** and unit vector **l** orthogonal to smectic layers. A full theoretical analysis of this problem is very difficult because one has to deal with highly nonlinear differential equations in a complex geometry (and besides functional form of the appropriate surface energy is not known and thermal fluctuations play a relevant role in two dimensions). With all these arguments in mind (and also guided by a desire to make formulas simpler to clarify major features of the physical behavior) in this work we combine experimental and semiqualitative heuristic tools.

The problem is well elaborated for SmC in the one constant approximation for infinitely strong surface anchoring. In this case one can easily find the defect position by simply equilibrating the forces acting on the defect. It is well known that in a good approximation to satisfy boundary conditions on the inclusion surface and at infinity, one has to include at least two singular points: one singularity outside the inclusion (at a distance r_d from the inclusion center) and its virtual image inside the inclusion at the distance $r_d' = r_0^2 / r_d$, where r_0 is the inclusion radius. This force balance equation for a singularity with topological charge S (in the case considered in this paper we are interested in $S = -1$) reads as

$$\frac{S(S-1)}{r_d} - \frac{S^2}{r_d - r_d'} = 0. \tag{1}$$

On a theory side, this problem was addressed first by Pettey, Lubensky, and Link [12]. The authors calculated the position of a topological defect with charge $S=-1$ near a circular inclusion in the limit of strong anchoring. The distance scales with the inclusion radius and equals $r_d = \sqrt{2}r_0$.

The question is what effects may change the position of the topological defect and so influence the structure of the inclusion-defect pairs and their interaction. Korolev and Nelson [18] predicted an essential increase in the average droplet-defect distance due to anharmonic asymmetry of the interaction potential between the droplet and the defect. The effect should depend on the dimensionless ratio $\varkappa = k_B T/K$ where K is the orientational two-dimensional (2D) elastic constant with dimension of energy (erg). Increase in $\varkappa$ from 0 to 1 should lead to an increase in the relative droplet-defect distance r_d/r_0 from 1.41 to more than 1.9.

Let us now consider the case of a finite anchoring strength W [19]. For a finite W we may assume the boundary condition to be satisfied at a certain smaller than r_0 circle. This is the essence of de Gennes extrapolation length $\xi = K/W$. In our case W is the anchoring energy per unit boundary length. Formally, for $\xi \neq 0$ the **c**-director deformation can be continued beyond the inclusion surface to $r_0 - \xi$. In this spirit one has to put the image defect at $r_d' = (r_0 - \xi)^2/r_d$. Therefore combining everything together we find the equilibrium condition

$$\frac{r_d}{r_0} = \left(1 - \frac{\xi}{r_0}\right)\sqrt{1-S}, \tag{2}$$

and for $S=-1$, $r_d = (r_0 - \xi)\sqrt{2}$. This simple consideration shows that finite anchoring W decreases the equilibrium inclusion-defect distance. Certainly the anchoring should not be too weak (i.e., ξ must be smaller than r_0), otherwise there is no need for defects at all. This is our scenario, and although a much more elaborated theory and numeric consideration is needed, as a first semiqualitative approach, we utilize it to confront with experimental observations.

So two opposite effects can be expected in liquid crystals. In the first one the inclusion and the topological defect are moving farther apart due to decreasing of the elastic constant with respect to temperature. The parameter characterizing this effect is $\varkappa = k_B T/K$, that should be of the order of unity for the effect to be observed. By contrast, in the second one the topological defect approaches the inclusion due to decreasing anchoring on the inclusion surface. The parameter characterizing this effect is the dimensionless ratio $\nu = K/W r_0$. Both effects have simple qualitative explanations. If $k_B T > K$ the fluctuations of the defect position become large and the defect-droplet distance increases. If W is much less than K/r_0 the defect must reside on the inclusion surface.

To clarify the question whether the defect position may differ from the well-known theoretical prediction $r_d = \sqrt{2}r_0$ [12] we have performed studies of droplets with companion defects in a wide temperature range. The results of investigations exceeded our expectations. We found that the dis-

tance between the droplet surface and the topological defect may change substantially with temperature and the defect jumps to the droplet boundary above a critical temperature. We relate this change to the decrease in the anchoring energy on heating.

II. EXPERIMENT

We investigate droplet-defect pairs in ferroelectric SmC^* free standing films. The liquid crystal used here is S-4′-undecyloxybiphenyl-4-yl 4-(1-methylheptyloxy) benzoate (11BMSHOB) [20]. In the bulk sample the sequence of phase transitions was $SmC^* - (108\ °C) - N^* - (123.9\ °C) - I$. Films were prepared by spreading the liquid crystal in the SmC^* phase across a 4 mm circular hole in a thin glass plate. Films with thickness N from 8 to 30 smectic layers were used. Due to the surface ordering [21,22] smectic films exist at the temperature higher than the bulk smectic-cholesteric transition. In thick films ($N>20$) at heating the cholesteric droplets nucleate in the films near the temperature of the bulk transition from smectic to cholesteric phase. In thinner films the droplets nucleate at higher temperatures below the thinning transitions [23,24]. Topological defects and droplets were observed with an optical microscope in the reflection mode using depolarized reflected light microscopy (DRLM) [25] and polarized reflected light microscopy (PRLM). The images of the films were taken with a charge-coupled device camera connected to a computer, then digital image processing was used.

Dipolar inclusions formed by a droplet (intrinsic topological charge $S=+1$) and a companion $S=-1$ topological defect were investigated. It has been shown before that the topological defect may be situated on the droplet boundary or on some distance from it in the bulk of the film [26]. In this paper we focus on inclusions with defects located in the bulk of the film. Attractive interaction of dipolar droplets with each other and with Coulombic droplets [26] leads to formation of various chainlike structures. Interaction between inclusions on large and short distances, the interparticle distances in self-assembled structures critically depend on the position of the topological defect near the droplet.

III. RESULTS AND DISCUSSION

We found that the position of the topological defect depends drastically on the temperature. At low temperature the position of the defect is in good agreement with the classical theory of Pettey, Lubensky, and Link [12] based on the electromagnetic analogy. This situation is illustrated by Fig. 1(a). The orientation of the **c** director on the droplet boundary is tangential, that is, the **c** director is parallel to the droplet boundary. The defects near isolated inclusions are situated on the distance $r_d/r_0 = 1.41 \pm 0.02$ from the droplet center. We remind that according to the model with strong anchoring on the inclusion boundary [12] $r_d = \sqrt{2}r_0$.

Figure 1(b) shows dipolar droplets self-organized in a linear chain with $S=-1$ topological defects between the droplets. The droplet-droplet distance in chains in which the droplets are bound by bulk $S=-1$ defects was numerically
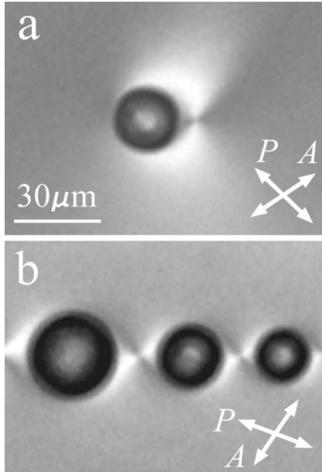
FIG. 1. A dipolar droplet-defect pair (a) and a chain of three droplets (b) in a smectic film. The topological defects are located between the inclusions. At low temperature the defect is situated at a relative distance $r_d/r_0 \approx 1.41$ from the isolated droplet (a). The droplet-droplet separation in the chain equals $D/r_0 = 2.86$ (b). The orientation of the polarizer and the analyzer are shown in the lower part of each frame. $T = 108.0$ °C (a), $T = 107.9$ °C (b).

calculated by Patricio *et al.* [17] and Korolev and Nelson [18]. According to their calculations, the distance between centers of adjacent droplets D equals approximately the same value $D/r_0 = 2.82 \pm 0.01$ [17] and $D/r_0 = 2.878$ [18]. The droplet-droplet separation in the chain in our case [Fig. 1(b)] equals $D/r_0 = 2.86 \pm 0.05$, where r_0 is the average radius of two droplets which is also consistent with the theory for droplet interaction with strong boundary conditions [17,18]. The position of the defect between the droplets correlates with the distance between a single droplet and a defect. If the droplets are of equal size, the defect is in the middle between them. The defect situates closer to the smaller droplet when the droplets in the chain differ in size.

With temperature the configuration of the droplet-defect pair changes. On heating the separation between the droplet and the defect diminishes (Fig. 2) and the defect approaches the inclusion boundary. In Fig. 2(a) the distance from the droplet center to the defect r_d is about $1.41r_0$. On heating the distance from the droplet to the topological defect continuously decreases [Fig. 2(b), $r_d/r_0 = 1.255$]. However at the temperature of about $T = 109.1$ °C a stepwise transition between two equilibrium defect positions occurs. At high temperatures ($T > 109.1$ °C) the equilibrium situation corresponds to the defect on the droplet boundary [Fig. 2(c)]. The transition takes some time to occur. At constant temperature the defect approaches the droplet boundary and resides on it. The movement occurs within several seconds. Figure 3(a) shows the temperature dependence of the distance r_d/r_0. Closed symbols correspond to equilibrium states. Around $T = 108.0$ °C the distance is $r_d/r_0 = 1.41$ within the accuracy of measurements. The dotted arrow shows the transition with a stepwise change in the equilibrium defect position on heating. At high temperature ($T > 109.1$ °C) a nonequilibrium state with position of the defect inside the gap $1 < r_d/r_0 \lesssim 1.25$ can be obtained on rapid heating. Figure 2(d) shows a
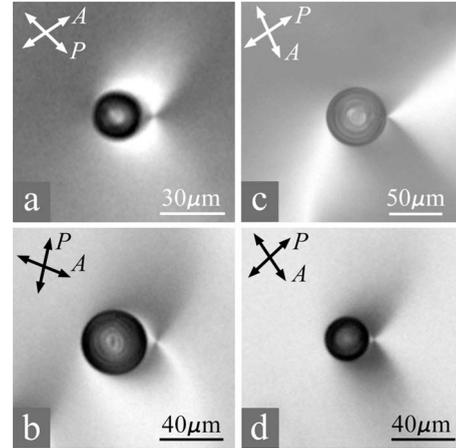

FIG. 2. Optical microscope pictures of dipolar droplets. The distance from the droplet center to the topological defect near an isolated droplet decreases with temperature: $r_d/r_0 = 1.41$, $T = 108.0$ °C (a) and $r_d/r_0 = 1.255$, $T = 108.9$ °C (b). At higher temperature above $T = 109.1$ °C the defect resides on the droplet boundary (c). Frame (d) shows a nonequilibrium situation when on rapid heating up to $T = 109.2$ °C the defect is moving toward the boundary of the droplet. This situation corresponds to the open circle in Fig. 3(a). The images were taken in DRLM.

nonequilibrium situation when after a rapid heating up to $T = 109.2$ °C the defect is moving toward the boundary of the droplet. The nonequilibrium state is shown by an open circle in Fig. 3(a). Such dynamic behavior is typical for discontinuous, first-order phase transitions, and it is consistent with observed stepwise jumps of the defect position and hysteresis phenomena. Although dynamic studies are likely to be more fruitful than static equilibrium ones, this field is still in a rather underdeveloped stage. This is largely accounted for by a complexity of dynamic nonequilibrium phenomena in smectic films with inclusions and topological defects. Defect motion is determined by a subtle interplay of hydrodynamic backflow and intrinsic dynamics of the orientational order parameter. We will not consider dynamic phenomena in this paper deferring detailed investigations to another work.

For samples with some change in the temperature of the bulk phase transition due to impurities the temperature of the transition of the defect to the droplet boundary also changes. At low temperature the defect position corresponding to strong anchoring could be observed in all investigated samples.

After the defect resides on the droplet boundary we observed two possibilities of the defect behavior on cooling. If the heating was only slightly higher than the temperature of the transition (about 0.1 °C above the transition temperature) and the defect was on the boundary for a short time (several seconds) as a rule on cooling the defect left the droplet boundary and moved back to a distance approximately equal to that before the transition. Typically a hysteresis occurs and in these cases the transition on cooling takes place at a slightly lower temperature than on heating (the difference is within 0.1 °C). If the heating was more sufficient and the film stayed at high temperature for a longer time (several tens of seconds) as a rule on cooling the defect
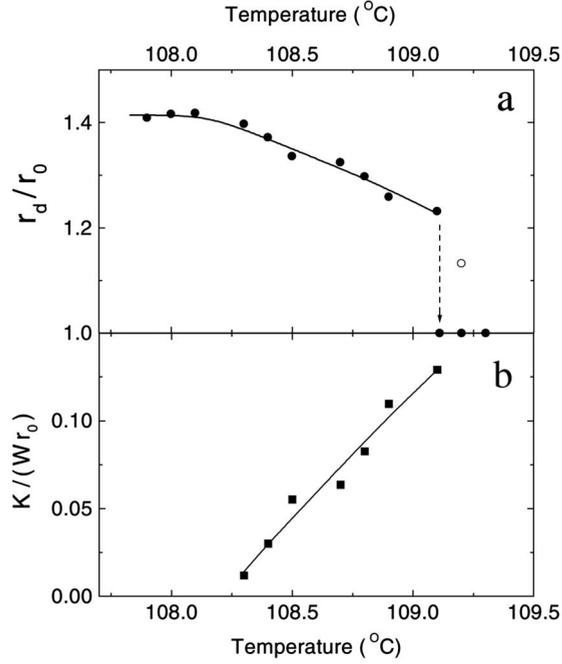
FIG. 3. Temperature dependence of the defect position near a droplet in a smectic film (a). On heating r_d/r_0 continuously decreases until the critical temperature $T=109.1$ °C above which the defect sits on the droplet boundary ($r_d=r_0$). The dotted arrow shows the transition on heating. The open circle shows r_d/r_0 in a state above $T=109.1$ °C that can be obtained on rapid heating. This state is nonequilibrium; the defect is moving toward the droplet boundary and within a short time sits onto it. Above $T=109.1$ °C the stable state corresponds to the defect on the droplet boundary, $r_d=r_0$. (b) The dimensionless ratio $\nu=K/Wr_0$ calculated from the measured defect position. At low temperature $T<108.2$ °C the ratio is $\nu=0$ within the accuracy of its determination. The thickness of the film is about 15 smectic layers. The droplet radius r_0 is about 27 μm.

remained on the droplet boundary. In this case the behavior of the droplet did not differ from the case of a droplet nucleated with a surface defect [26]. So two states of the topological defect on the droplet boundary exist: the first state in which the defect can leave the boundary and the second state in which the defect does not leave the droplet boundary. We suppose that the transition between the two states is kinetically driven and takes place when the defect is kept near the droplet boundary for a long time. The local structure of these two states is not yet clear, as well as the structure of the defect core in SmC^* films. We may guess that in one case the core of the topological defect remains separated from the droplet, whereas, in the other case the core of the defect joins with the droplet. We may guess also that between these two states there is a potential barrier which is somewhat higher than $k_B T$ at high temperature (so the transition between two states may take place). At large distances the **c**-director configuration in these two states is similar. At small distances the van der Waals interaction and ionic impurities may be essential and play an important role in the interaction and aggregation of the defect and the droplet. At low temperature the potential barrier becomes substantially higher than $k_B T$. This explains the existence of dipolar droplets at low temperature
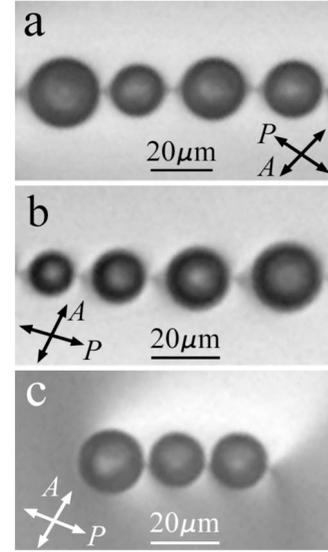

FIG. 4. The distance between droplets in self-organized chains depends on temperature. At $T=108.9$ °C (a) the interparticle distance for the droplets with defect between them is $D/r_0=2.5$ [compare with Fig. 1(b) where $D/r_0=2.86$]. A similar decrease in the distance between droplet centers is observed in chains with the defect on the droplet boundary (b,c). At low temperature $T=108.2$ °C (b) the droplet-droplet distance $D/r_0=2.6$. At high temperature $T=109.2$ °C (c) the droplet-droplet distance $D/r_0 \lesssim 2.2$.

both with boundary and bulk topological defects. At low temperature we did not observe a transition between them.

Based on our observations (Figs. 2 and 3) we can propose that the anchoring strength on the droplet boundary W decreases with temperature. This suggestion is supported by the fact that at high temperature a transition from tangential to radial **c**-director orientation on the inclusion boundary may take place [9]. Using our qualitative approach [Eq. (2)] from the experimental data of Fig. 3(a) the ratio $\nu=\xi/r_0=K/Wr_0$ was estimated [Fig. 3(b)]. This dimensionless ratio shows the relative interplay of bulk elastic and surface anchoring energies. The actual **c**-director orientation is such that the elastic torque transmitted by the liquid crystal to inclusion surface is balanced by the surface torque related to the anisotropic surface anchoring. Below 108.2 °C the ratio is $\nu=0$ within the accuracy of measurements. On heating ν essentially increases with temperature. From the value of ν the surface anchoring energy may be estimated. Since 2D elastic moduli and anchoring energy for a film scale approximately like film thickness, taking the bulk elastic constant to be 2×10^{-6} erg/cm [27,28] we get that the anchoring strength for unit boundary area decreases to about 6×10^{-3} erg/cm^2 near the temperature of the stepwise transition. It is worth noting that this value correlates with measurements of the anchoring energy of nematic on a solid surface [29,30].

We found that self-organized structures formed by droplets are also affected by the change in anchoring. A substantial transformation of the interparticle distances with temperature was observed in droplet chains. Figure 4(a) shows a chain of droplets near the temperature of the stepwise transition in dipolar droplets. The distance between centers of droplets separated by the bulk defect [three right droplets in

Fig. 4(a)] decreases to $D/r_0 = 2.5$, which is consistent with the decrease in the droplet-defect distance for individual droplets [Fig. 2(b)]. We remind that $D/r_0 = 2.86$ at low temperature [Fig. 1(b)]. The spacing between droplet surfaces in a chain is about twice the distance from the defect to the boundary of an isolated dipolar droplet.

We also found that the distance in the chains between droplets with defects on their boundary changes with temperature. For such droplets at low temperature the distance between droplet centers in a chain is $D/r_0 = 2.6 \pm 0.1$ [Fig. 4(b)]. On heating the interparticle distance decreases [Fig. 4(a), two left droplets, $D/r_0 = 2.3$] and becomes $D/r_0 \lesssim 2.2$ at higher temperatures [Fig. 4(c), $T = 109.2 \,^\circ\text{C}$]. The decrease in the interparticle distance in the chains formed by droplets with boundary defects may indicate a further decrease in the surface anchoring energy above 109.1 $^\circ$C. For the chains with the defect located between the droplets at such temperature the defect remains in the bulk, although the interparticle distance decreases. This seems to be related to the attraction of the defect from two droplets on the opposite sides.

We may speculate about the origin of the observed effect. On the boundary of the droplet there exists a meniscus of the smectic phase. The mechanism of the decrease in the anchoring energy may be related to the modification of the meniscus with temperature. The **c** director tends to orient tangential on the meniscus boundary (smectic layer step). On heating, the size of the meniscus decreases and the value of the anchoring energy may become smaller. We can assume that the qualitative consideration using the notion of the extrapolation length ξ is valid if the topological defect is not in the vicinity of the droplet surface, namely, $r_d - r_0 > \xi/2$. In our case when the defect is in the bulk of the film this condition is fulfilled [Figs. 3(a) and 3(b)]. When $r_d - r_0 \sim \xi/2$ our consideration becomes invalid and the transition of the defect to the droplet surface takes place.

Let us note several other aspects that were not included in our semiqualitative approach. Whereas in this paper we use the one constant approximation (which enables to simplify the analysis) in reality two-dimensional bend and splay elastic constants can differ in several times [27,28,31,32]. This could influence the equilibrium position of the defect, especially for small droplet-defect separations. It would be interesting to analyze the director configuration for different values of the elastic anisotropy, however it will definitely require employing numerical methods.

Previously the position of a point topological defect near a spherical inclusion in nematic liquid crystal was calculated numerically for different surface anchoring [33,34]. The calculations predict that the inclusion-defect separation depends on the ratio $K^0/W^0 r_0$, where K^0 and W^0 are the elastic constant and anchoring energy in the three-dimensional (3D) system. We remind that K^0/W^0 and K/W have the same dimension and the same sense. As the value $K^0/W^0 r_0$ increases, the defect should approach the inclusion [33,34]. To the best of our knowledge, these theoretical predictions have not been observed experimentally. According to theory, the spacing between the inclusion surface and the defect decreases in two times with respect to the strong anchoring limit as $K^0/W^0 r_0$ increases from 0 to about 0.13 [33,34]. In our experiments (Fig. 3) the spacing between the inclusion

surface and the defect decreases from 0.41 to about 0.23 when $\nu = K/W r_0$ becomes approximately 0.13. So the results of our experiments correlate fairly well with theoretical predictions. We should note that calculations were performed for a 3D system and in the experiment we deal with 2D geometry of the **c** director. However, there is essential difference in the behavior of 3D and 2D systems at further decrease in the anchoring. For the 3D system the calculations predict that at the ratio $K^0/W^0 r_0 > 0.14 - 0.1$ the dipolar configuration with a point topological defect becomes unstable and a transition to the quadrupolar configuration with an equatorial defect (Saturn ring) should occur [33,34]. The transition from the dipolar to the quadrupolar configuration upon changing the size of inclusion was observed in experiment [35]. The transition related to the change in anchoring was predicted for 3D nematic but not observed before. In 2D geometry there is no transition to quadrupolar configuration in the temperature range of our observations. In our experiment on heating the defect resides on the droplet boundary (Figs. 2 and 3) but the configuration remains dipolar.

Let us now discuss the possibility for the opposite effect, namely, increase in the defect-droplet distance predicted by Korolev and Nelson [18]. Assuming a reasonable accuracy of measurements, we estimate that this effect can be observed if $\varkappa = k_B T/K \geq 0.1$. In smectic liquid crystals the typical value of $\varkappa$ for a film with thickness $N \sim 15$ is less than 10^{-2}. Liquid crystal phases exist in a limited temperature range that does not allow changing the temperature essentially. So if this effect exists it may be observed only by means of decreasing K in the vicinity of the temperatures of SmC–SmA or thinning transitions [23] where the elastic modulus becomes small.

The properties of the droplet boundary are mainly described by two physical quantities: the anchoring energy W and the surface tension σ. A dimensionless ratio W/σ shows the relative importance of the two energies. If W/σ changes with temperature, one would expect a competition of the anchoring energy and the surface tension with liquid crystal elasticity [36,37]. If σ dominates, it corresponds to the situation discussed in this paper. The droplets have circular shape due to the tendency to minimize the length of the boundary. However, the orientation of the director on the inclusion boundary for small W may change and the elastic-mediated repulsion between the droplet boundary and the defect decreases. In the limit of very small W the companion topological defects disappear. We remind that the topological defect can disappear only on the droplet surface. In the opposite case of large ratio W/σ the droplet shape may differ from circular, but the director orientation on the boundary remains planar. Such behavior was observed in films of nonpolar SmC [38].

In summary, many experimental and theoretical studies were performed in recent years to resolve challenging questions of the defect-mediated interaction and self-organization of inclusions in liquid crystals. The key point of this topic is the interaction of inclusions with liquid crystal media. Most theories and numerical simulations are based on the strong anchoring model of the inclusion boundary that determines the equilibrium distance between the inclusion and the topological defect. We demonstrate how temperature influences

the structure of the dipole formed by the inclusion and the defect. The distance between the inclusion boundary and the defect changes and above the critical temperature the defect resides on the inclusion boundary. This phenomenon, namely, the stepwise jump of the defect position from the smectic film to the surface of the inclusion, to the best of our knowledge, has never been observed before this work. The interparticle distance in chains formed by the droplets decreases substantially with increasing temperature. The change in the position of the topological defect is attributable to the competition between elastic and boundary energies. Our simple model, which uses the notion of the surface extrapolation length, helps to understand the experimental ob-

servations but for a full quantitative description of the data numerical calculations are necessary. There is also another challenge in this field, namely, the reason of tangential and radial director orientation at the droplet boundary in smectic films. To solve these problems, further theoretical and experimental efforts are needed.

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