



The finite-size effect in thin liquid crystal systems

I. Śliwa

Institute of Molecular Physics, Polish Academy of Sciences, Smoluchowskiego 17, 60-179 Poznań, Poland

HIGHLIGHTS

- Local phase transitions in thin liquid crystals are theoretically studied.
- Both surface and intermolecular long-range interactions are taken into account.
- New type of scaling properties at local phase transitions is found.

ARTICLE INFO

Article history:

Received 17 June 2017

Received in revised form 16 October 2017

Available online 1 February 2018

Keywords:

Liquid crystals

Phase transitions

Finite-size scaling

Critical exponents

ABSTRACT

Effects of surface ordering in liquid crystal systems confined between cell plates are of great theoretical and experimental interest. Liquid crystals introduced in thin cells are known to be strongly stabilized and ordered by cell plates. We introduce a new theoretical method for analyzing the effect of surfaces on local molecular ordering in thin liquid crystal systems with planar geometry of the smectic layers. Our results show that, due to the interplay between pair long-range intermolecular forces and nonlocal, relatively short-range, surface interactions, both orientational and translational orders of liquid crystal molecules across confining cells are very complex. In particular, it is demonstrated that the SmA, nematic, and isotropic phases can coexist. The phase transitions from SmA to nematic, as well as from nematic to isotropic phases, occur not simultaneously in the whole volume of the system but begin to appear locally in some regions of the LC sample. Phase transition temperatures are demonstrated to be strongly affected by the thickness of the LC system. The dependence of the corresponding shifts of phase transition temperatures on the layer number is shown to exhibit a power law character. This new type of scaling behavior is concerned with the coexistence of local phases in finite systems. The influence of a specific character of interactions of molecules with surfaces and other molecules on values of the resulting critical exponents is also analyzed.

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1. Introduction

Confining surfaces are known to affect locally critical phenomena of finite systems. As has already been shown, depending on whether the surface couplings are sufficiently stronger or weaker than the bulk interactions, surfaces confining various systems can induce order or disorder [1–7]. Essentially, in both these cases, long-range correlations in directions parallel to surfaces can occur, even if respective bulk phase transitions are discontinuous. Furthermore, surfaces can lead to the coexistence of different phases, separated by sharp fronts that displace under temperature changes. In general, temperature ranges for which particular phases coexist shrinks with the size of systems and ultimately vanish as the bulk limit is

E-mail address: izasliwa@ifmpan.poznan.pl.

approached [8]. Clearly, any resulting bulk phase transition occurs at a definite (critical) temperature. Consequently, finite-size scaling effects associated with the deviation of the temperature, at which a given local phase forms in a finite system, from the corresponding temperature of the bulk (first-order or second-order) phase transition is expected.

A spectacular example of surface confined systems exhibiting the coexistence of different phases are a free-standing smectic films (FSSFs) [9–11]. In these thin liquid crystal (LC) systems, the thickness is quantized in units of layers. As a result, a new class of phase transitions, consisting in the spontaneous layer-by-layer thinning transitions, has been observed [9,10]. This interesting phenomenon has been reported both for compounds exhibiting a first-order phase transition from the smectic A to the isotropic phase [9], as well as for compounds which possess a second-order smectic A–nematic (SmA–N) phase transition [10]. In both types of systems, the central layers of films begin to melt under increasing temperature above the bulk transition temperature. Thus, melting originates in the interior of the film and penetrates towards surfaces. Accordingly, at each step of the layer-thinning process we can observe the coexistence of different LC phases, separated by phase limits (fronts) which move under temperature changes. The movement of these phase fronts consist in successive spontaneous layer-by-layer thinning transitions (local transformations of smectic layers into nematic or isotropic phases) of films. Recently, the layer-thinning transition has been modeled as the local melting and subsequent removal of interior layers of the film to the meniscus as the temperature is increased above the bulk transition temperature [12–16].

In an earlier paper [8], we have presented a theoretical method for analyzing the effect of surfaces on molecular ordering in thin LC systems with planar geometry of the smectic layers. This method, being an extension of the McMillan's mean-field approach [17] to finite systems, allows us to study the local ordering in LC systems of rather large thicknesses, corresponding to thicknesses of real sample cells. Our results, being in agreement with experimental data obtained by applying the technique of fluorescence scanning laser confocal microscopy, show that orientational and translational order of LCs across confining cells are very complex. This anticipates the coexistence of different LC phases, which can be a consequence of the interplay between pair long-range intermolecular forces and nonlocal, relatively short-range, surface interactions. The phase transitions from SmA to nematic, as well as from nematic to isotropic phases, under increasing temperature, do not occur simultaneously in the entire volume of the system but only in some regions of a given LC sample. As a consequence, smectic layers formed in the vicinity of surfaces can coexist not only with nematic, but also with centrally formed smectic phases, or can coexist with isotropic and centrally formed nematic phases [8]. When temperature grows, the phase limits between different LC phases move, mainly towards the center of the system.

The present paper concerns the study on how temperatures of phase transitions, at which particular phases start to occur or vanish, are affected by the thickness of the LC system, governed by two types of interactions: nonlocal, short-range interactions of the smectic layers with surfaces and interlayer couplings (van der Waals interactions). It turns out that the size effect is very distinct and the dependence of the phase transition temperatures on the system thickness (the maximal number n of smectic layers that can planarly form in a given cell) has a power law character. The corresponding critical exponents are argued to reflect the type of interactions existing in the system.

2. Model

We consider a LC system initially exhibiting the SmA phase, consisting of n smectic layers (each of the thickness d) oriented parallel to the confining surfaces. The geometry of the system is shown in Fig. 1. The model involves two types of interactions: interactions of the smectic layers with planar surfaces and with other layers [8]. Surface interactions are assumed as being nonlocal, although rapidly decaying with distance, while different layers are assumed to be coupled via van der Waals forces. Thus, our model describes interactions between large objects (smectic layers) and surfaces, as well as interactions between the layers themselves [8,17]. Therefore, the only space variable, z , denotes the distance from surfaces. Both interactions between different layers, as well as interactions between smectic layers and surfaces are assumed to be attractive. Of course, interactions between molecules within smectic layers are much stronger than the above-mentioned two types of interactions. The intralayer interactions are implicitly accounted by assuming that the center of mass of molecules undergoes an appropriate probability distribution within each of the smectic layers [8,17].

The mean-field expression for the energy of the i th smectic layer (due to couplings of this layer with surfaces and with remaining $n - 1$ layers) has the form [8]:

$$U_i(\theta_i, z_i) = (\bar{V}_i + a_i) \left(\frac{3}{2} \cos^2 \theta_i - \frac{1}{2} \right) + \alpha (\bar{V}_i + \alpha b_i) \left(\frac{3}{2} \cos^2 \theta_i - \frac{1}{2} \right) \cos(2\pi z_i/d) \quad (1)$$

with

$$a_i = \eta_i \sum_{\substack{j=1 \\ (j \neq i)}}^n V_{ij}, \quad b_i = \sigma_i \sum_{\substack{j=1 \\ (j \neq i)}}^n V_{ij}, \quad (2)$$

where

$$\eta_i = \left\langle \frac{3}{2} \cos^2 \theta_i - \frac{1}{2} \right\rangle \quad (3)$$

is the local orientational order parameter, while

$$\sigma_i = \alpha^{-1} \left\langle \left(\frac{3}{2} \cos^2 \theta_i - \frac{1}{2} \right) \cos(2\pi z_i/d) \right\rangle \quad (4)$$

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